Marine Monitoring: Confidence Building and Data Quality Assurance Project

Inter-laboratory comparisons 2014-2016
Determination of radionuclides in seawater, sediment and fish

IAEA
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IAEA Environment Laboratories

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SUMMARY REPORT

With a view to assisting the Government of Japan in its objective of making the Sea Area Monitoring Plan comprehensive, credible and transparent, the IAEA, through its Environment Laboratories, is helping to ensure the high quality of data and to prove the comparability of the results. A 3-year project "Marine Monitoring: Confidence Building and Data Quality Assurance" (2014 – 2016) was initiated as a follow-up activity to recommendations made on marine radioactivity monitoring in a report issued by the IAEA in 2013 (https://www.iaea.org/sites/default/files/final_report120214.pdf) related to the decommissioning of the Fukushima Daiichi Nuclear Power Station, which reviewed Japan's efforts to plan and implement the decommissioning of the plant. Six interlaboratory comparisons (ILCs) and three proficiency tests (PTs) were organized during this project. The PT results published so far can be accessed on the IAEA web pages[1]. This report focuses on ILCs, which are a standard means for laboratories to assess the quality of their measurement results as compared to that of other participating laboratories and identify any potentially needed improvements. Within this project, joint sampling campaigns to collect seawater (Fig A), sediment and fish samples were undertaken. The resulting samples were subsequently analysed in each participating laboratory and the results were compared.

FIG. A. Collection of seawater samples for interlaboratory comparison immediately offshore Fukushima Daiichi Nuclear Power Plant (November 2014).

In total, ten laboratories participated in at least one of the ILCs: seven from Japan (participating on behalf of the Japanese authorities); the IAEA Environment Laboratories in Monaco; and, for the purpose of additional transparency, one laboratory from Ireland and one from New Zealand, both members of the network of Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA).

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[1] 2014 Proficiency Test report accessible at http://www-pub.iaea.org/MTCD/Publications/PDF/IAEA_AQ_43_web.pdf

Surface seawater samples were collected during each of the six sampling missions at five sampling stations in the proximity of the Fukushima Daiichi nuclear power plant. The sampling of marine sediment was done during missions in May 2015 and May 2016 at five sampling stations in the proximity of the Fukushima Daiichi nuclear power plant. Five batches of fresh fish samples (cod, flounder and mackerel) caught at locations in the vicinity of Fukushima Daiichi Nuclear Power Plant were collected during missions in November 2015, while six batches of fresh fish samples (olive flounder, chum salmon, Japanese Spanish mackerel and John Dory) were caught in November 2016.

Radionuclides in seawater, sediment and fish were determined by participating laboratories using radioanalytical methods, including liquid scintillation counting (for ^{3}H), gas-flow proportional counting (for ^{90}Sr), alpha-ray spectrometry (for ^{238}Pu and $^{239+240}$Pu) and gamma-ray spectrometry (for ^{134}Cs and ^{137}Cs).

The laboratories' results were collected by the IAEA, compared and evaluated with statistical tests. For one of the exercises an independent ALMERA laboratory in Hungary assisted in the collection and assessment of the data. From the statistical analysis it can be concluded that the overwhelming majority of the results are not significantly different from each other (see examples in Figs. B – D for seawater, sediment and fish samples, respectively). An analysis of the whole data set demonstrated that 362 out of the 369 statistical tests applied to the data for different radionuclides and samples, i.e. over 98%, were passed with a high level of confidence (99%). Given that departures found are minor, i.e. for 1.9% of the statistical tests applied to the data, it can be said with confidence that the respective 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.

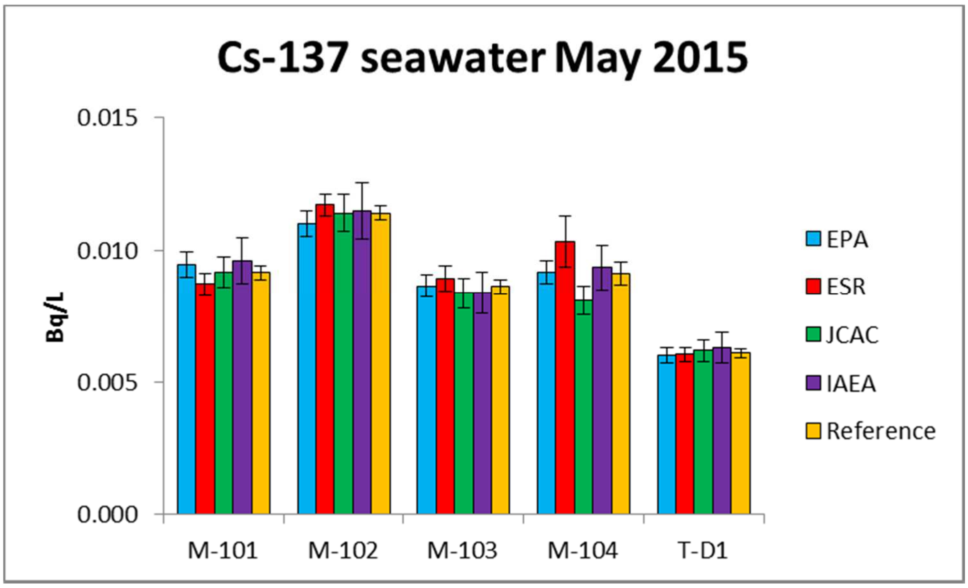

FIG. B. Example of results of an interlaboratory comparison exercise organised in May 2015: Good agreement was obtained between activity concentrations of ^{137}*Cs in five seawater samples reported by laboratories in Japan (Japan Chemical Analysis Center - JCAC), Ireland (Environmental Protection Agency – EPA), New Zealand (Institute of Environmental Science and Research – ESR) and the IAEA (for details please refer to extended report).*

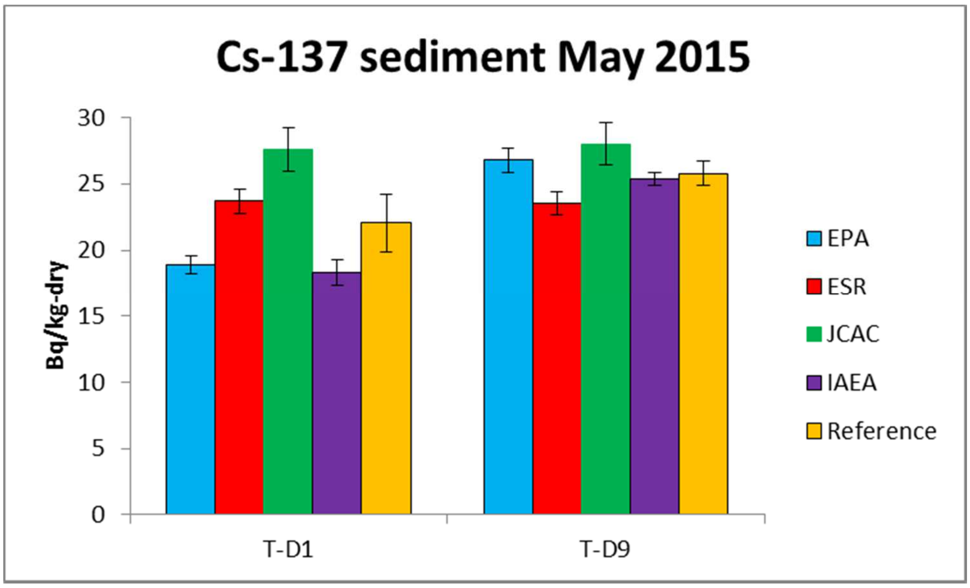

FIG. C. Example of results of an interlaboratory comparison exercise organised in May 2015: Good agreement was obtained between massic activities of ^{137}Cs in two sediment samples reported by laboratories in Japan (Japan Chemical Analysis Center - JCAC), Ireland (Environmental Protection Agency – EPA), New Zealand (Institute of Environmental Science and Research – ESR) and the IAEA (for details please refer to extended report).

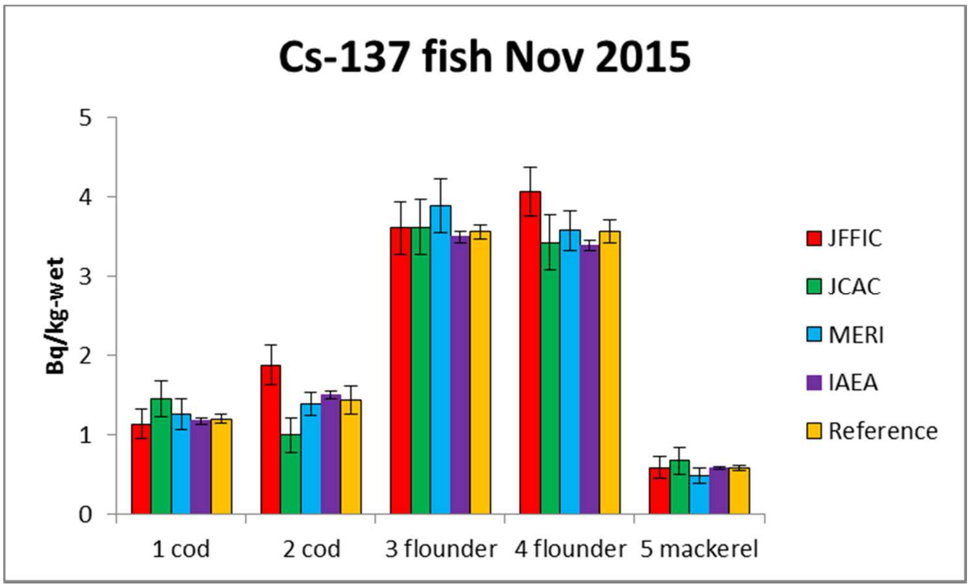

FIG. D. Example of results of an interlaboratory comparison exercise organised in November 2015: Good agreement was obtained between massic activities of ^{137}Cs in five fish samples reported by laboratories in Japan (Japan Frozen Foods Inspection Corporation – JFFIC; Japan Chemical Analysis Center – JCAC; Marine Ecology Research Institute – MERI) and the IAEA (for details please refer to extended report).

Following the six missions organized in 2014 – 2016, the IAEA can report that Japan's sample collection procedures follow the appropriate methodological standards required to obtain representative samples. The results obtained in ILCs demonstrate a 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.

1. BACKGROUND

With a view to assisting the Government of Japan in its objective of making the Sea Area Monitoring Plan comprehensive, credible and transparent, the IAEA, through its Environment Laboratories, is helping to ensure the high quality of data and to prove the comparability of the results. Six sampling missions were organized in 2014 – 2016 to collect seawater, sediment and fish samples for interlaboratory comparisons (ILCs); see Table 1. The missions were carried out within the project "Marine Monitoring: Confidence Building and Data Quality Assurance".

TABLE 1. OVERVIEW OF INTERLABORATORY COMPARISONS

Intercomparison	Seawater	Sediment	Fish	Participating laboratories
September 2014	^{3}H ^{90}Sr ^{134}Cs ^{137}Cs	–	–	GSL (^{3}H) IAEA KANSO (^{90}Sr, ^{134}Cs and ^{137}Cs)
November 2014	^{3}H ^{90}Sr ^{134}Cs ^{137}Cs	–	–	GSL (^{3}H) IAEA KANSO (^{90}Sr, ^{134}Cs and ^{137}Cs)
May 2015	^{3}H ^{90}Sr ^{134}Cs ^{137}Cs	^{134}Cs ^{137}Cs $^{239/240}$Pu	–	EPA ESR IAEA JCAC
November 2015	^{3}H ^{90}Sr ^{134}Cs ^{137}Cs	–	^{134}Cs ^{137}Cs	GSL (seawater: ^{3}H) IAEA (seawater and fish) JCAC (seawater and fish) KANSO (seawater: ^{90}Sr, ^{134}Cs and ^{137}Cs) JFFIC (fish) MERI (fish)
May 2016	^{3}H ^{90}Sr ^{134}Cs ^{137}Cs	^{134}Cs ^{137}Cs ^{238}Pu $^{239/240}$Pu	–	GSL (seawater: ^{3}H) IAEA (seawater and sediment) JCAC (seawater and sediment) KANSO (seawater: ^{90}Sr, ^{134}Cs and ^{137}Cs) FP (sediment) TPT (sediment: ^{134}Cs and ^{137}Cs)

November 2016	^{3}H ^{90}Sr ^{134}Cs ^{137}Cs	–	^{134}Cs ^{137}Cs	GSL (seawater: ^{3}H) IAEA (seawater and fish) JCAC (seawater and fish) KANSO (seawater: ^{90}Sr, ^{134}Cs and ^{137}Cs) JFFIC (fish) MERI (fish)

Participating laboratories:

EPA	Environmental Protection Agency, Ireland
ESR	Institute of Environmental Science and Research, New Zealand
FP	Fukushima Prefecture, Japan
GSL	Geo Science Laboratory, Japan
IAEA	IAEA Environment Laboratories, Monaco
JCAC	Japan Chemical Analysis Center, Japan
JFFIC	Japan Frozen Foods Inspection Corporation, Japan
KANSO	KANSO Co., LTD., Japan
MERI	Marine Ecology Research Institute, Japan
TPT	Tokyo Power Technology, Japan

2. SEAWATER, SEDIMENT AND FISH SAMPLING AND PREPARATION

2.1. SEAWATER

Surface seawater samples were collected during all six sampling missions at five sampling stations (M-101, M-102, M-103, M-104 and T-D1) offshore Fukushima Daiichi nuclear power plant. The coordinates of the sampling stations are given in Table 2 and shown in Figure 1.

TABLE 2. COORDINATES OF THE 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 and sediment)	37°30′00″	141°04′20″
T-D9 (sediment)	37°20′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″

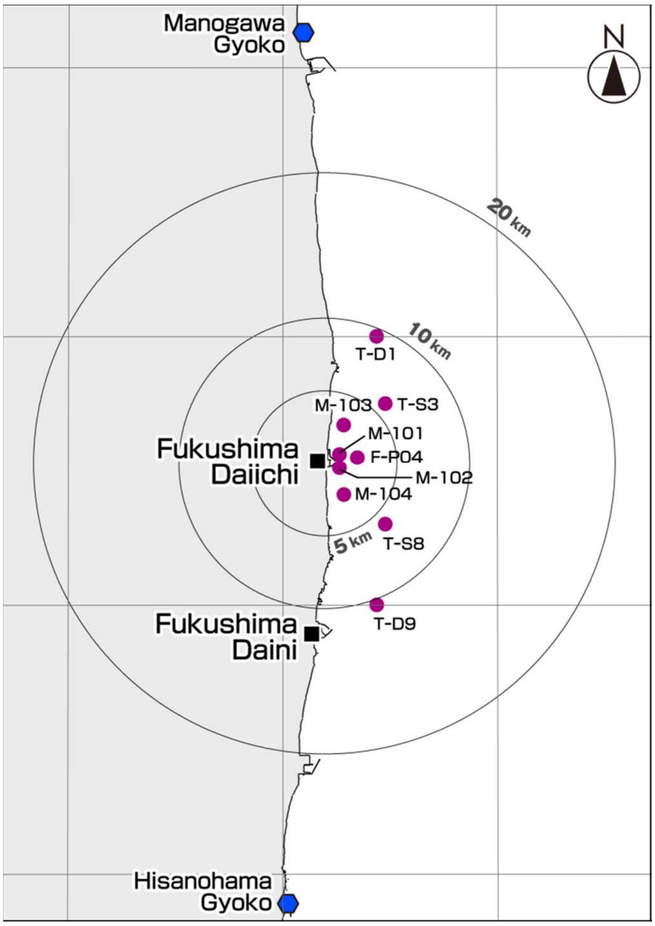

FIG. 1. Locations of sampling stations offshore Fukushima Daiichi nuclear power plant.

During the September and November 2014 missions, two surface seawater samples were collected from each sampling location for subsequent analysis for ^{90}Sr, ^{134}Cs and ^{137}Cs at a Japanese laboratory (KANSO) and the IAEA Environment Laboratories in Monaco. Two separate surface seawater samples were collected at the same locations for subsequent analysis for ^{3}H at a Japanese laboratory (GSL) and the IAEA laboratories. Identical and homogeneous samples were collected using the procedures outlined below. The surface seawater was collected in Van-Dorn water samplers, mixed in a 50 L container and subsequently divided into two 20 L samples. This procedure was repeated three times at each sampling station, resulting in two 60 L of seawater samples being collected. Each sample was acidified to pH 1-2 with concentrated HCl. Upon arrival at the laboratory, the 60 L of seawater sample collected from each sampling station was mixed again before analysis. In the case of samples for analysis for ^{3}H, two 2 L surface seawater samples were collected for the determination of tritium at each sampling station. These samples were not acidified.

FIG 2. Collection of seawater samples immediately offshore Fukushima Daiichi Nuclear Power Plant.

During the May 2015, November 2015, May 2016 and November 2016 missions, seawater samples were again collected from each sampling location for subsequent analysis for ^{90}Sr, ^{134}Cs and ^{137}Cs and, separately, for ^{3}H. As more laboratories participated in each of these exercises, a greater number of seawater samples were collected from each sampling location. In both missions, the procedure regarding the collection of 2 L seawater samples for analysis of ^{3}H was identical to the 2014 exercises described above. In the case of samples for analysis for ^{90}Sr, ^{134}Cs and ^{137}Cs, the collection and distribution method was changed however. A 400 L plastic container with four valves was first filled with seawater. 20 L containers were filled, three at a time, from each of the three/four valves (depending on the number of laboratories participating), resulting in a total of nine/twelve 20 L samples from each sampling station. Each sample was acidified to pH 1–2 with concentrated HCl, as for the 2014 missions. Three 20 L samples were provided to each laboratory. The seawater sampling procedure and the distribution matrix, meant to ensure the homogenisation of the samples distributed to the four laboratories participating in the May 2015 exercise, is shown in Table 3, while the distribution matrix distributed to the three laboratories participating in the November 2015, May 2016 and November 2016 exercises is shown in Table 4.

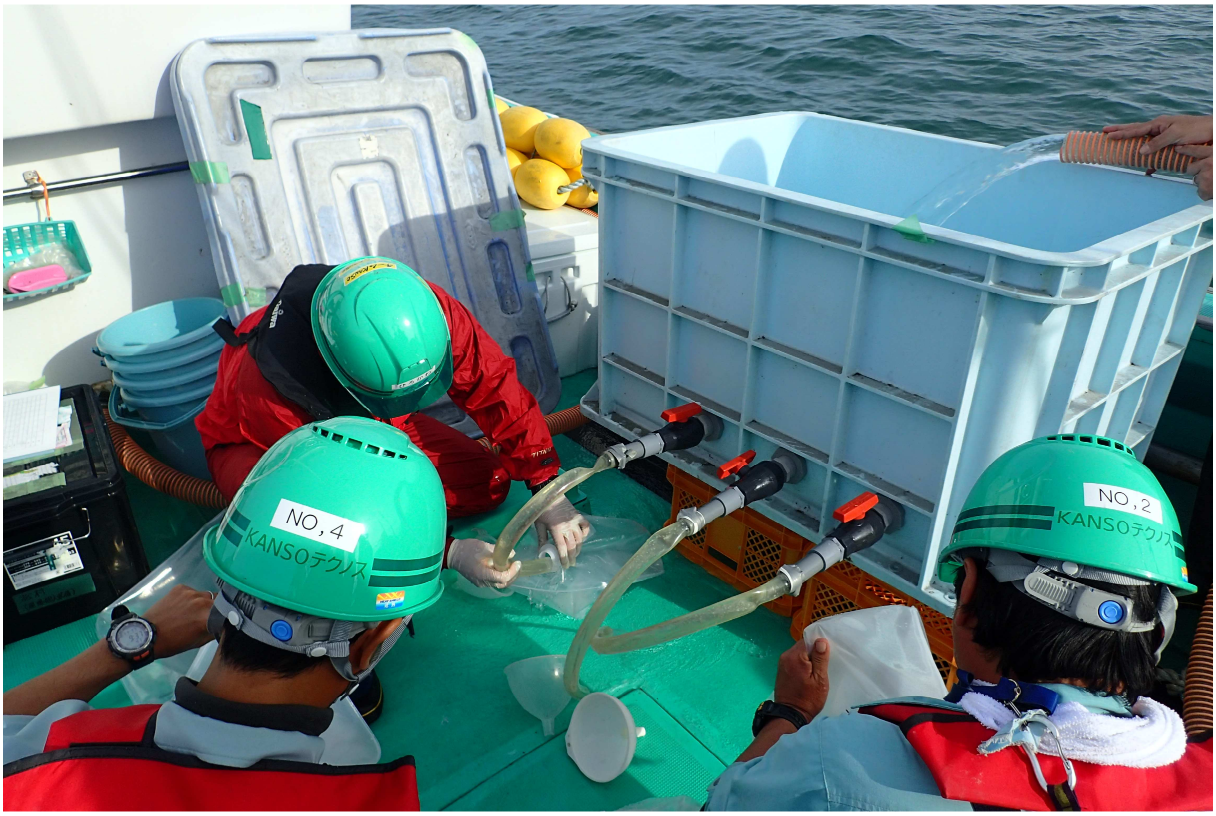

FIG 3. Collection of seawater samples.

TABLE 3. SAMPLE DISTRIBUTION BETWEEN FOUR LABORATORIES (MAY 2015)

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

TABLE 4. SAMPLE DISTRIBUTION BETWEEN THREE LABORATORIES (NOV 2015, MAY 2016 AND NOV 2016)

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

FIG 4. Distribution and preparation of transport arrangements for collected seawater samples.

2.2. SEDIMENT

The sampling of sediment was done during May 2015 and May 2016 offshore Fukushima Daiichi nuclear power plant. Sediment samples at stations T-D1 (in 2015), T-D9 (in 2015), F-P04 (in 2016), T-S3 (in 2016) and T-S8 (in 2016) off Fukushima were collected using a grab sampler. The coordinates of the sampling stations are given in Table 2. The sediment samples were oven-dried at 105 °C on large stainless steel trays, crushed using stainless steel spatulae and sieved through a 2-mm mesh sieve at the KANSO laboratory. No grinding was required prior to sieving due to the sandy nature of the sediments. The sieved sample was grinded using mortars and pestles, 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 grinded sample. The samples were then bottled in 500 mL plastic bottles and their homogeneity was re-checked using gamma-ray spectrometry with high purity germanium (HPGe) detectors. Approximately 350 g of dried sediment from each station was delivered to each participant.

FIG 5. Collection of sediment samples.

2.3. FISH

In 2015, five batches of fresh fish samples (two batches of cod, two batches of flounder and one batch of mackerel) caught at locations in the vicinity of Fukushima Daiichi Nuclear Power Plant were collected from the fish landing port of Onahama on 18 November 2015 (see Table 5). The fresh fish samples were prepared the next day at the Marine Ecology Research Institute (MERI) in Onjuku (Japan) and subsequently analysed for ^{134}Cs and ^{137}Cs at MERI. After subsequent measurement on 20 November 2015 at the Japan Chemical Analysis Center (JCAC) in Chiba (Japan) and the Japan Frozen Foods Inspection Corporation (JFFIC) in Yokohama (Japan), the fresh fish samples were frozen and shipped to the IAEA Environmental Laboratories in Monaco in early January 2016. After defrosting, the fresh fish samples were measured at the IAEA Environmental Laboratories between 26 and 30 January 2016. All measurements were done by gamma-ray spectrometry with HPGe detectors. The measurement time per sample was 1 hour for MERI, JCAC and JFFIC and between 22 and 47 hours for the IAEA. All massic activities were reported at a reference time of 18 November 2015 0:00 UTC.

FIG 6. Collection of the fish samples.

In 2016, six batches of fresh fish samples (two batches of olive flounder, two batches of chum salmon, one batch of Japanese Spanish mackerel and one batch of John Dory) caught at locations in the vicinity of Fukushima Daiichi Nuclear Power Plant were collected from the fish landing port of Onahama on 16 November 2016 (see Table 5). As before, the fresh fish samples were prepared the next day at MERI and subsequently analysed for ^{134}Cs and ^{137}Cs. After subsequent measurement on 18 November 2016 at JCAC and JFFIC, the fresh fish samples were frozen and shipped to the IAEA Environmental Laboratories in Monaco in January 2017. After defrosting, the fresh fish samples were measured at the IAEA Environmental Laboratories between 26 January and 2 March 2017. All measurements were done by gamma-ray spectrometry with HPGe detectors. The measurement time per sample was 1 hour for MERI, JCAC and JFFIC and between 20 and 88 hours for the IAEA. All massic activities were reported at a reference time of 16 November 2016 0:00 UTC.

TABLE 5. COORDINATES OF THE CATCH POSITION

Batch	Latitude (N)	Longitude (E)
15FA0001 cod	36°59′19″	141°28′44″
15FA0002 cod	36°59′19″	141°28′44″
15FA0003 flounder	37°06′16″	141°07′49″
15FA0004 flounder	36°54′42″	141°02′16″
15FA0005 mackerel	37°04′12″	141°16′22″
16FA0001 olive flounder	36°54′56″	140°53′36″
16FA0002 olive flounder	36°53′12″	140°52′07″
16FA0003 chum salmon (male)	37°04′14″	140°58′50″
16FA0004 chum salmon (female)	37°05′16″	140°59′04″
16FA0005 Japanese Spanish mackerel	36°54′22″	140°56′12″
16FA0006 John Dory	36°52′32″	140°47′34″

FIG 7. Preparation of the fish samples before measurement.

3. METHODOLOGY OF RADIONUCLIDE DETERMINATION

3.1. SEAWATER

Radionuclides of interest in seawater were determined by six laboratories participating in one or more inter-laboratory comparison: GSL, KANSO and JCAC, all participating on behalf of the Nuclear Regulation Authority, Japan, and EPA, ESR and IAEA (see Table 1).

3.1.1. GSL methodology for seawater

3.1.1.1. Tritium analysis

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

3.1.2. KANSO methodology for seawater

3.1.2.1. ^{90}Sr analysis

KANSO used an ion exchange resin for pre-concentration of strontium from seawater sample followed by precipitation of carbonates and barium chromate. After reaching of secular equilibrium, ^{90}Y was separated using a ferric hydroxide co-precipitation technique and measured by a gas-flow counter (Aloka LBC-472/LBC-4202B).

3.1.2.2. ^{134}Cs and ^{137}Cs analysis

Chemical separation of caesium by using ammonium phosphomolybdate (AMP) followed by gamma-ray spectrometry with a HPGe detector was used for ^{134}Cs and ^{137}Cs determination in seawater.

3.1.3. JCAC methodology for seawater

3.1.3.1. Tritium analysis

JCAC used liquid scintillation counter (Aloka LB-5/LB-7) for measurement of tritium in seawater after distillation, electrolytic enrichment and second distillation.

3.1.3.2. ^{90}Sr analysis

For the determination of ^{90}Sr, a cation exchange resin column was used for pre-concentration of strontium from seawater sample followed by precipitation of carbonates and additional cation exchange resin column for separation of calcium. ^{90}Y was removed by scavenging and measurement was done after reaching of secular equilibrium by low background beta counter (Aloka LBC-4211).

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

Chemical separation of caesium by using AMP followed by gamma-ray spectrometry with a HPGe detector was used for ^{134}Cs and ^{137}Cs determination in seawater.

3.1.4. EPA methodology for seawater

3.1.4.1. ^{134}Cs and ^{137}Cs analysis

At EPA, the previously acidified seawater samples were filtered through 0.45 µm filters and caesium was extracted using Triskem AMP-PAN resin based on a method published by Pike et al. [1]. The

AMP-PAN resin, containing ^{134}Cs and ^{137}Cs, was counted by gamma-ray spectrometry using a high-purity germanium (HPGe) well type detector (Canberra GCW2023).

3.1.5. ESR methodology for seawater

3.1.5.1. ^{90}Sr analysis

ESR used a method based on Eichrom Sr resin for the determination of ^{90}Sr in seawater.

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

ESR used the Eichrom potassium nickel ferrocyanate (KNiFC-PAN) resin method for the determination of ^{134}Cs and ^{137}Cs from 20 L of seawater. The resin containing caesium radioisotopes was counted by gamma-ray spectrometry.

3.1.6. IAEA methodology for seawater

3.1.6.1. Tritium analysis

Tritium was determined by liquid scintillation counting after double vacuum distillation (at 35°C) and electrolytic enrichment followed by a second distillation (under atmospheric pressure). An ultra-low level liquid scintillation counter (Quantulus 1220) was used for the counting of an aliquot of the enriched and distilled sample mixed with Quicksafe 400 scintillation cocktail.

3.1.6.2. ^{90}Sr analysis

At the IAEA, 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). Only for the samples from sampling stations M-101 and M-102 in the September 2014 mission, sequential separation by precipitation of MnO_2 (Pu and Am), AMP (Cs) and oxalate (Sr) was used. The ^{90}Sr activity concentration is calculated based on the measurement of ^{90}Y (yttrium oxalate source) beta activity using a proportional counter (Risø National Laboratory model GM-25-5, Roskilde, Denmark) with an efficiency of up to 44%.

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

^{134}Cs and ^{137}Cs were measured by low-level HPGe gamma-ray spectrometry (EURISYS SYSTEME coaxial HPGe detector model EGC 50-200-R with 47.5 % relative efficiency). The counting time was set to obtain a counting uncertainty of less than 5%.

3.2. SEDIMENT

Radionuclides of interest in sediment samples were determined by four laboratories participating in the May 2015 inter-laboratory comparison: JCAC, participating on behalf of the Nuclear Regulation Authority, Japan, and EPA, ESR and IAEA (see Table 1). Four laboratories participated in the May 2016 inter-laboratory comparison: JCAC, FP, and TPT, participating on behalf of the Nuclear Regulation Authority, Japan, and IAEA (see Table 1).

3.2.1. JCAC methodology for sediment

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

JCAC used direct counting by a p-type coaxial HPGe detector with a relative efficiency 31% for the determination of ^{134}Cs and ^{137}Cs in sediment.

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

Plutonium isotopes in sediments were determined by alpha-ray 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. EPA methodology for sediment

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

At EPA, ^{134}Cs and ^{137}Cs in re-homogenised and re-dried sediment samples were counted on a germanium detector (Canberra GC7520/S). Canberra ApexGamma (Genie 2k) software was used for the analysis of the obtained spectra, while coincidence summing and attenuation corrections were applied using the GESPECOR Monte-Carlo software.

3.2.2.2. $^{239/240}$Pu analysis

The method for the determination of plutonium radioisotopes used at EPA is based on work by Luisier et al. [2]. This technique utilises Triskem DGA resin in association with TEVA resin for the sequential determination of plutonium and americium radioisotopes in environmental samples. Alpha spectrometry was used for the measurement of $^{239+240}$Pu in sediments.

3.2.3. ESR methodology for sediment

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

ESR counted ^{134}Cs and ^{137}Cs in sediments in a cylinder geometry by gamma-ray spectrometry.

3.2.3.2. $^{239/240}$Pu analysis

An Eichrom method based on the use of TEVA and TRU resins followed by alpha spectrometry was used for the determination of plutonium isotopes.

3.2.4. FP methodology for sediment

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

FP determined ^{134}Cs and ^{137}Cs in sediments by gamma-ray spectrometry with a HPGe detector.

3.2.4.2. $^{239/240}$Pu analysis

Plutonium isotopes in sediments were determined by alpha-ray 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.

3.2.5. TPT methodology for sediment

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

TPT determined ^{134}Cs and ^{137}Cs in sediment by gamma-ray spectrometry with a p-type coaxial HPGe detector.

3.2.6. IAEA methodology for sediment

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

At the IAEA, 40 g of sediment was sealed in a tin can and ^{134}Cs and ^{137}Cs were measured by a germanium detector (EURISYS SYSTEME coaxial HPGe detector model EGPC 170-210-R) with cosmic veto shielding and relative efficiency of 170 %.

3.2.6.2. ^{238}Pu and $^{239/240}$Pu analysis

The method for the determination of plutonium radioisotopes in sediments was based on 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 1X4) 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.

3.3. FISH

Radionuclides of interest in fish samples were determined by four laboratories participating in the November 2015 and November 2016 inter-laboratory comparisons: MERI, JCAC and JFFIC, all participating on behalf of the Japan Fisheries Agency, and IAEA (see Table 1).

FIG 8. Measurement of fish samples by gamma-ray spectrometry.

3.3.1. MERI methodology for fish

MERI used direct counting by p-type coaxial HPGe detectors with relative efficiencies between 28% and 46% for the determination of ^{134}Cs and ^{137}Cs in fish. The sample mass was 2 kg-wet and each sample was measured for 1 hour.

3.3.2. JCAC methodology for fish

JCAC used direct counting by p-type coaxial HPGe detectors with relative efficiencies between 29% and 33% for the determination of ^{134}Cs and ^{137}Cs in fish. The sample mass was 2 kg-wet and each sample was measured for 1 hour.

3.3.3. JFFIC methodology for fish

JFFIC used direct counting by p-type coaxial HPGe detectors with relative efficiencies between 22% and 32% for the determination of ^{134}Cs and ^{137}Cs in fish. The sample mass was 2 kg-wet and each sample was measured for 1 hour.

3.3.4. IAEA methodology for fish

IAEA used direct counting by a coaxial HPGe detector with relative efficiency of 48% for the determination of ^{134}Cs and ^{137}Cs in fish. The sample mass was 1 kg-wet and the samples were measured between 22 and 47 hours (2015) and 20 and 88 hours (2016). As the IAEA was the final recipient of the fish samples in both exercises, it was possible to measure the fish samples for a longer time period resulting in smaller counting uncertainties.

4. RESULTS

4.1. GENERAL

4.1.1. Uncertainties

Uncertainties quoted in this report are combined standard uncertainties 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 14, 15A and 16).

4.1.2. Reference time

All activity concentrations and massic activities were reported at reference times specific for each of the six sampling missions.

4.2. SEAWATER

Tables 6–8, 9A and 9B contain the results reported by the participating laboratories for the activity concentrations of ^{3}H, ^{90}Sr, ^{134}Cs and ^{137}Cs in the seawater samples. Figures 2 to 25 show the activity concentrations of these radionuclides in the seawater samples.

TABLE 6. ^3H ACTIVITY CONCENTRATIONS (Bq L^{-1}) IN SEAWATER

		IAEA	GSL	JCAC
Sep 2014	M-101	0.17 ± 0.04	0.171 ± 0.016	–
	M-102	0.14 ± 0.03	0.129 ± 0.015	–
	M-103	0.16 ± 0.04	0.101 ± 0.014	–
	M-104	0.15 ± 0.03	0.151 ± 0.016	–
	T-D1	0.10 ± 0.03	0.123 ± 0.015	–
Nov 2014	M-101	0.12 ± 0.04	0.175 ± 0.016	–
	M-102	0.16 ± 0.04	0.114 ± 0.013	–
	M-103	0.12 ± 0.04	0.118 ± 0.014	–
	M-104	0.14 ± 0.04	0.117 ± 0.013	–
	T-D1	0.14 ± 0.04	0.083 ± 0.013	–
May 2015	M-101	0.14 ± 0.05	–	0.08 ± 0.02
	M-102	0.08 ± 0.04	–	0.09 ± 0.02
	M-103	0.07 ± 0.03	–	0.09 ± 0.02
	M-104	0.08 ± 0.04	–	0.08 ± 0.02
	T-D1	0.06 ± 0.03	–	0.08 ± 0.02
Nov 2015	M-101	0.10 ± 0.04	0.093 ± 0.014	0.099 ± 0.021
	M-102	0.09 ± 0.04	0.109 ± 0.014	0.101 ± 0.021
	M-103	0.14 ± 0.03	0.092 ± 0.014	0.083 ± 0.020
	M-104	0.12 ± 0.03	0.110 ± 0.014	0.090 ± 0.021
	T-D1	0.08 ± 0.03	0.093 ± 0.014	0.076 ± 0.020
May 2016	M-101	< 0.17	0.075 ± 0.010	0.069 ± 0.019
	M-102	< 0.14	0.090 ± 0.011	0.106 ± 0.019
	M-103	< 0.13	0.117 ± 0.014	0.148 ± 0.020
	M-104	< 0.13	0.079 ± 0.010	0.072 ± 0.018
	T-D1	< 0.12	0.107 ± 0.013	0.110 ± 0.020
Nov 2016	M-101	0.17 ± 0.04	0.104 ± 0.014	0.112 ± 0.020
	M-102	0.21 ± 0.04	0.147 ± 0.015	0.135 ± 0.021
	M-103	0.12 ± 0.03	0.130 ± 0.015	0.082 ± 0.020
	M-104	0.08 ± 0.03	0.131 ± 0.015	0.094 ± 0.020
	T-D1	0.13 ± 0.03	0.107 ± 0.015	0.104 ± 0.020

TABLE 7. ^{90}Sr ACTIVITY CONCENTRATIONS (Bq L^{-1}) IN SEAWATER

		IAEA	KANSO	JCAC
Sep 2014	M-101	0.026 ± 0.003	0.0190 ± 0.0011	–
	M-102	0.0110 ± 0.0014	0.0100 ± 0.00007	–
	M-103	0.00125 ± 0.00009	0.00110 ± 0.00020	–
	M-104	0.0080 ± 0.0006	0.0069 ± 0.0006	–
	T-D1	0.00101 ± 0.00008	0.00086 ± 0.00017	–
Nov 2014	M-101	0.0100 ± 0.0005	0.0087 ± 0.0007	–
	M-102	0.00377 ± 0.00020	0.0036 ± 0.0004	–
	M-103	0.00276 ± 0.00015	0.0025 ± 0.0003	–
	M-104	0.00386 ± 0.00020	0.0039 ± 0.0004	–
	T-D1	0.00098 ± 0.00006	0.00110 ± 0.00021	–
May 2015*	M-101	0.00153 ± 0.00008	–	0.00160 ± 0.00016
	M-102	0.00103 ± 0.00006	–	0.00105 ± 0.00013
	M-103	0.00129 ± 0.00007	–	0.00162 ± 0.00017
	M-104	0.00084 ± 0.00005	–	0.00109 ± 0.00014
	T-D1	0.00128 ± 0.00007	–	0.00168 ± 0.00017
Nov 2015	M-101	0.00174 ± 0.00010	0.00150 ± 0.00020	0.00177 ± 0.00017
	M-102	0.0180 ± 0.0009	0.0170 ± 0.0010	0.0174 ± 0.0009
	M-103	0.00161 ± 0.00009	0.00160 ± 0.00020	0.00134 ± 0.00015
	M-104	0.00119 ± 0.00007	0.00120 ± 0.00020	0.00112 ± 0.00014
	T-D1	0.00135 ± 0.00008	0.00110 ± 0.00020	0.00110 ± 0.00014
May 2016	M-101	0.00103 ± 0.00007	0.00150 ± 0.00024	0.00103 ± 0.00013
	M-102	0.00166 ± 0.00009	0.0026 ± 0.0003	0.00203 ± 0.00018
	M-103	0.00178 ± 0.00010	0.0020 ± 0.0003	0.00166 ± 0.00016
	M-104	0.00101 ± 0.00007	0.00120 ± 0.00023	0.00095 ± 0.00013
	T-D1	0.00077 ± 0.00006	0.00110 ± 0.00018	0.00077 ± 0.00011
Nov 2016	M-101	0.00174 ± 0.00010	0.00260 ± 0.00024	0.00199 ± 0.00018
	M-102	0.00219 ± 0.00012	0.00210 ± 0.00021	0.00215 ± 0.00019
	M-103	0.00155 ± 0.00009	0.00120 ± 0.00017	0.00116 ± 0.00013
	M-104	0.00127 ± 0.00007	0.00150 ± 0.00018	0.00092 ± 0.00012
	T-D1	0.00112 ± 0.00007	0.00094 ± 0.00015	0.00101 ± 0.00013

* additional values reported by ESR (in Bq L^{-1}) were < 0.015 (M-101), < 0.014 (M-102), < 0.033 (M-103), < 0.046 (M-104) and < 0.022 (T-D1)

TABLE 8. ^{134}Cs ACTIVITY CONCENTRATIONS (Bq L^{-1}) IN SEAWATER

		IAEA	KANSO	ESR	JCAC
Sep 2014	M-101	0.046 ± 0.005	0.040 ± 0.005	–	–
	M-102	0.037 ± 0.004	0.039 ± 0.005	–	–
	M-103	0.0179 ± 0.0018	0.0160 ± 0.0021	–	–
	M-104	0.035 ± 0.004	0.030 ± 0.004	–	–
	T-D1	0.0023 ± 0.0003	0.0023 ± 0.0004	–	–
Nov 2014	M-101	0.0238 ± 0.0021	0.021 ± 0.003	–	–
	M-102	0.0170 ± 0.0016	0.0150 ± 0.0020	–	–
	M-103	0.0111 ± 0.0008	0.0094 ± 0.0013	–	–
	M-104	0.0120 ± 0.0009	0.0096 ± 0.0013	–	–
	T-D1	0.0072 ± 0.0006	0.0059 ± 0.0009	–	–
May 2015	M-101	0.00227 ± 0.00023	–	0.00196 ± 0.00022	0.0025 ± 0.0003
	M-102	0.0030 ± 0.0003	–	0.00288 ± 0.00018	0.0027 ± 0.0003
	M-103	0.00195 ± 0.00019	–	0.00174 ± 0.00013	0.0022 ± 0.0003
	M-104	0.00246 ± 0.00024	–	0.0025 ± 0.0005	0.0018 ± 0.0003
	T-D1	0.00143 ± 0.00016	–	0.0013 ± 0.0004	0.0015 ± 0.0003
Nov 2015	M-101	0.0066 ± 0.0006	0.0069 ± 0.0003	–	0.0074 ± 0.0005
	M-102	0.0154 ± 0.0015	0.0140 ± 0.0006	–	0.0141 ± 0.0009
	M-103	0.0083 ± 0.0008	0.0093 ± 0.0004	–	0.0111 ± 0.0007
	M-104	0.0047 ± 0.0004	0.0051 ± 0.0003	–	0.0052 ± 0.0004
	T-D1	0.0038 ± 0.0004	0.00420 ± 0.00025	–	0.0054 ± 0.0004
May 2016	M-101	0.00197 ± 0.00023	0.00170 ± 0.00017	–	0.0026 ± 0.0003
	M-102	0.0029 ± 0.0003	0.00250 ± 0.00020	–	0.0024 ± 0.0004
	M-103	0.0053 ± 0.0005	0.0053 ± 0.0003	–	0.0050 ± 0.0004
	M-104	0.00195 ± 0.00021	0.00170 ± 0.00017	–	0.0021 ± 0.0003
	T-D1	0.00133 ± 0.00014	0.00140 ± 0.00017	–	0.0018 ± 0.0003
Nov 2016	M-101	0.0063 ± 0.0006	0.0064 ± 0.0003	–	0.0079 ± 0.0006
	M-102	0.0111 ± 0.0011	0.00100 ± 0.0004	–	0.00122 ± 0.0008
	M-103	0.0067 ± 0.0007	0.0060 ± 0.0003	–	0.0062 ± 0.0005
	M-104	0.0042 ± 0.0004	0.00410 ± 0.00024	–	0.0039 ± 0.0004
	T-D1	0.00164 ± 0.00018	0.00150 ± 0.00017	–	0.0019 ± 0.0003

TABLE 9A. ^{137}Cs ACTIVITY CONCENTRATIONS (Bq L^{-1}) IN SEAWATER

		IAEA	KANSO	JCAC
Sep 2014	M-101	0.134 ± 0.013	0.120 ± 0.007	–
	M-102	0.113 ± 0.011	0.120 ± 0.006	–
	M-103	0.052 ± 0.005	0.052 ± 0.003	–
	M-104	0.104 ± 0.010	0.094 ± 0.005	–
	T-D1	0.0085 ± 0.0009	0.0070 ± 0.0005	–
Nov 2014	M-101	0.076 ± 0.007	0.070 ± 0.004	–
	M-102	0.052 ± 0.005	0.0470 ± 0.0025	–
	M-103	0.0357 ± 0.0025	0.0350 ± 0.0019	–
	M-104	0.038 ± 0.003	0.0350 ± 0.0018	–
	T-D1	0.0214 ± 0.0016	0.0210 ± 0.0012	–
May 2015	See Table 9B			
Nov 2015	M-101	0.031 ± 0.003	0.0310 ± 0.0010	0.0326 ± 0.0017
	M-102	0.067 ± 0.006	0.0610 ± 0.0020	0.066 ± 0.003
	M-103	0.041 ± 0.004	0.0410 ± 0.0013	0.0472 ± 0.0024
	M-104	0.0219 ± 0.0020	0.0230 ± 0.0008	0.0251 ± 0.0013
	T-D1	0.0187 ± 0.0017	0.0190 ± 0.0007	0.0240 ± 0.0013
May 2016	M-101	0.0111 ± 0.0011	0.0100 ± 0.0004	0.0144 ± 0.0008
	M-102	0.0144 ± 0.0014	0.0130 ± 0.0005	0.0152 ± 0.0009
	M-103	0.028 ± 0.003	0.0290 ± 0.0010	0.0286 ± 0.0015
	M-104	0.0108 ± 0.0011	0.0100 ± 0.0004	0.0116 ± 0.0007
	T-D1	0.0082 ± 0.0008	0.0079 ± 0.0003	0.0086 ± 0.0005
Nov 2016	M-101	0.037 ± 0.004	0.0400 ± 0.0013	0.0458 ± 0.0024
	M-102	0.065 ± 0.006	0.0640 ± 0.0021	0.068 ± 0.004
	M-103	0.038 ± 0.004	0.0360 ± 0.0012	0.0413 ± 0.0021
	M-104	0.026 ± 0.003	0.0250 ± 0.0008	0.0269 ± 0.0014
	T-D1	0.0114 ± 0.0011	0.0110 ± 0.0004	0.0119 ± 0.0007

TABLE 9B. ^{137}Cs ACTIVITY CONCENTRATIONS (Bq L^{-1}) IN SEAWATER (MAY 2015)

	IAEA	JCAC	EPA	ESR	Reference value*
M-101	0.0096 ± 0.0009	0.0092 ± 0.0006	0.0095 ± 0.0005	0.0087 ± 0.0004	0.0092 ± 0.0003
M-102	0.0115 ± 0.0011	0.0114 ± 0.0007	0.0110 ± 0.0005	0.0117 ± 0.0004	0.0114 ± 0.0003
M-103	0.0084 ± 0.0008	0.0084 ± 0.0005	0.0086 ± 0.0004	0.0089 ± 0.0005	0.0086 ± 0.0003
M-104	0.0093 ± 0.0009	0.0081 ± 0.0005	0.0092 ± 0.0004	0.0103 ± 0.0010	0.0091 ± 0.0004
T-D1	0.0063 ± 0.0006	0.0062 ± 0.0004	0.0060 ± 0.0003	0.0060 ± 0.0003	0.00609 ± 0.00017

* The reference value, a power-moderated mean of the combined results, was calculated by a method currently being used by the Consultative Committee for Ionizing Radiation, Section II: Measurement of radionuclides, CCRI(II) [3].

4.3. SEDIMENT

Table 10A contains the results reported by the four participating laboratories (EPA, ESR, IAEA and JCAC) for the massic activities of radionuclides in the May 2015 sediment samples. Table 10B contains the results reported by the four participating laboratories (FP, IAEA, JCAC and TPT) for the massic activities of radionuclides in the May 2016 sediment samples. Figures 26 to 31 show the massic activities of ^{134}Cs, ^{137}Cs and $^{239+240}$Pu in the sediment samples.

TABLE 10A. RADIONUCLIDE MASSIC ACTIVITIES (Bq kg^{-1}-dry) IN SEDIMENT (MAY 2015)

		IAEA	EPA	JCAC	ESR	Reference value*
^{134}Cs	T-D1	5.0 ± 0.3	5.20 ± 0.25	6.5 ± 0.5	6.6 ± 0.4	5.8 ± 0.4
	T-D9	7.15 ± 0.20	7.6 ± 0.4	7.1 ± 0.6	6.6 ± 0.3	7.12 ± 0.20
^{137}Cs	T-D1	18.3 ± 1.0	18.9 ± 0.7	27.6 ± 1.6	23.7 ± 1.0	22.0 ± 2.2
	T-D9	25.4 ± 0.5	26.8 ± 0.9	28.0 ± 1.6	23.6 ± 0.9	25.8 ± 0.9
$^{239+240}$Pu	T-D1	0.232 ± 0.012	0.32 ± 0.04	0.283 ± 0.016	0.5 ± 0.4	0.27 ± 0.03
	T-D9	0.503 ± 0.022	0.54 ± 0.09	0.490 ± 0.022	0.4 ± 0.4	0.499 ± 0.019

* The reference value, a power-moderated mean of the combined results, was calculated by a method currently being used by the Consultative Committee for Ionizing Radiation, Section II: Measurement of radionuclides, CCRI(II) [3].

TABLE 10B. RADIONUCLIDE MASSIC ACTIVITIES (Bq kg^{-1}-dry) IN SEDIMENT (MAY 2016)

		IAEA	Fukushima Prefecture	JCAC	Tokyo Power Technology
^{134}Cs	F-P04	4.5 ± 0.4	5.7 ± 0.5	5.8 ± 0.5	–
	T-S3	151 ± 4	–	165 ± 8	147 ± 4
	T-S8	5.84 ± 0.17	–	5.8 ± 0.5	6.1 ± 0.3
^{137}Cs	F-P04	25.7 ± 1.7	28.0 ± 1.6	28.9 ± 1.5	–
	T-S3	776 ± 16	–	851 ± 42	762 ± 22
	T-S8	31.1 ± 0.3	–	29.7 ± 1.6	32.0 ± 1.1
^{238}Pu	F-P04	< 0.004	< 0.02	0.0041 ± 0.0014	–
	T-S3	0.009 ± 0.003	–	0.0083 ± 0.0017	–
	T-S8	0.0064 ± 0.0022	–	0.0079 ± 0.0014	–
$^{239+240}$Pu	F-P04	0.381 ± 0.020	0.43 ± 0.04	0.370 ± 0.015	–
	T-S3	0.577 ± 0.025	–	0.591 ± 0.020	–
	T-S8	0.536 ± 0.024	–	0.507 ± 0.016	–

4.4. FISH

Tables 11 and 12 contain the results reported by the four participating laboratories (IAEA, JFFIC, JCAC and MERI) for the massic activities of radionuclides in the fish samples. Figures 32 to 35 show the massic activities of ^{134}Cs and ^{137}Cs in the fish samples.

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

	IAEA	JFFIC	JCAC	MERI	Reference value*
15FA0001 cod	0.29 ± 0.03	0.27 ± 0.08	0.27 ± 0.15	0.40 ± 0.11	0.30 ± 0.03
15FA0002 cod	0.36 ± 0.03	< 0.6	< 0.6	0.31 ± 0.07	–
15FA0003 flounder	0.76 ± 0.04	0.74 ± 0.16	1.00 ± 0.21	0.88 ± 0.15	0.79 ± 0.05
15FA0004 flounder	0.74 ± 0.03	0.88 ± 0.15	1.02 ± 0.22	0.83 ± 0.11	0.79 ± 0.04
15FA0005 mackerel	0.108 ± 0.017	0.19 ± 0.13	< 0.5	0.16 ± 0.06	–
16FA0001 olive flounder	0.35 ± 0.04	0.67 ± 0.13	0.26 ± 0.15	< 0.6	–
16FA0002 olive flounder	0.64 ± 0.04	0.43 ± 0.12	0.31 ± 0.18	0.51 ± 0.09	0.51 ± 0.07
16FA0003 chum salmon (male)	< 0.07	< 0.4	< 0.6	< 0.5	–
16FA0004 chum salmon (female)	< 0.03	0.18 ± 0.14	0.20 ± 0.11	0.15 ± 0.08	–
16FA0005 Japanese Spanish mackerel	< 0.10	< 0.6	0.19 ± 0.12	< 0.5	–
16FA0006 John Dory	0.21 ± 0.03	0.32 ± 0.10	< 0.5	0.13 ± 0.09	–

* The reference value, a power-moderated mean of the combined results, was calculated by a method currently being used by the Consultative Committee for Ionizing Radiation, Section II: Measurement of radionuclides, CCRI(II) [3].

TABLE 12. Cs-137 MASSIC ACTIVITIES (Bq kg^{-1}-wet) IN FISH

	IAEA	JFFIC	JCAC	MERI	Reference value*
15FA0001 cod	1.18 ± 0.04	1.14 ± 0.19	1.45 ± 0.22	1.26 ± 0.20	1.20 ± 0.05
15FA0002 cod	1.51 ± 0.04	1.88 ± 0.25	1.00 ± 0.21	1.39 ± 0.15	1.44 ± 0.17
15FA0003 flounder	3.50 ± 0.07	3.6 ± 0.3	3.6 ± 0.3	3.9 ± 0.3	3.56 ± 0.08
15FA0004 flounder	3.39 ± 0.07	4.1 ± 0.3	3.4 ± 0.3	3.58 ± 0.25	3.57 ± 0.15
15FA0005 mackerel	0.585 ± 0.022	0.59 ± 0.13	0.68 ± 0.17	0.49 ± 0.10	0.58 ± 0.03
16FA0001 olive flounder	1.99 ± 0.07	2.67 ± 0.25	2.1 ± 0.3	2.06 ± 0.23	2.18 ± 0.15
16FA0002 olive flounder	3.34 ± 0.09	4.2 ± 0.4	3.9 ± 0.4	3.38 ± 0.23	3.64 ± 0.21
16FA0003 chum salmon (male)	0.11 ± 0.03	< 0.5	< 0.5	< 0.6	–
16FA0004 chum salmon (female)	0.104 ± 0.019	< 0.6	< 0.5	< 0.4	–
16FA0005 Japanese Spanish mackerel	0.48 ± 0.04	0.50 ± 0.19	0.40 ± 0.14	0.19 ± 0.16	0.43 ± 0.06
16FA0006 John Dory	1.25 ± 0.05	1.47 ± 0.21	1.13 ± 0.20	1.30 ± 0.14	1.27 ± 0.06

* The reference value, a power-moderated mean of the combined results, was calculated by a method currently being used by the Consultative Committee for Ionizing Radiation, Section II: Measurement of radionuclides, CCRI(II) [3].

5. STATISTICAL EVALUATION OF THE RESULTS

For the May 2015 mission (seawater and sediment), the collection of the data was carried out by the Radioanalytical Reference Laboratory of the National Food Chain Safety Office, Hungary, while the data evaluation was done by the IAEA. For all other missions, the data was collected and evaluated by the IAEA.

The statistical evaluation method used for the results depended on the size of the data set. If two or three results were received, one or three zeta tests [4] were performed.

The zeta test was defined as:

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

where:

x_i is the value of laboratory i [Bq L^{-1}]
x_j is the value of laboratory j [Bq L^{-1}]
u_i is the standard uncertainty (at $k = 1$) for the value of laboratory i [Bq L^{-1}]
u_j is the standard uncertainty (at $k = 1$) for the value of laboratory j [Bq L^{-1}]

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 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:

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

where: x_{lab} is the individual laboratory result
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 [4]. 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.

5.1. SEAWATER

Table 13A and 13B contain the zeta scores for the activity concentrations of radionuclides in the seawater samples.

TABLE 13A. RADIONUCLIDES IN SEAWATER; ZETA SCORES

		Sep 2014	Nov 2014	May 2015
^{3}H	M-101	0.02	1.29	1.17
	M-102	0.28	1.17	0.32
	M-103	1.34	0.03	0.39
	M-104	0.10	0.48	0.10
	T-D1	0.61	1.29	0.55
^{90}Sr	M-101	1.97	1.59	0.40
	M-102	0.66	0.38	0.17
	M-103	0.70	0.70	1.81
	M-104	1.39	0.09	1.68
	T-D1	0.82	0.55	2.16
^{134}Cs	M-101	0.86	0.82	0.98 / 0.57 / 1.39
	M-102	0.36	0.78	0.35 / 0.61 / 0.40
	M-103	0.67	1.10	0.89 / 0.68 / 1.38
	M-104	0.87	1.51	0.05 / 1.71 / 1.17
	T-D1	0.08	1.25	0.25 / 0.33 / 0.44
^{137}Cs	M-101	0.91	0.75	See Table 14
	M-102	0.51	1.03	
	M-103	0.04	0.23	
	M-104	0.84	0.91	
	T-D1	1.55	0.21	

NOTE: Format “x.xx” refers to $\zeta_{1,2}$; format “x.xx / y.yy / z.zz” refers to $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$. Number 1 refers to IAEA, number 2 refers to GSL/KANSO and number 3 refers to JCAC.

TABLE 13B. RADIONUCLIDES IN SEAWATER; ZETA SCORES

		Nov 2015	May 2016	Nov 2016
^{3}H	M-101	0.17 / 0.03 / 0.22	– / – / 0.29	1.73 / 1.42 / 0.33
	M-102	0.45 / 0.24 / 0.32	– / – / 0.73	1.63 / 1.82 / 0.46
	M-103	1.45 / 1.59 / 0.38	– / – / 1.27	0.31 / 0.90 / 1.91
	M-104	0.30 / 0.82 / 0.79	– / – / 0.33	1.44 / 0.39 / 1.50
	T-D1	0.39 / 0.12 / 0.71	– / – / 0.13	0.75 / 0.78 / 0.12
^{90}Sr	M-101	1.07 / 0.17 / 1.03	1.91 / 0.01 / 1.74	**3.35** / 1.18 / 2.06
	M-102	0.70 / 0.44 / 0.30	**2.76** / 1.85 / 1.52	0.36 / 0.16 / 0.18
	M-103	0.06 / 1.57 / 1.04	0.75 / 0.64 / 1.06	1.87 / 2.47 / 0.20
	M-104	0.04 / 0.46 / 0.33	0.78 / 0.47 / 0.97	1.15 / 2.52 / **2.65**
	T-D1	1.15 / 1.55 / 0.00	1.71 / 0.05 / 1.56	1.11 / 0.75 / 0.37
^{134}Cs	M-101	0.44 / 1.00 / 0.80	0.94 / 1.54 / 2.40	0.20 / 1.92 / 2.33
	M-102	0.88 / 0.75 / 0.10	1.11 / 1.08 / 0.25	0.94 / 0.80 / 2.50
	M-103	1.09 / **2.61** / 2.26	0.00 / 0.53 / 0.68	0.98 / 0.61 / 0.38
	M-104	0.83 / 0.77 / 0.10	0.93 / 0.50 / 1.21	0.25 / 0.65 / 0.55
	T-D1	0.77 / **2.60** / 2.33	0.32 / 1.40 / 1.13	0.56 / 0.77 / 1.20
^{137}Cs	M-101	0.00 / 0.46 / 0.81	0.95 / 2.42 / **4.94**	0.65 / 1.89 / 2.12
	M-102	0.95 / 0.20 / 1.19	0.95 / 0.49 / 2.27	0.08 / 0.46 / 0.96
	M-103	0.00 / 1.33 / 2.27	0.32 / 0.18 / 0.22	0.61 / 0.66 / 2.20
	M-104	0.51 / 1.34 / 1.39	0.69 / 0.62 / 2.09	0.35 / 0.32 / 1.16
	T-D1	0.16 / 2.48 / **3.43**	0.35 / 0.36 / 1.06	0.31 / 0.39 / 1.13

NOTE: Format “x.xx” refers to $\zeta_{1,2}$; format “x.xx / y.yy / z.zz” refers to $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$. Number 1 refers to IAEA, number 2 refers to GSL/KANSO and number 3 refers to JCAC.

Table 14 contains the degrees of relative equivalence for the activity concentrations of ^{137}Cs in the seawater samples in the May 2015 mission.

TABLE 14. RADIONUCLIDES IN SEAWATER; RELATIVE DEGREE OF EQUIVALENCE (%)

		IAEA	EPA	JCAC	ESR
^{137}Cs May 2015	M-101	5 ± 23	3 ± 11	0 ± 14	–5 ± 10
	M-102	1 ± 22	–4 ± 10	0 ± 14	3 ± 8
	M-103	–2 ± 21	0 ± 10	–3 ± 14	4 ± 13
	M-104	3 ± 23	1 ± 14	–11 ± 16	13 ± 25
	T-D1	3 ± 22	–2 ± 11	2 ± 16	–1 ± 9

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

5.2. SEDIMENT

Table 15A contains the degrees of relative equivalence for the massic activities of the radionuclides in the sediment samples. Table 15B contains the zeta scores for the massic activities of the radionuclides in the sediment samples.

TABLE 15A. RADIONUCLIDES IN SEDIMENT (MAY 2015); RELATIVE DEGREE OF EQUIVALENCE (%)

		IAEA	EPA	JCAC	ESR
^{134}Cs	T-D1	–13 ± 21	–10 ± 20	12 ± 26	14 ± 22
	T-D9	0 ± 8	7 ± 12	0 ± 20	–7 ± 11
^{137}Cs	T-D1	–17 ± 27	–14 ± 26	25 ± 29	7 ± 27
	T-D9	–2 ± 10	4 ± 11	9 ± 16	–9 ± 11
$^{239+240}$Pu	T-D1	–15 ± 30	15 ± 39	3 ± 31	82 ± 353
	T-D9	1 ± 11	7 ± 43	–2 ± 11	–14 ± 215

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

TABLE 15B. RADIONUCLIDES IN SEDIMENT (MAY 2016); ZETA SCORES

		May 2016
^{134}Cs	F-P04	1.87 / 1.98 / 0.11
	T-S3	0.68 / 1.50 / 1.94
	T-S8	0.60 / 0.06 / 0.44
^{137}Cs	F-P04	1.00 / 1.42 / 0.41
	T-S3	0.53 / 1.64 / 1.86
	T-S8	0.82 / 0.89 / 1.24
^{238}Pu	F-P04	–
	T-S3	0.35
	T-S8	0.60
$^{239+240}$Pu	F-P04	1.10 / 0.44 / 1.40
	T-S3	0.45
	T-S8	1.01

NOTE: Format “x.xx” refers to $\zeta_{1,3}$; format “x.xx / y.yy / z.zz” refers to $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$. Number 1 refers to IAEA, number 2 refers to Fukushima Prefecture/Tokyo Power Technology and number 3 refers to JCAC.

5.3. FISH

Table 16 contains the degrees of relative equivalence for the massic activities of the radionuclides in the fish samples.

TABLE 16. RADIONUCLIDES IN FISH; RELATIVE DEGREE OF EQUIVALENCE (%)

		IAEA (1)	JFFIC (2)	JCAC (3)	MERI (4)
^{134}Cs	15FA0001 cod	–3 ± 22	–10 ± 65	–8 ± 122	35 ± 84
	15FA0002 cod	Note 1	DL	DL	Note 1
	15FA0003 flounder	–4 ± 13	–7 ± 49	26 ± 63	12 ± 45
	15FA0004 flounder	–6 ± 13	11 ± 45	30 ± 68	6 ± 33
	15FA0005 mackerel	Note 2	Note 2	DL	Note 2
	16FA0001 olive flounder	Note 3	Note 3	Note 3	DL
	16FA0002 olive flounder	24 ± 35	–16 ± 55	–39 ± 85	–1 ± 46
	16FA0003 chum salmon (male)	DL	DL	DL	DL
	16FA0004 chum salmon (female)	DL	Note 4	Note 4	Note 4
	16FA0005 Japanese Spanish mackerel	DL	DL	–	DL
	16FA0006 John Dory	Note 5	Note 5	DL	Note 5
^{137}Cs	15FA0001 cod	–2 ± 10	–5 ± 38	20 ± 45	5 ± 39
	15FA0002 cod	5 ± 31	31 ± 45	–31 ± 42	–3 ± 36
	15FA0003 flounder	–2 ± 5	1 ± 22	2 ± 23	9 ± 22
	15FA0004 flounder	–5 ± 11	14 ± 20	–4 ± 23	0 ± 17
	15FA0005 mackerel	1 ± 12	2 ± 56	17 ± 73	–15 ± 40
	16FA0001 olive flounder	–9 ± 19	22 ± 29	–3 ± 30	–6 ± 27
	16FA0002 olive flounder	–8 ± 15	16 ± 25	6 ± 25	–7 ± 18
	16FA0003 chum salmon (male)	–	DL	DL	DL
	16FA0004 chum salmon (female)	–	DL	DL	DL
	16FA0005 Japanese Spanish mackerel	13 ± 36	18 ± 108	–6 ± 79	–55 ± 88
	16FA0006 John Dory	–1 ± 10	16 ± 39	–11 ± 38	2 ± 26

NOTE: 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 0.71 for $\zeta_{1,4}$

Note 2; Values of 0.61, 0.82 and 0.23 for $\zeta_{1,2}$, $\zeta_{1,4}$ and $\zeta_{2,4}$, respectively

Note 3; Values of 2.44, 0.55 and 2.06 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively

Note 4; Values of 0.12, 0.17 and 0.38 for $\zeta_{2,3}$, $\zeta_{2,4}$ and $\zeta_{3,4}$, respectively

Note 5; Values of 1.05, 0.96 and 1.49 for $\zeta_{1,2}$, $\zeta_{1,4}$ and $\zeta_{2,4}$, respectively

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

Number 1 refers to IAEA, number 2 refers to JFFIC, number 3 refers to JCAC and number 4 refers to MERI.

6. CONCLUSION

A detailed data analysis was performed on activity concentrations reported for ^{3}H, ^{90}Sr, ^{134}Cs and ^{137}Cs in thirty seawater samples, massic activities reported for ^{134}Cs, ^{137}Cs and $^{239+240}$Pu in five sediment samples and massic activities reported for ^{134}Cs and ^{137}Cs in eleven fish samples, all collected offshore Fukushima Daiichi nuclear power plant between September 2014 and November 2016. The samples were shared between the ten laboratories [EPA (Ireland), ESR (New Zealand), Fukushima Prefecture (Japan), GSL (Japan), IAEA (Monaco), JCAC (Japan), JFFIC (Japan), KANSO (Japan), MERI (Japan) and Tokyo Power Technology (Japan)]. From this analysis it can be concluded that the overwhelming majority of the results are not significantly different from each other. A global analysis of the whole data set demonstrated that 362 out of the 369 statistical tests applied to the data, i.e. over 98%, were passed with a high level of confidence (99%). The only exceptions were four zeta scores that were just marginally higher than the critical limit of 2.58:

$\zeta = 2.60$ for ^{134}Cs in seawater sample T-D1 in Nov 2015 between IAEA and JCAC
$\zeta = 2.61$ for ^{134}Cs in seawater sample M-103 in Nov 2015 between IAEA and JCAC
$\zeta = 2.65$ for ^{90}Sr in seawater sample M-101 in Nov 2016 between IAEA and KANSO
$\zeta = 2.76$ for ^{90}Sr in seawater sample M-102 in May 2016 between IAEA and KANSO

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

$\zeta = 3.35$ for ^{90}Sr in seawater sample M-104 in Nov 2016 between KANSO and JCAC
$\zeta = 3.43$ for ^{137}Cs in seawater sample T-D1 in Nov 2015 between KANSO and JCAC
$\zeta = 4.94$ for ^{137}Cs in seawater sample M-101 in May 2016 between KANSO and JCAC

Despite these minor 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 the six sampling missions organized in 2014–2016, the IAEA can confidently report that Japan's sample collection procedures follow the appropriate methodological standards required to obtain representative samples. The results obtained in ILCs demonstrate a 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] PIKE, S., et al., Extraction of cesium in seawater off Japan using AMP-PAN resin and quantification via gamma spectrometry and inductively coupled mass spectrometry. J. Radioanal. Nucl. Chem. **296** (2013) 369.

[2] LUISIER, F., ALVARADO, J.A.C., STEINMANN, P., KRACHLER, M., FROIDEVAUX, P., A new method for the determination of plutonium and americium using high pressure microwave digestion and alpha-spectrometry or ICP-SMS, J. Radioanal. Nucl. Chem. **281** (2009) 425.

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

[4] INTERNAL ORGANIZATION FOR STANDARDIZATION, Statistical methods for use in proficiency testing by interlaboratory comparisons, ISO 13528:2005, Geneva (2005).

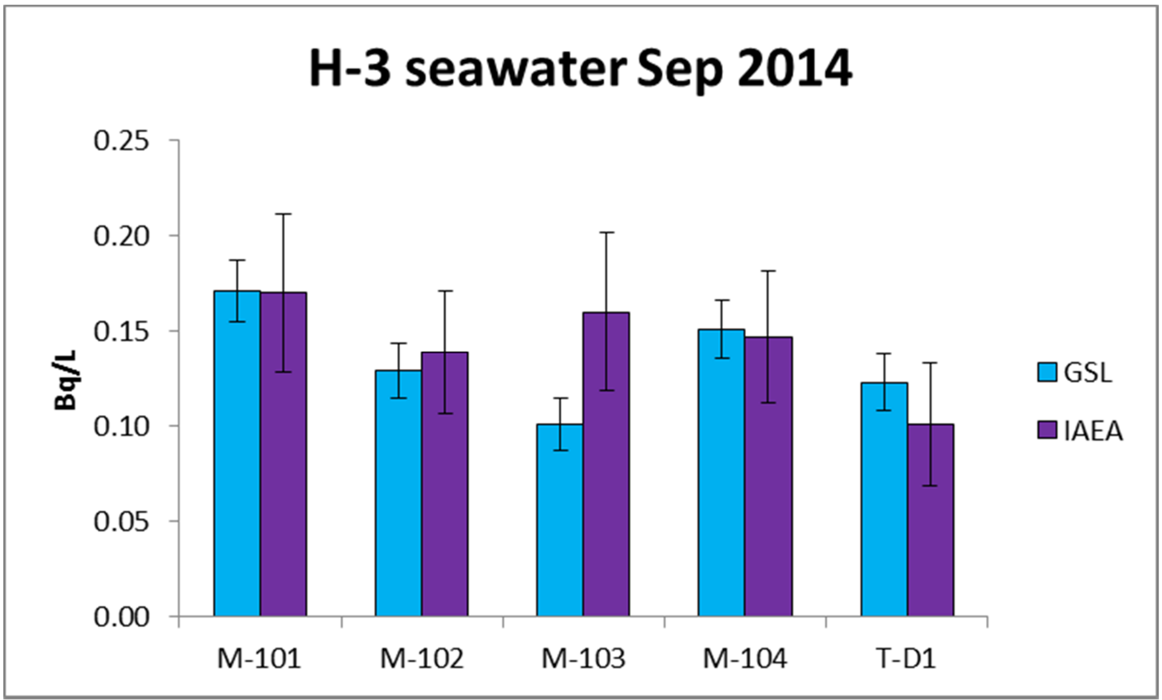

FIG. 2. Activity concentrations of ^{3}H in seawater samples.

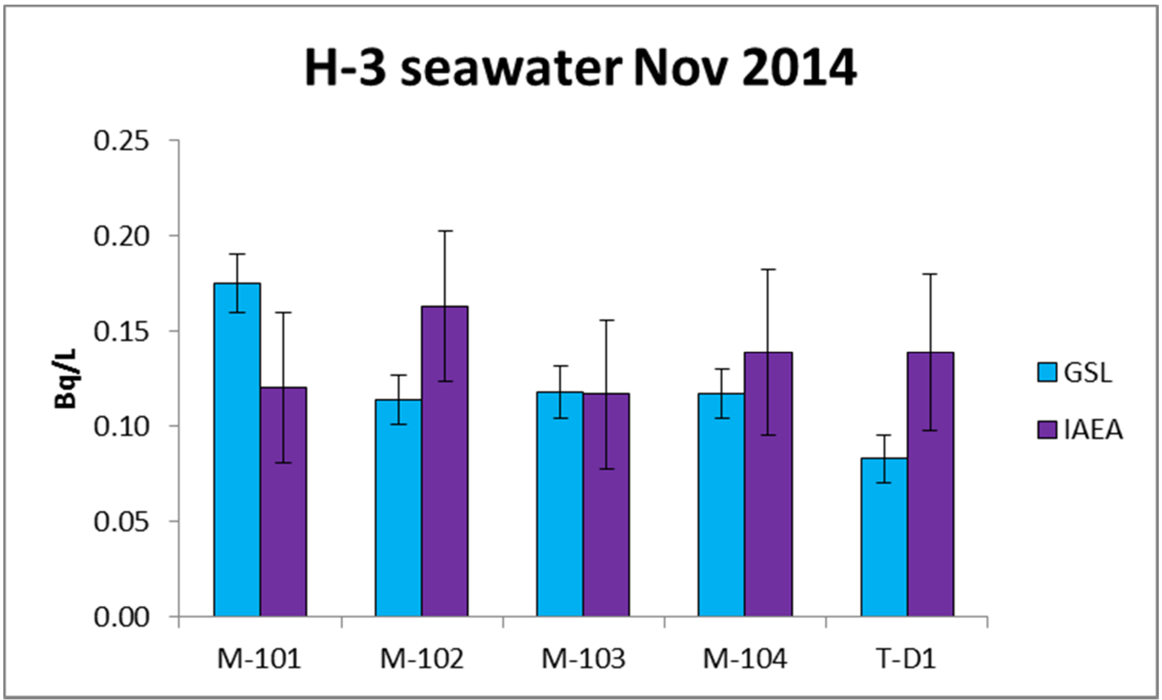

FIG. 3. Activity concentrations of ^{3}H in seawater samples.

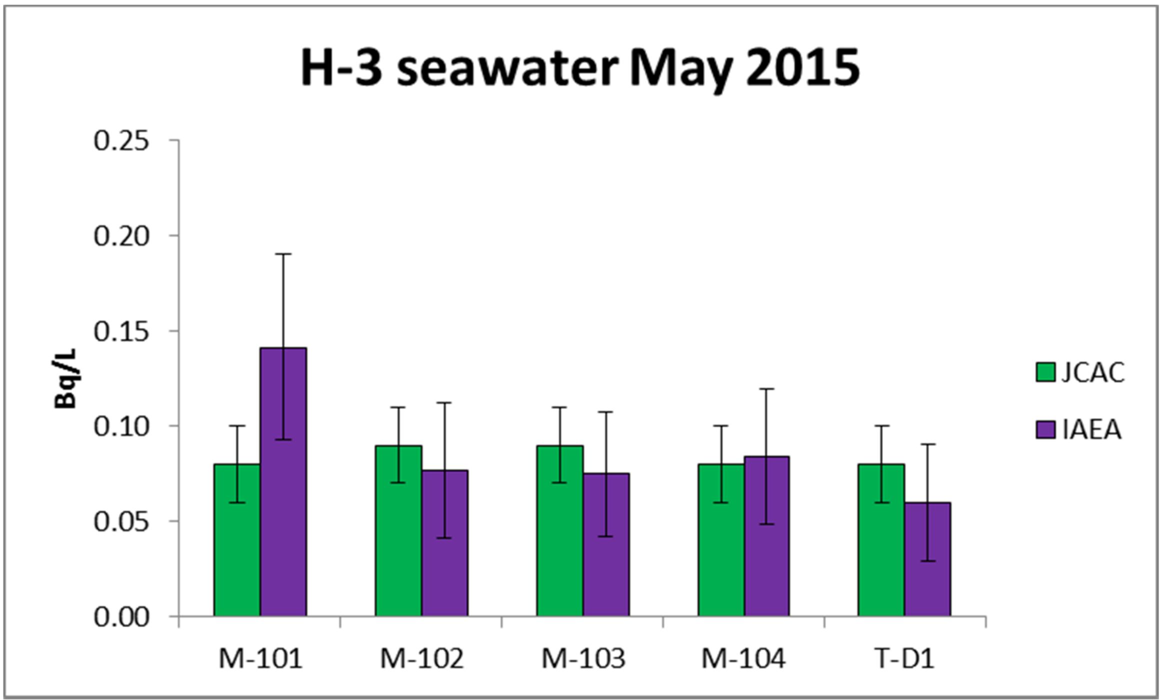

FIG. 4. Activity concentrations of ^{3}H in seawater samples.

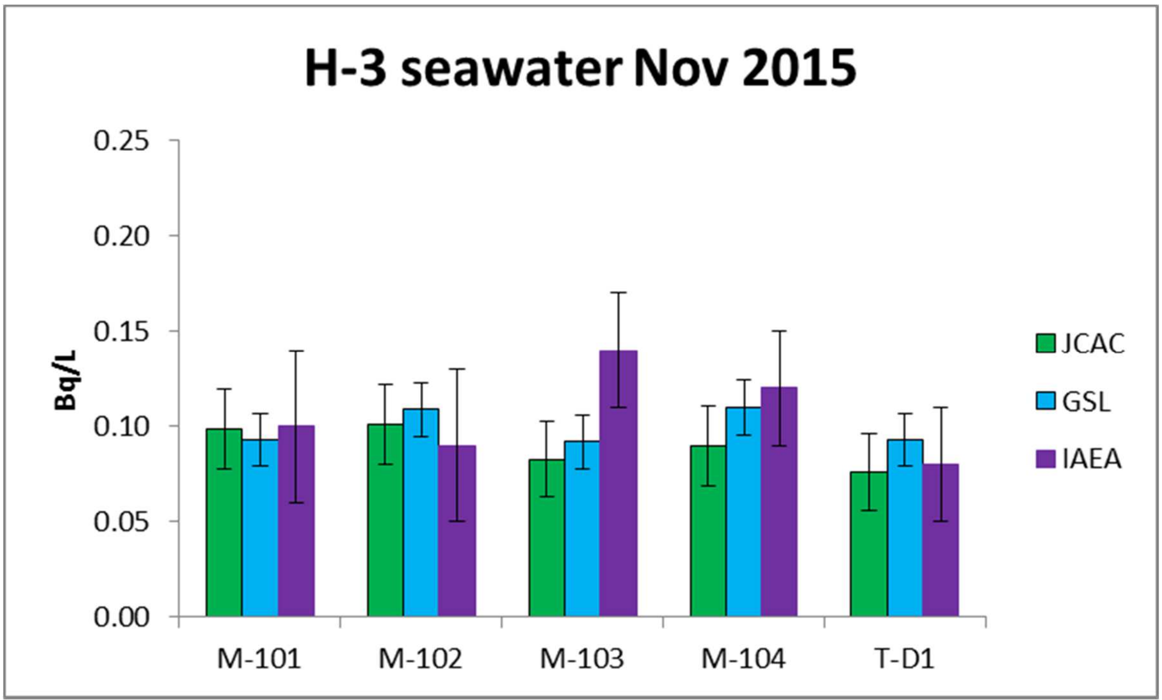

FIG. 5. Activity concentrations of ^{3}H in seawater samples.

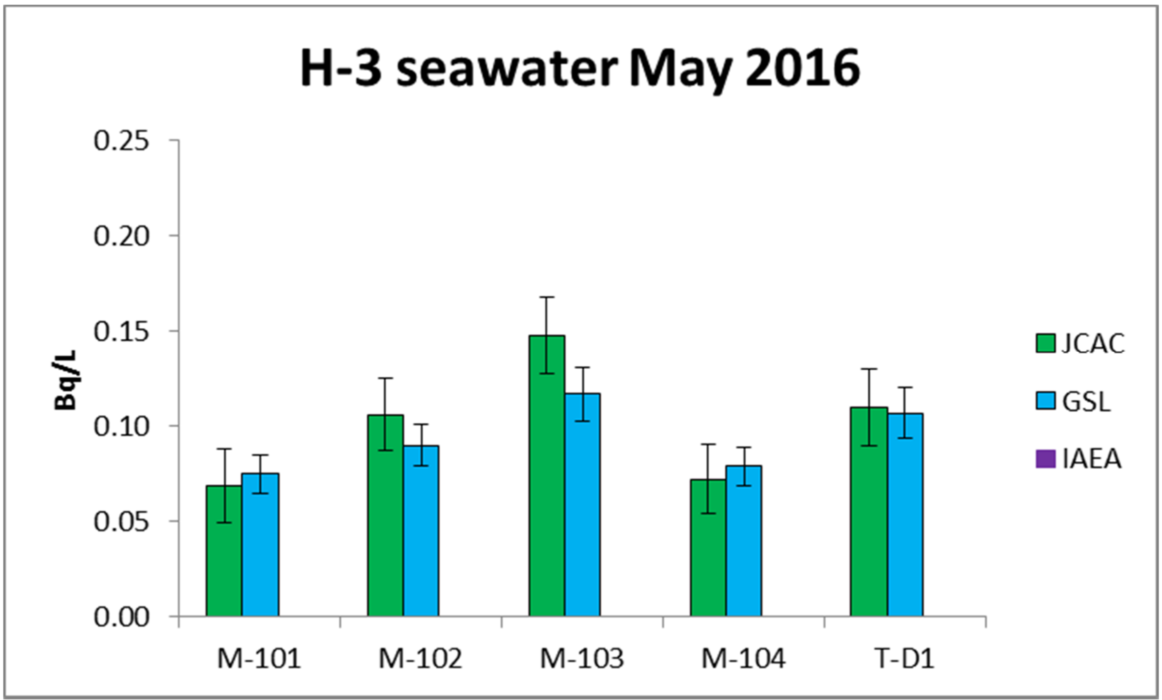

FIG. 6. Activity concentrations of ^{3}H in seawater samples.

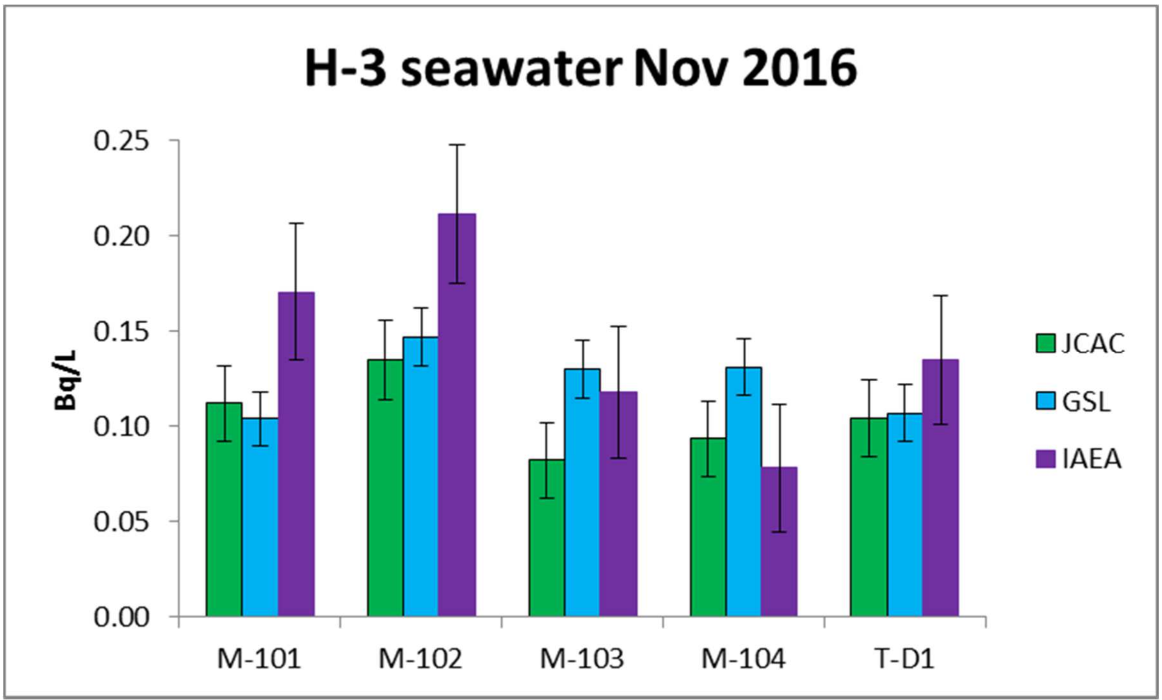

FIG. 7. Activity concentrations of ^{3}H in seawater samples.

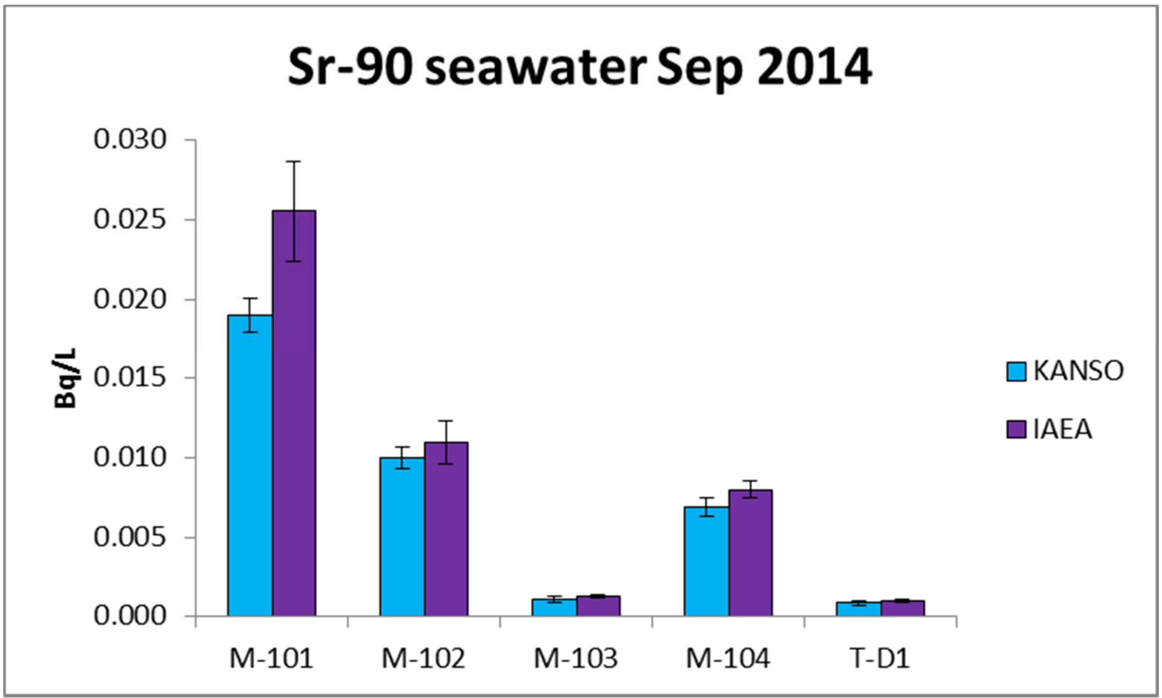

FIG. 8. Activity concentrations of ^{90}Sr in seawater samples.

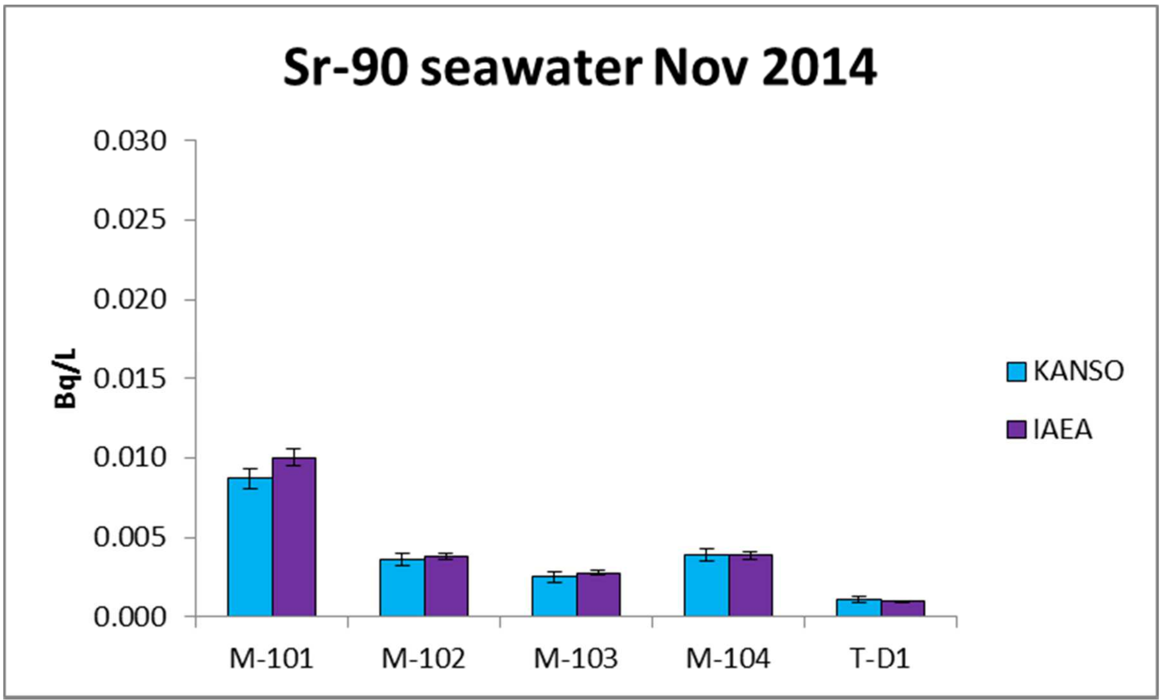

FIG. 9. Activity concentrations of ^{90}Sr in seawater samples.

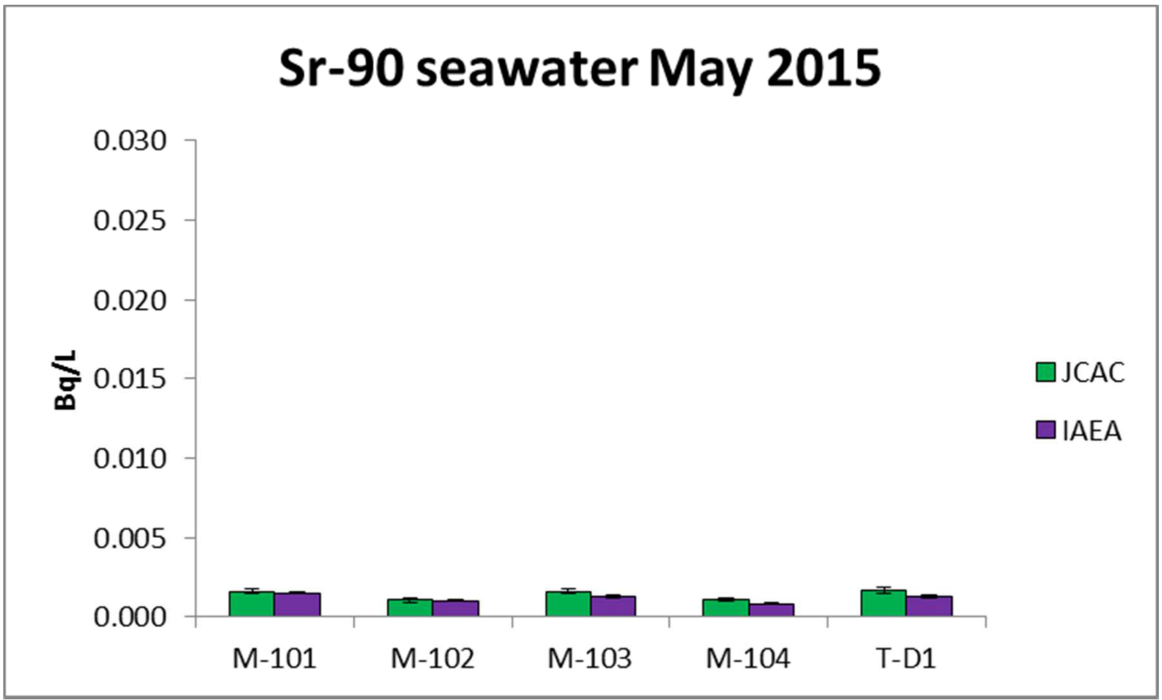

FIG. 10. Activity concentrations of ^{90}*Sr in seawater samples.*

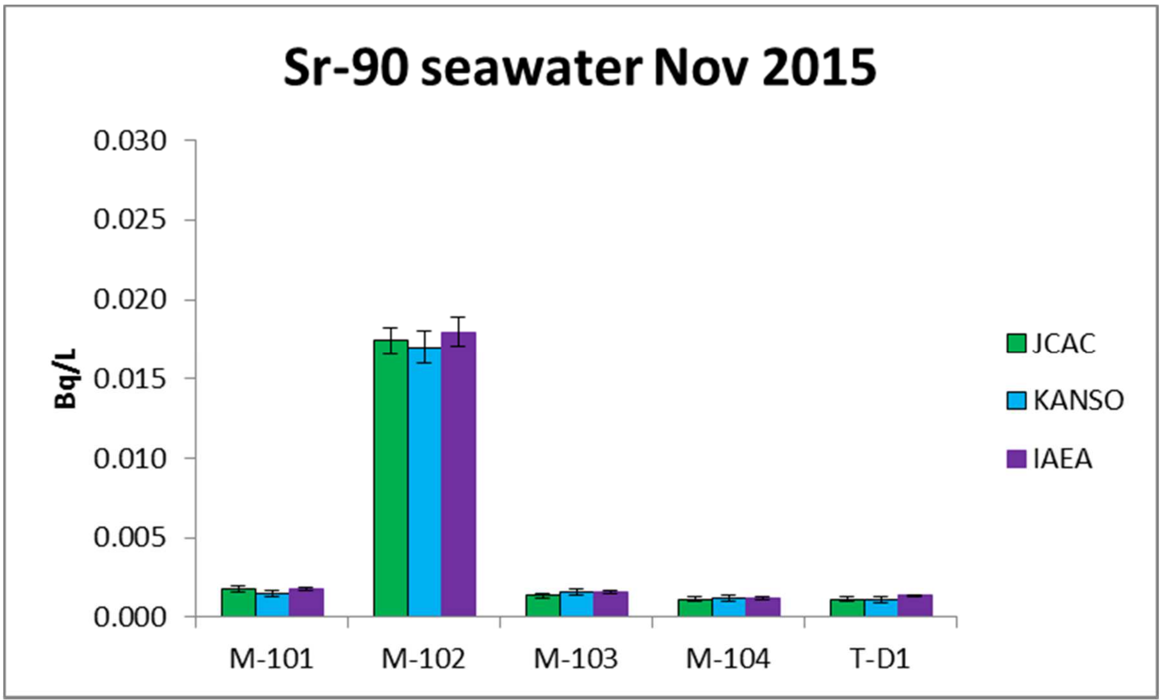

FIG. 11. Activity concentrations of ^{90}*Sr in seawater samples.*

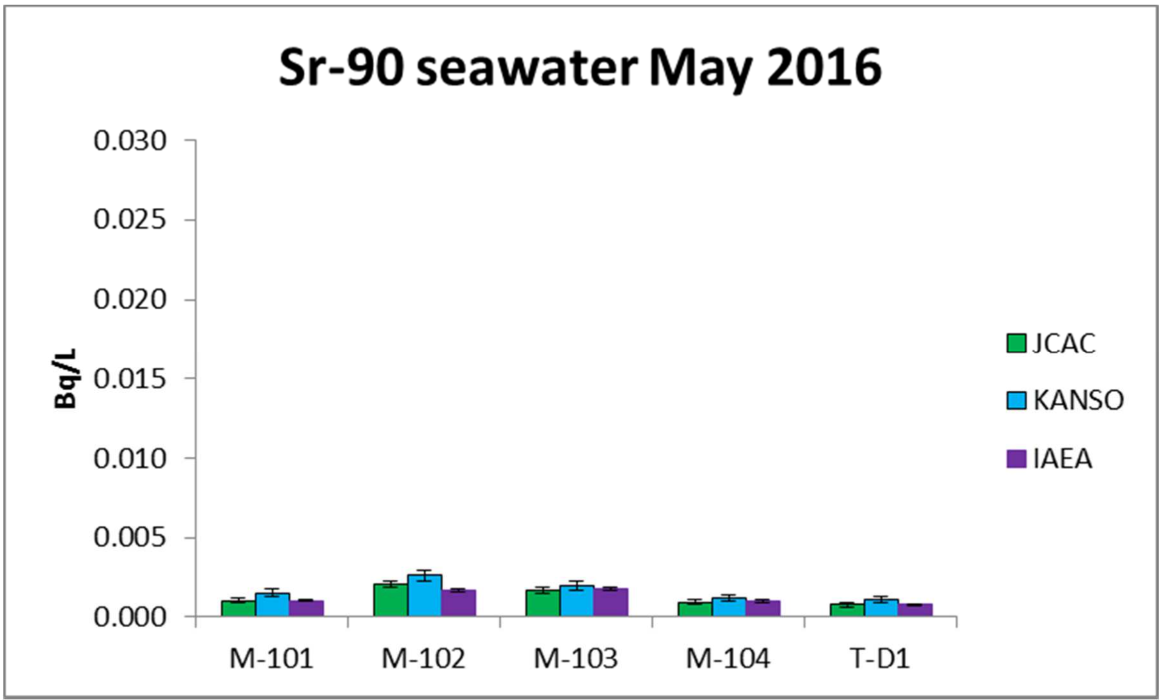

FIG. 12. Activity concentrations of ^{90}Sr in seawater samples.

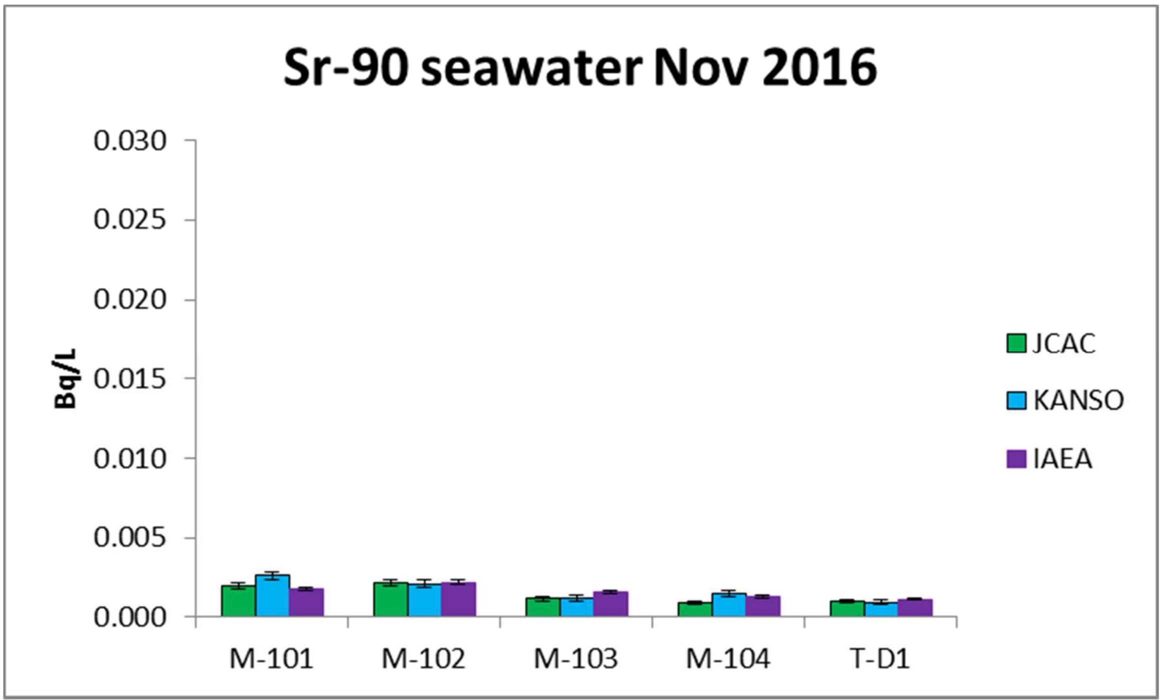

FIG. 13. Activity concentrations of ^{90}Sr in seawater samples.

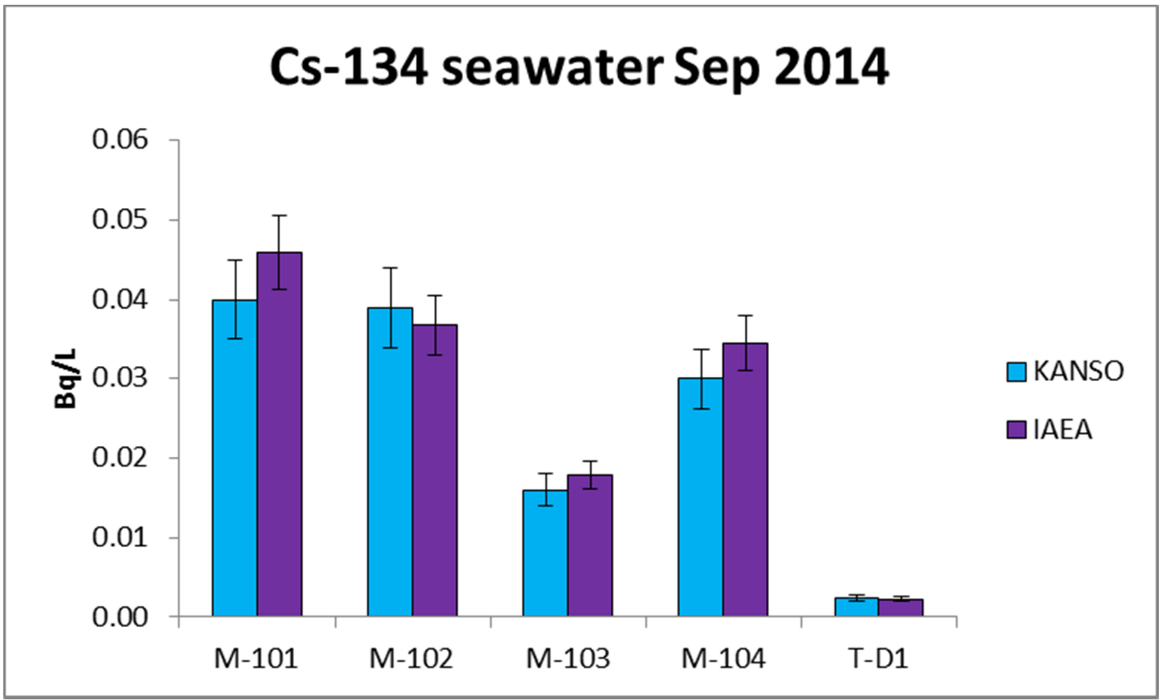

FIG. 14. Activity concentrations of ^{134}Cs in seawater samples.

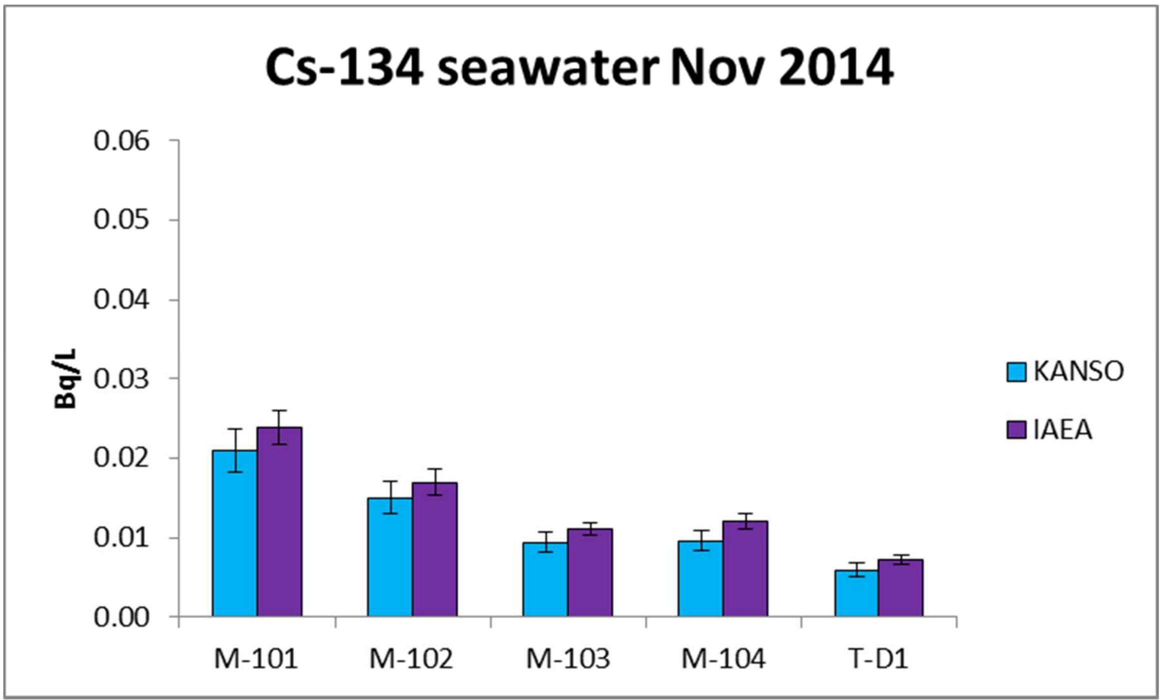

FIG. 15. Activity concentrations of ^{134}Cs in seawater samples.

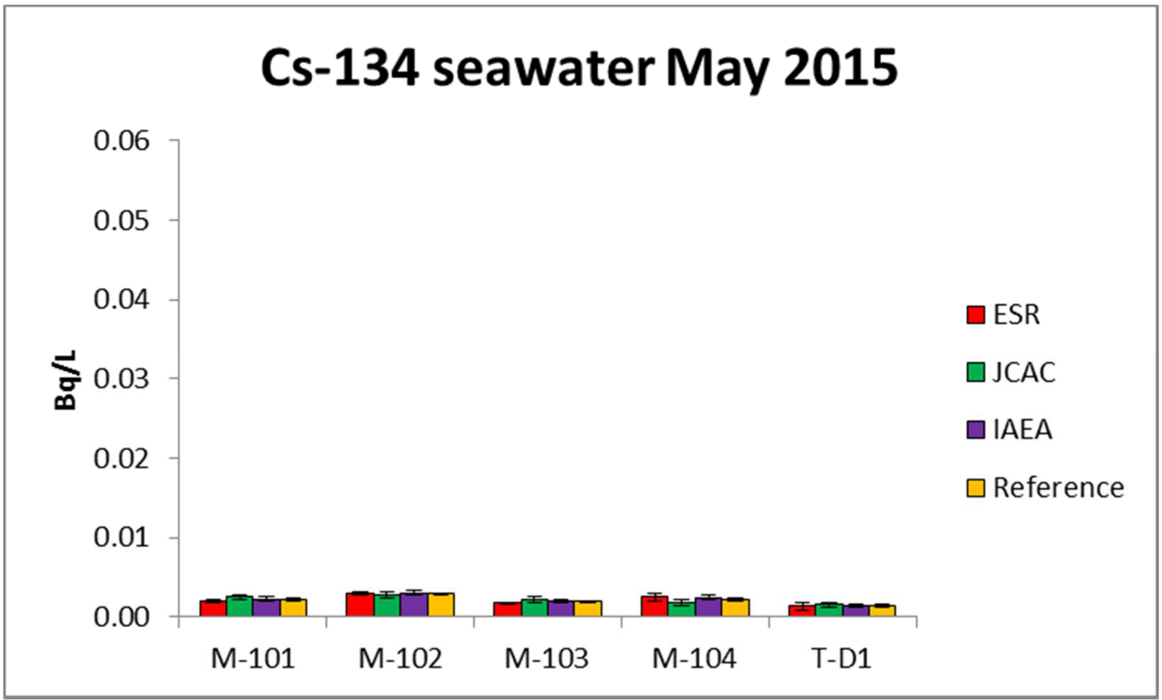

FIG. 16. Activity concentrations of ^{134}*Cs in seawater samples.*

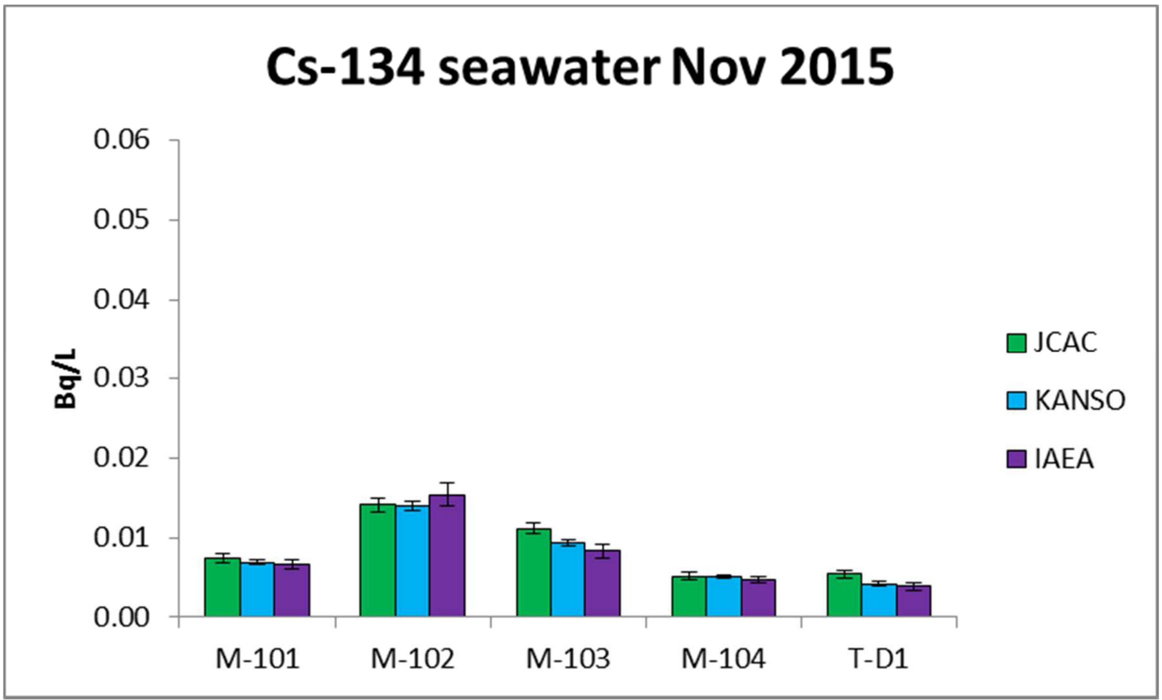

FIG. 17. Activity concentrations of ^{134}*Cs in seawater samples.*

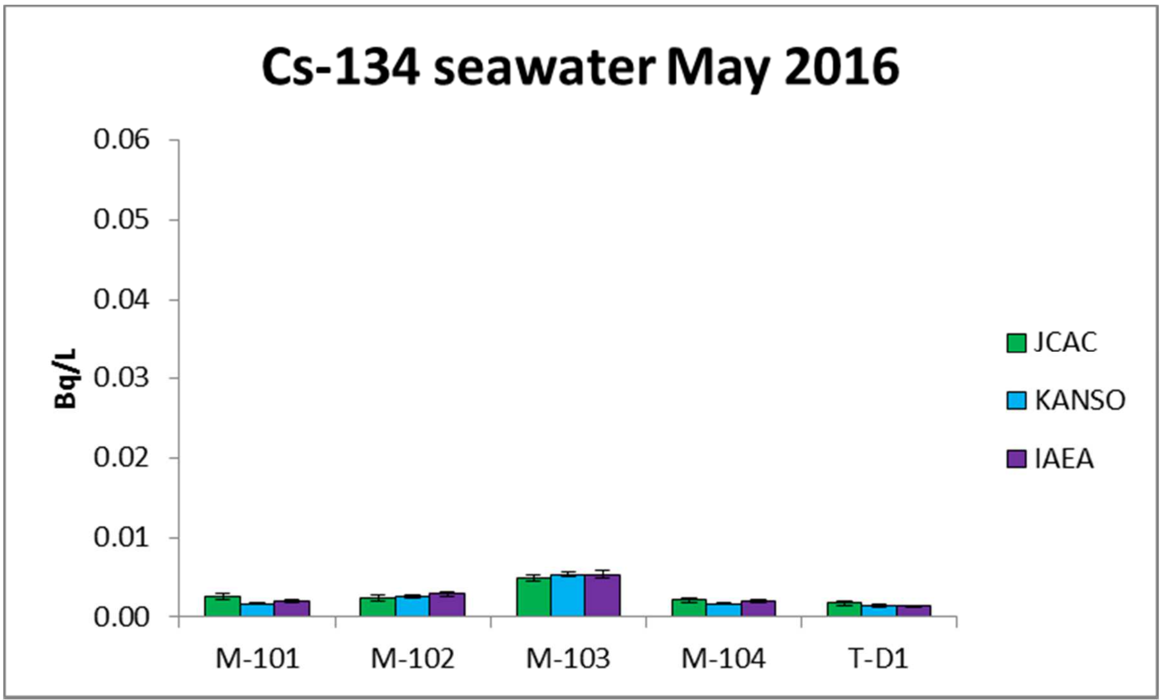

FIG. 18. Activity concentrations of ^{134}Cs in seawater samples.

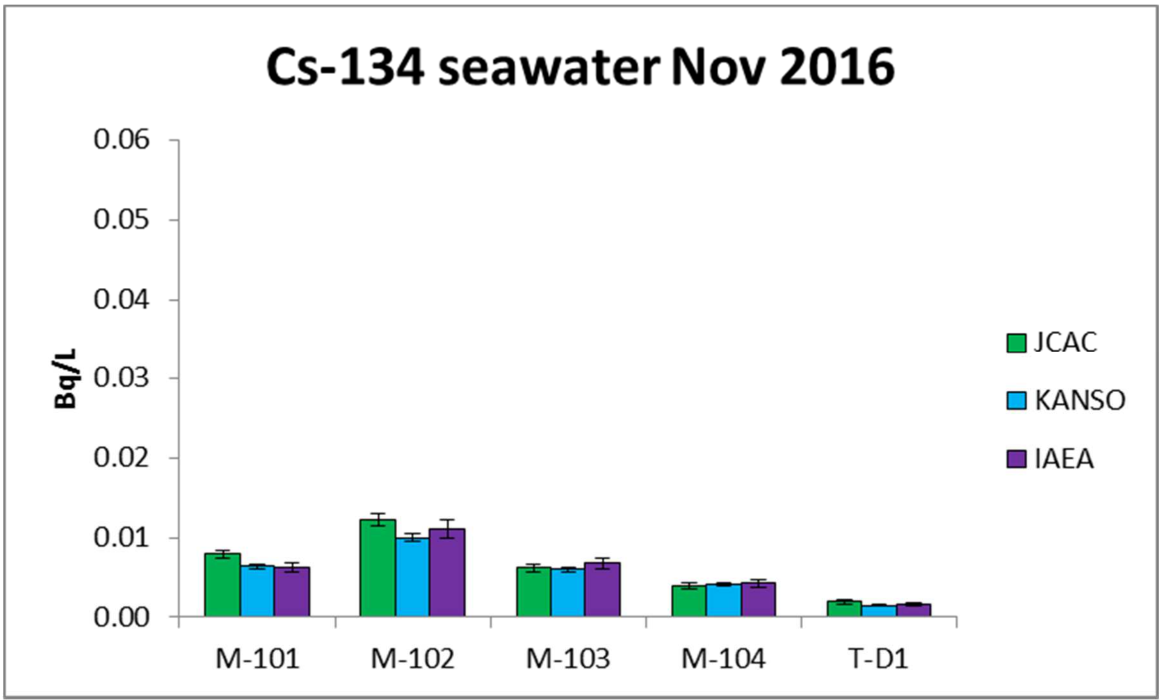

FIG. 19. Activity concentrations of ^{134}Cs in seawater samples.

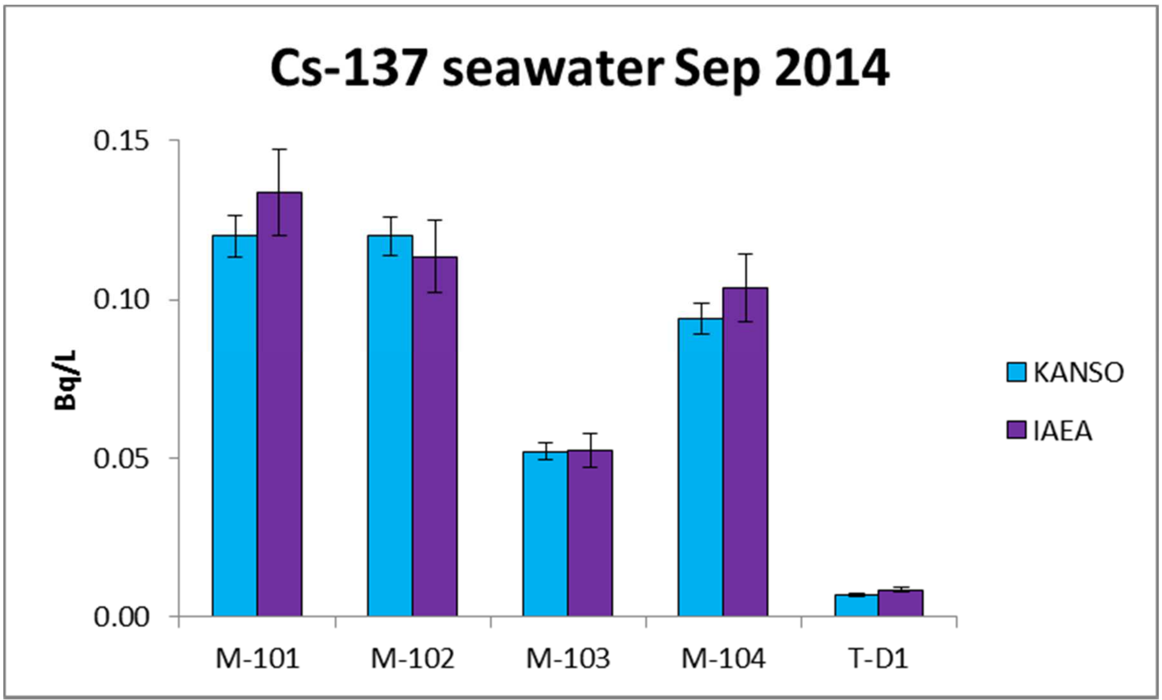

FIG. 20. Activity concentrations of ^{137}Cs in seawater samples.

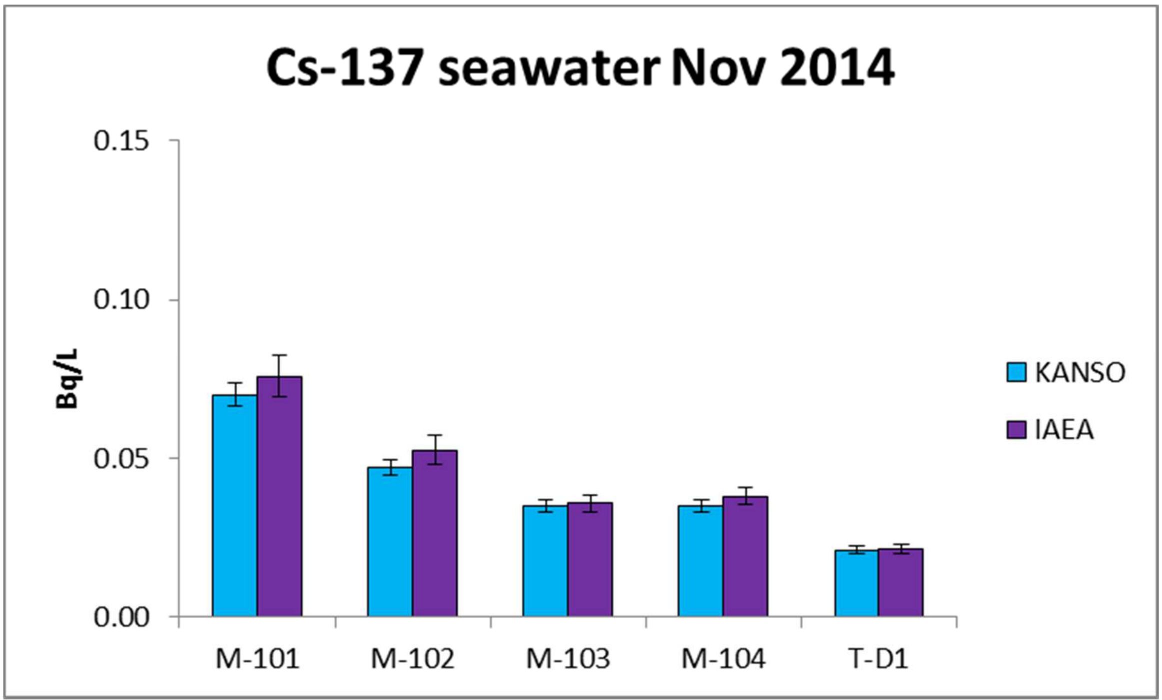

FIG. 21. Activity concentrations of ^{137}Cs in seawater samples.

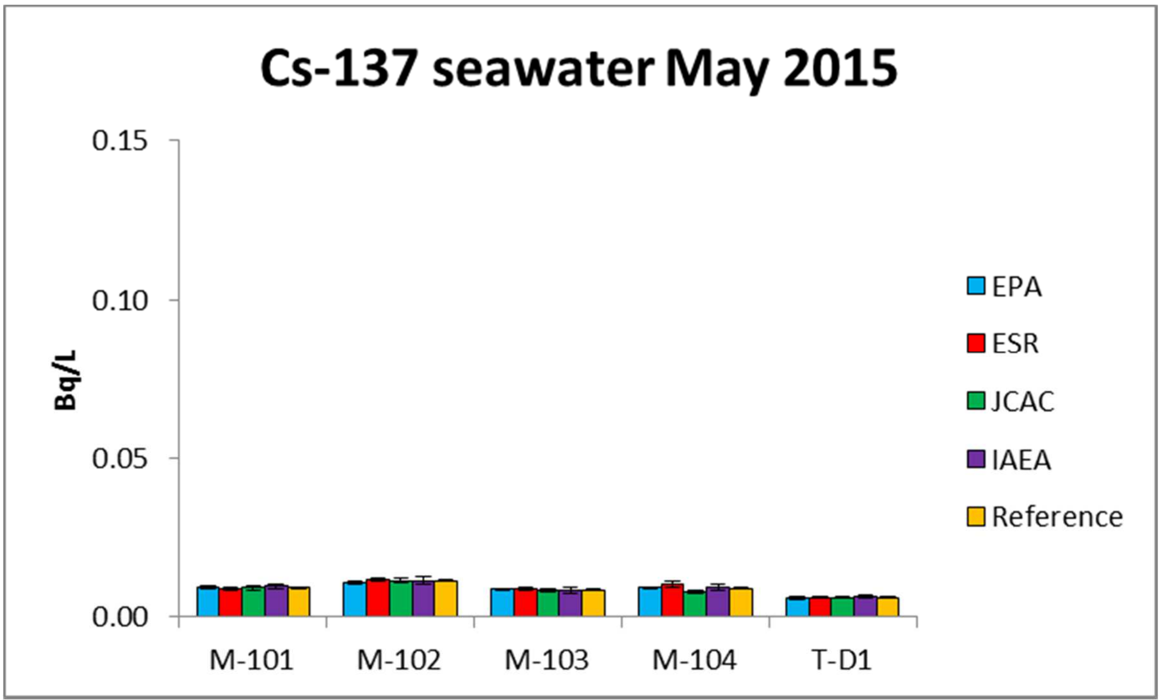

FIG. 22. Activity concentrations of ^{137}Cs in seawater samples.

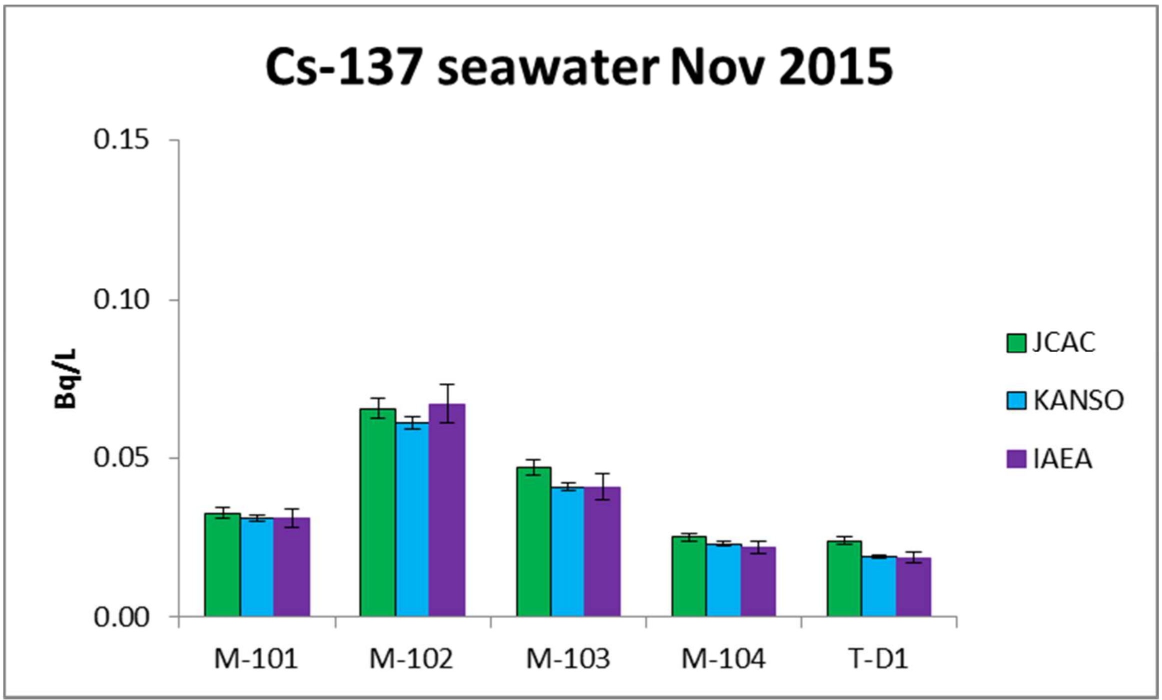

FIG. 23. Activity concentrations of ^{137}Cs in seawater samples.

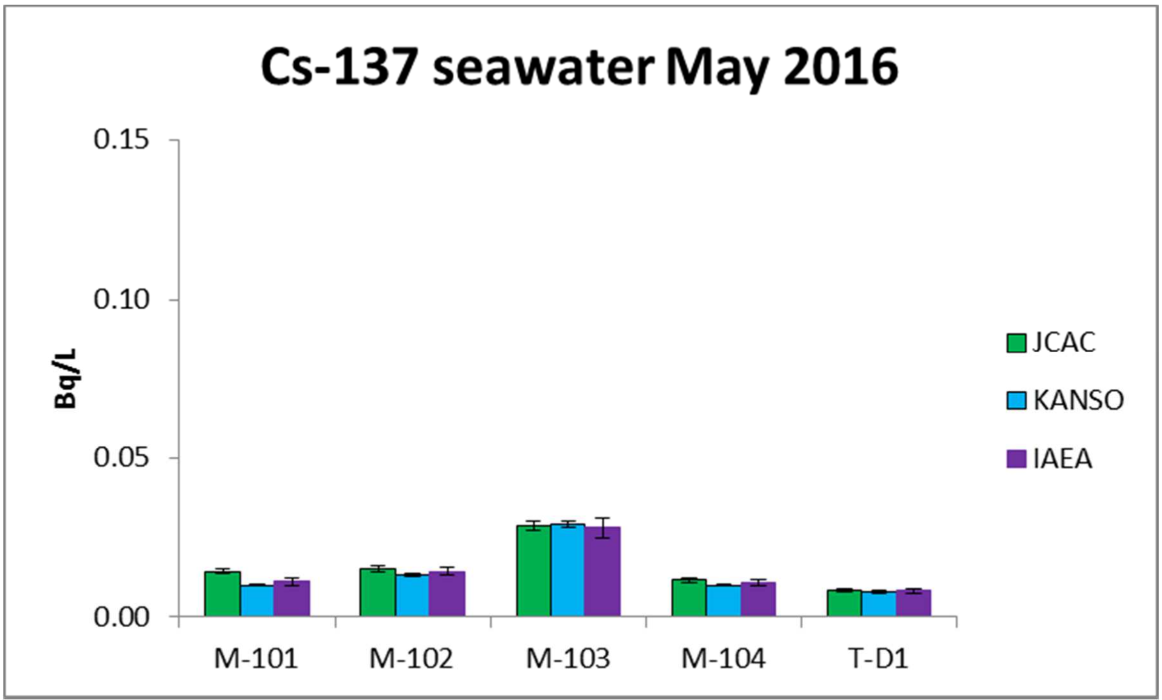

FIG. 24. Activity concentrations of ^{137}*Cs in seawater samples.*

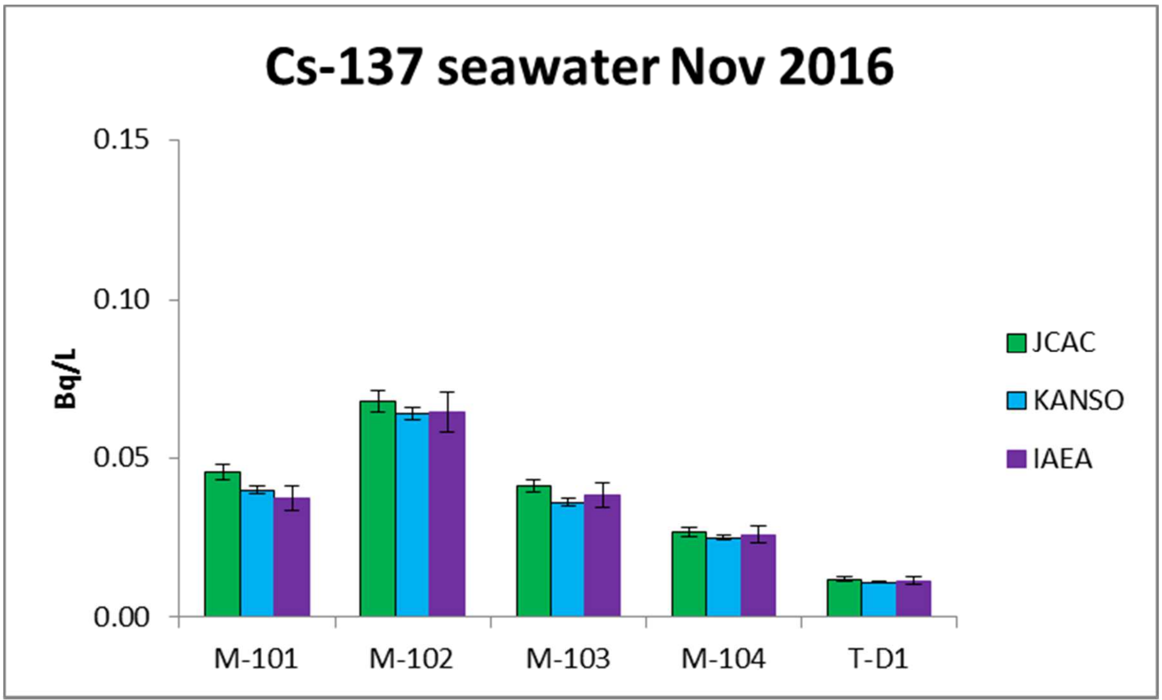

FIG. 25. Activity concentrations of ^{137}*Cs in seawater samples.*

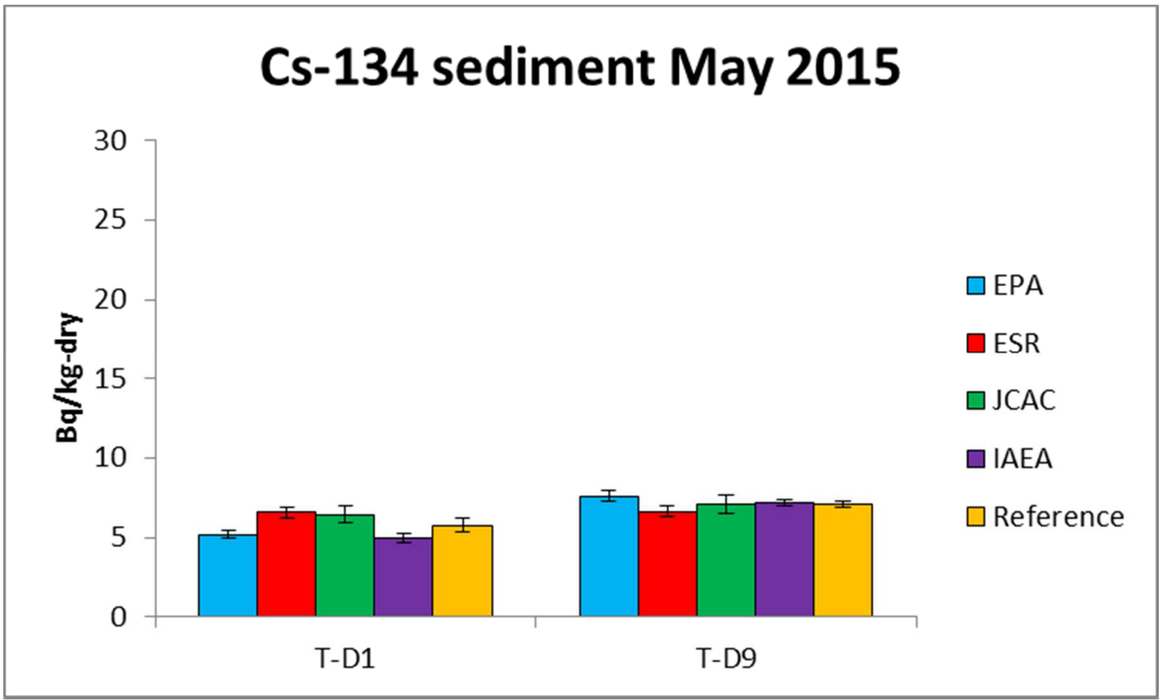

FIG. 26. Massic activities of ^{134}*Cs in sediment samples.*

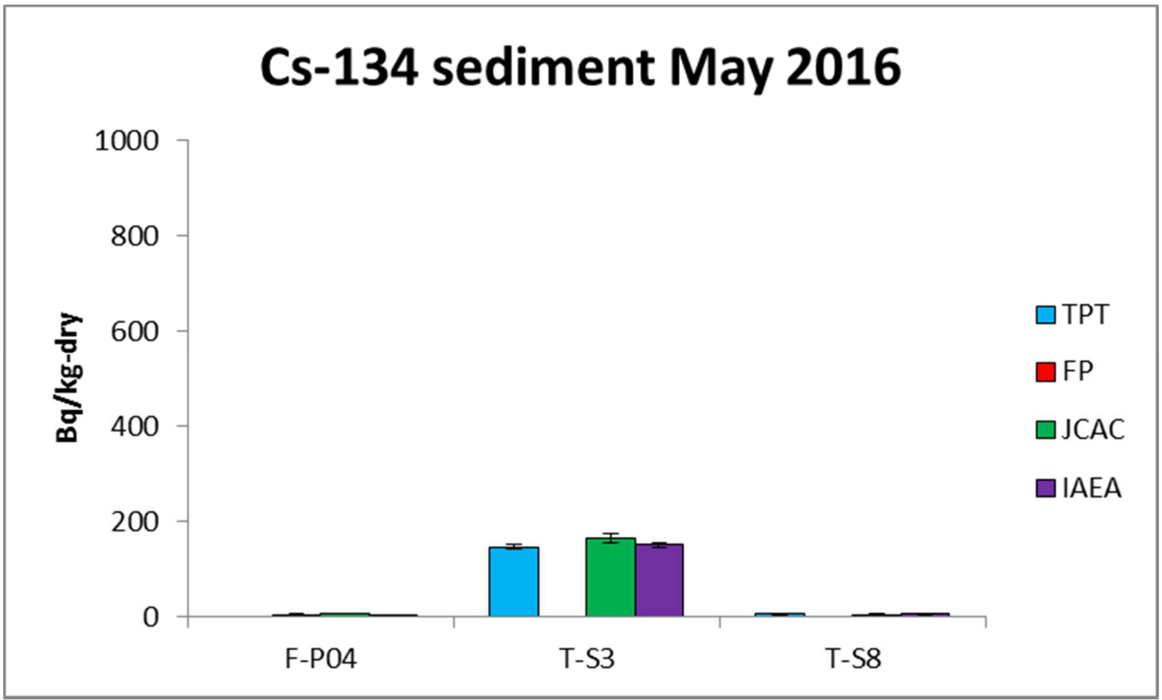

FIG. 27. Massic activities of ^{134}*Cs in sediment samples.*

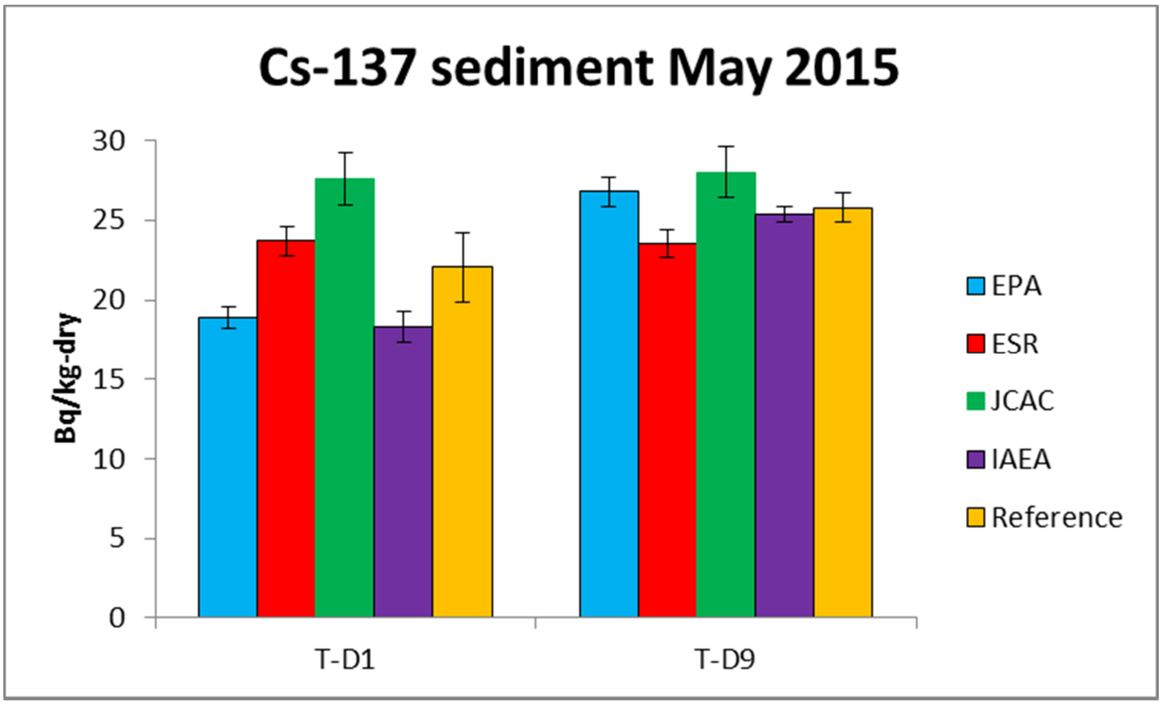

FIG. 28. Massic activities of ^{137}Cs in sediment samples.

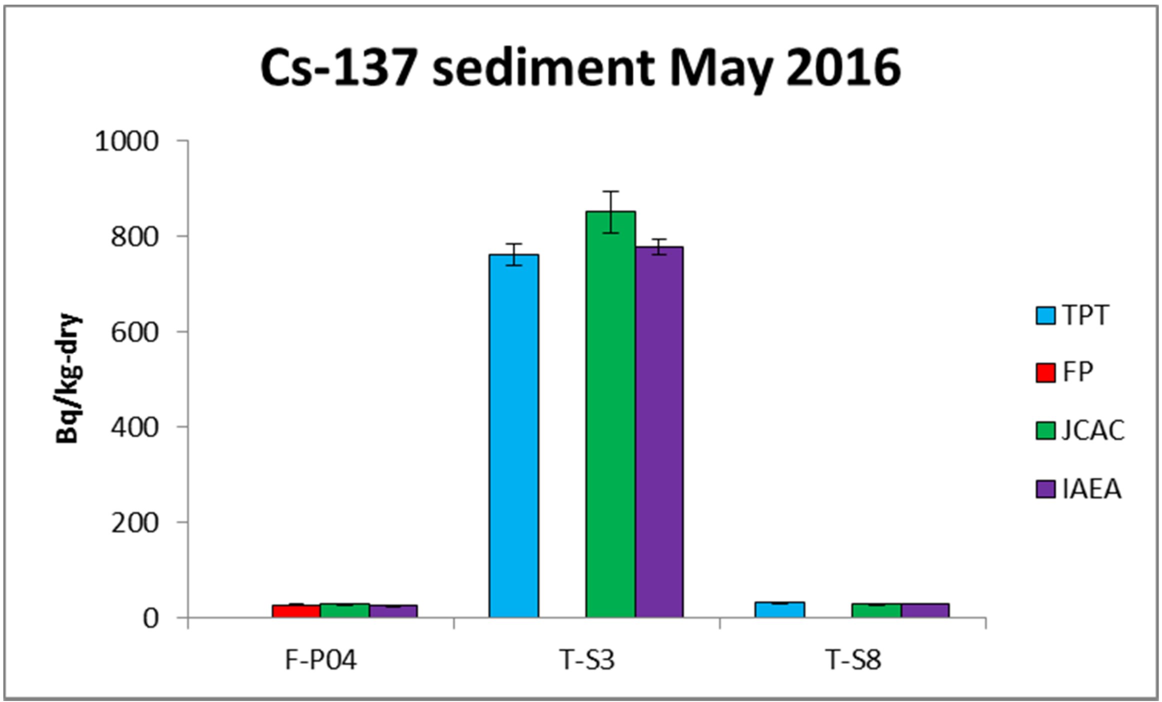

FIG. 29. Massic activities of ^{137}Cs in sediment samples.

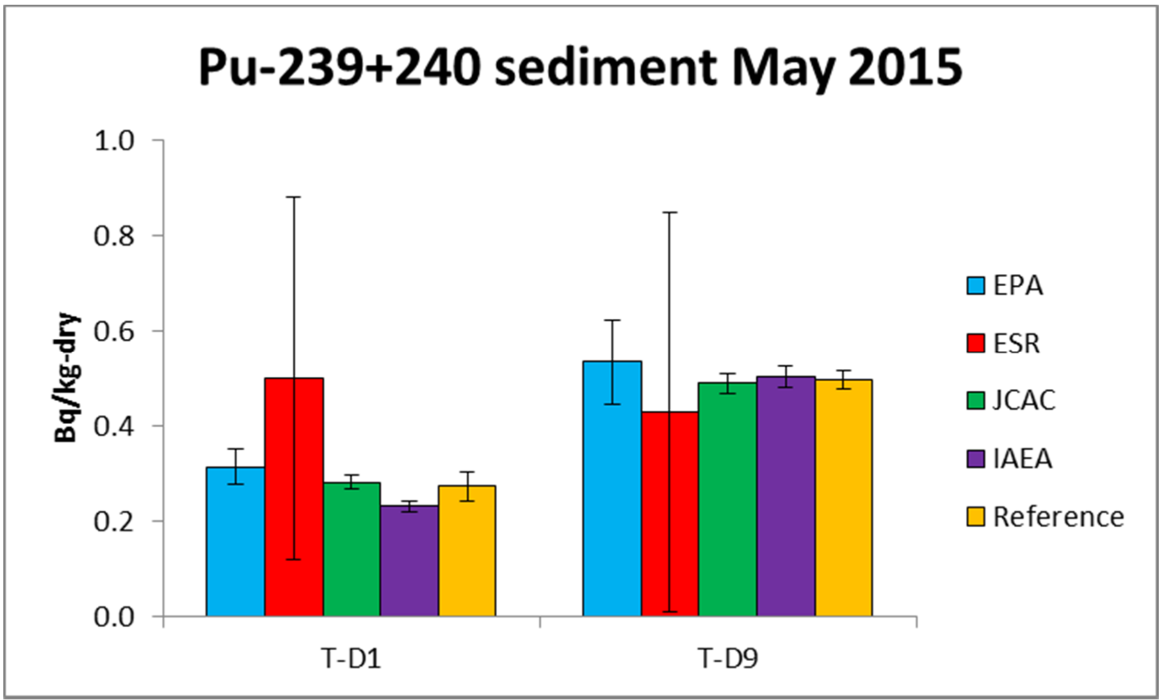

FIG. 30. Massic activities of $^{239+240}$*Pu in sediment samples.*

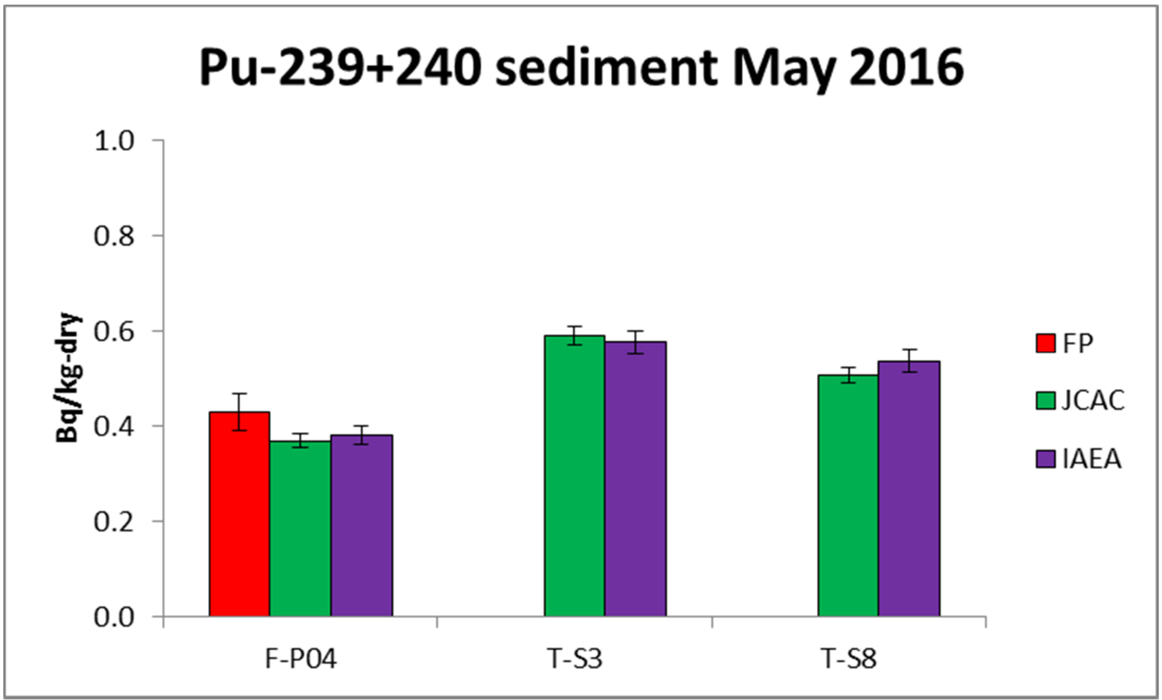

FIG. 31. Massic activities of $^{239+240}$*Pu in sediment samples.*

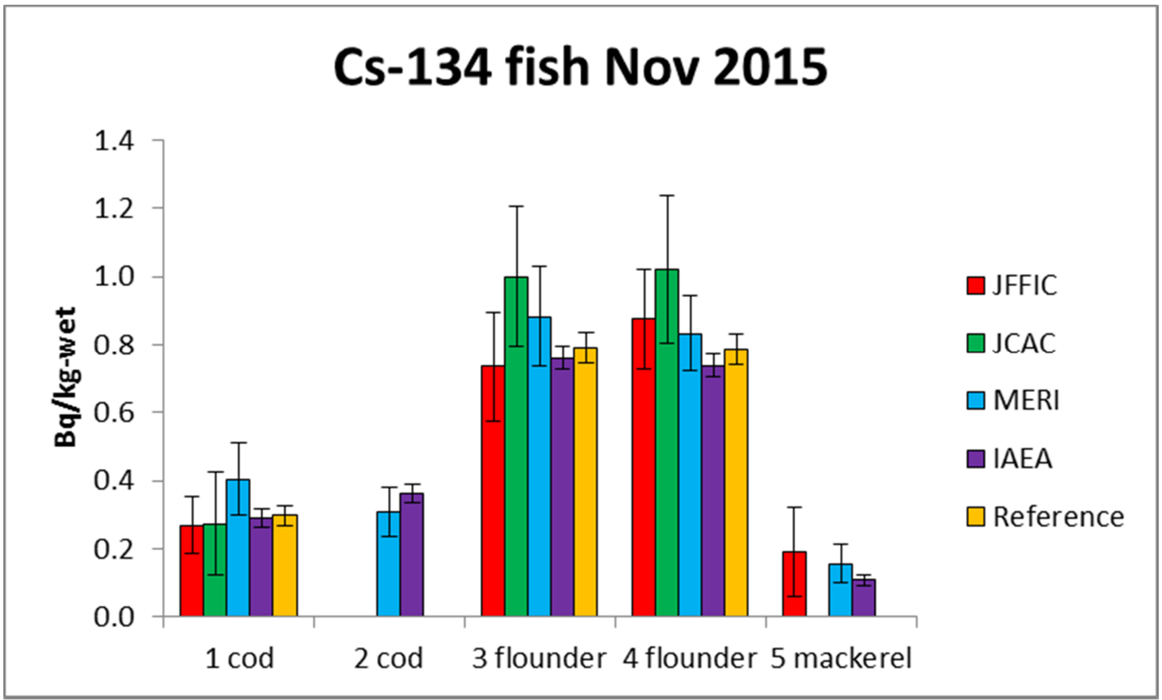

FIG. 32. Massic activities of ^{134}Cs in fish samples.

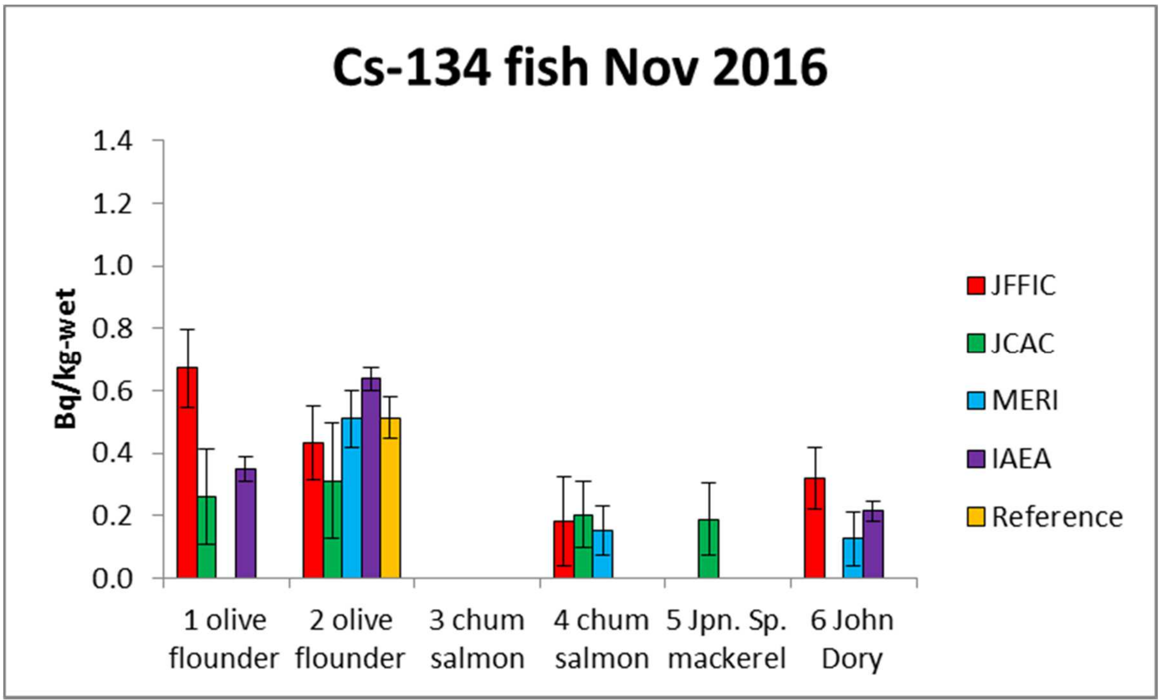

FIG. 33. Massic activities of ^{134}Cs in fish samples.

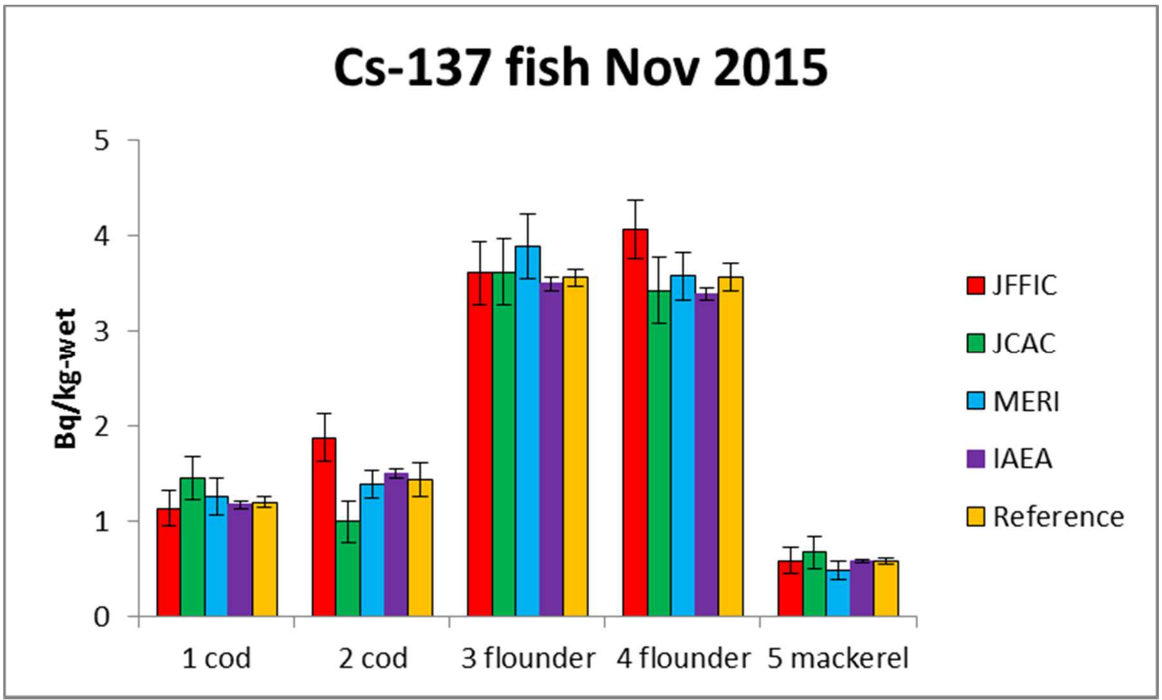

FIG. 34. Massic activities of ^{137}Cs in fish samples.

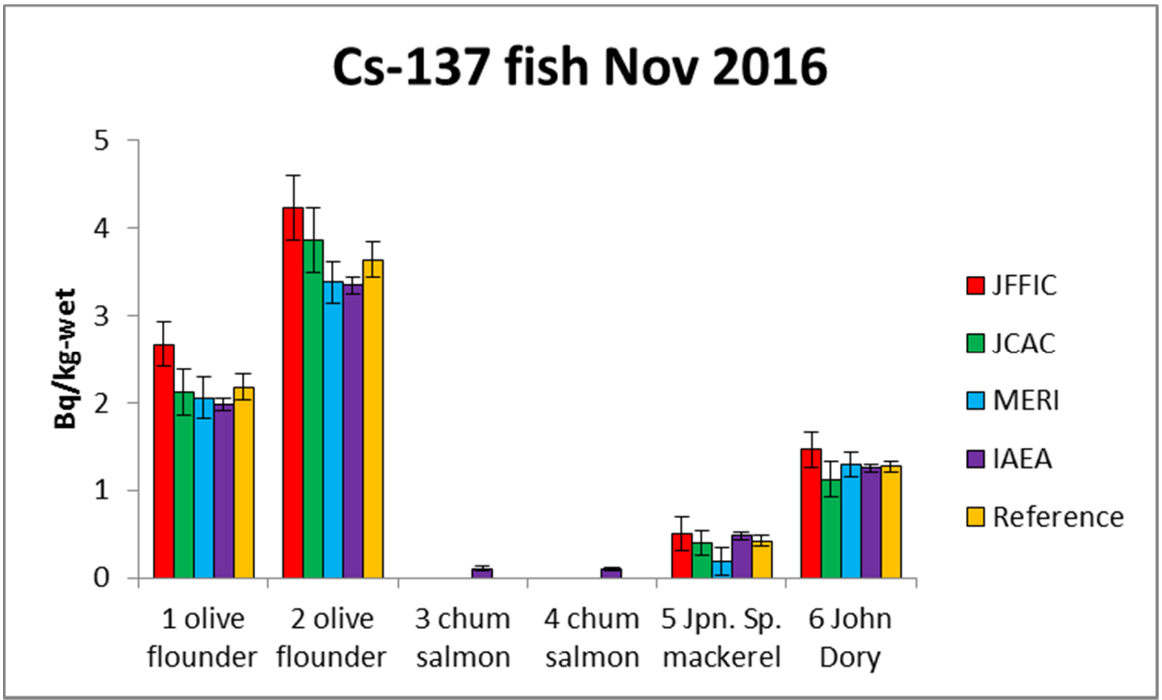

FIG. 35. Massic activities of ^{137}Cs in fish samples.