IAEA Review of Safety Related Aspects of Handling ALPS Treated Water at TEPCO's Fukushima Daiichi Nuclear Power Station

Third Interlaboratory Comparison on the Determination of Radionuclides in ALPS Treated Water

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EXECUTIVE SUMMARY

In 2021, the IAEA started its review of safety related aspects of handling ALPS (Advanced Liquid Processing System) treated water at TEPCO's Fukushima Daiichi Nuclear Power Station (FDNPS). Consistent with the request from the Government of Japan, the IAEA statutory functions and the mandate of the Task Force, the scope of the IAEA review is tailored to assessing safety related aspects of the implementation of Japan's *Basic Policy on Handling of ALPS Treated Water at the Tokyo Electric Power Company's Holdings' Fukushima Daiichi Nuclear Power Station* against the IAEA's Safety Standards. The current approach outlined in the Basic Policy is to conduct a series of controlled discharges of ALPS treated water into the sea ('batch discharges') over a period of decades.

Consistent with the relevant IAEA's Safety Standards, TEPCO is required to determine the characteristics and activity of the ALPS treated water (e.g., through the radiological environmental impact assessment) to be discharged into the sea, and to establish and implement monitoring programmes to ensure that public exposure due to the discharges is adequately assessed and that the assessment is sufficient to verify and demonstrate compliance with the authorization granted by the NRA.

To conduct its safety review, the IAEA has organized the work of the Task Force into three main components, the assessment of protection and safety; regulatory activities and processes; and sampling, independent analysis and data corroboration. The latter activities include three elements:

- Sampling, analysis and interlaboratory comparison for ALPS treated water from the FDNPS.
- Sampling, analysis and interlaboratory comparison for environmental samples (e.g., seawater, fish) from the surrounding environment of FDNPS.
- Assessment of the capabilities of dosimetry service providers involved in the monitoring of internal and external radiation exposure of workers at FDNPS.

The IAEA's sampling, independent analysis and data corroboration activities also include a review of sampling and analytical methods used by TEPCO and any other relevant technical institutions.

The corroboration of source and environmental monitoring conducted by TEPCO and other Japanese ministries and organizations is based on interlaboratory comparisons (ILCs). ILCs, along with proficiency tests (PTs), are standard methods for laboratories to assess the quality of their measurement results in comparison with those of other participating laboratories, and to identify any potential improvements. PTs involve the evaluation of performance against pre-established criteria whereas ILCs involve the organization, performance, and evaluation of measurements on the same or similar items by two or more laboratories in accordance with predetermined conditions.

For this third ILC to corroborate the results of source monitoring under the IAEA's ALPS safety review, ALPS treated water samples were taken in June 2024 from the K4-C tank group, part of the measurement and confirmation facility at FDNPS. The water contained in the K4-C tank group was being prepared to be the eighth batch of ALPS treated water to be discharged, subject to a compliance with authorized limits as demonstrated by source monitoring.

The focus of the analysis efforts for this ILC were on the radionuclides from the source term (Table 1) which are included in the radiological environmental impact assessment conducted by TEPCO. A source term refers to the amount and isotopic composition of radioactive material that is released, in this case as part of a controlled discharge. It is used for modelling releases of radionuclides into the environment and allows for the estimation of doses to be included as part of the radiological environmental impact assessment. The source term also defines the radionuclides that should be included in the respective source monitoring plan and informs the requirements for environmental monitoring. The objective of the ILC was to assess TEPCO's capability to undertake analyses relevant to source monitoring with respect to discharges of ALPS treated water to the required standard and to report high quality and comparable results.

Analyses were undertaken by TEPCO and by the following three participating IAEA Nuclear Sciences and Applications Laboratories:

- IAEA Marine Environment Laboratories, Radiometrics Laboratory (RML), Monaco;
- Terrestrial Environmental Radiochemistry Laboratory (TERC), Seibersdorf, Austria;
- Isotope Hydrology Laboratory (IHL), Vienna, Austria.

Additionally, under the coordination of the participating IAEA laboratories, selected third-party laboratories, members of the network of Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA) with demonstrable competence in the methods required, also conducted analyses of samples as participants in the ILCs. ALMERA is a network comprising more than 200 member laboratories globally. It is coordinated jointly by RML and TERC and provides a platform for maintaining and developing capability on the determination of radionuclides in air, water, soil, sediment and vegetation that can be used for both routine and environmental emergency monitoring in the IAEA Member States.

The laboratories participating in this ILC were:

- China Institute for Radiation Protection (CIRP), People's Republic of China
- Los Alamos National Laboratory (LANL), United States of America
- Korea Institute of Nuclear Safety (KINS), Republic of Korea
- Spiez Laboratory (Labor Spiez, SPIEZ), Switzerland

The results of the analyses undertaken at each laboratory were reported to the IAEA. For results that could be intercompared (i.e. for radionuclides for which activity concentrations above detection limits were reported by at least two laboratories) a statistical evaluation to assess agreement was carried out by the IAEA. The method used for the statistical evaluation was based on techniques currently used by the International Bureau of Weights and Measures' (BIPM) Consultative Committee for Ionizing Radiation, Section II: Measurement of Radionuclides, CCRI(II) and, thus, adhered to best international practice.

For other radionuclides, the detection limits reported by participating laboratories were compared to evaluate whether the analytical methods used by TEPCO were broadly equivalent and thus appropriate and fit for purpose.

The results are presented in tables and charts in this report. Reference is made to the relevant regulatory limit for discharge to sea for each radionuclide as appropriate.

The key findings of this ILC are:

- TEPCO has reported accurate results that demonstrate a high degree of proficiency.
- TEPCO's sample collection procedures follow the appropriate methodological standards required to obtain representative samples.
- The selected analytical methods utilized by TEPCO for different radionuclides were appropriate and fit for purpose. The reported detection limits were less than 1% of the respective regulatory limits for all radionuclides included in TEPCO’s source term.

The IAEA notes that these findings provide confidence in TEPCO’s capability for conducting reliable and high-quality source monitoring related to the discharge of ALPS treated water. Furthermore, based on the observations of the IAEA, TEPCO has demonstrated that they have a sustainable and robust analytical system in place to support the ongoing technical needs at FDNPS during the discharge of ALPS treated water.

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1. INTRODUCTION

1.1. BACKGROUND

The corroboration of a representative subset of the radioactivity measurement results reported by TEPCO and relevant Japanese authorities during both the pre-operational and the operational phases of discharge of ALPS treated water to the sea, and a review of the methods for related sampling and analysis used by TEPCO and relevant Japanese authorities is being undertaken by the IAEA. This corroboration provides an independent check of the veracity of the radiological data resulting from source and environmental monitoring programmes related to the ALPS discharges upon which the safety related aspects of the discharges of ALPS treated water are being evaluated. The scope of the corroboration includes an independent check of the radiological characterization of the discharges. A primary objective is to promote transparency and provide sound information to enable interested parties to evaluate the radiological data used as the basis for planning the discharges of ALPS treated water into the sea.

The corroboration of source and environmental monitoring is based on interlaboratory comparisons (ILCs). ILCs, along with proficiency tests (PTs), are standard methods for laboratories to assess the quality of their measurement results in comparison with those of other participating laboratories, and to identify any potential improvements. PTs involve the evaluation of performance against pre-established criteria whereas ILCs involve the organization, performance and evaluation of measurements on the same or similar items by two or more laboratories in accordance with predetermined conditions [1].

This publication reports the results of the third ILC to corroborate source monitoring.

This ILC was based on samples of ALPS treated water collected from the K4-C tank group in the measurement and confirmation facility at FDNPS in June 2024 by TEPCO personnel with IAEA observation. The results of the analyses were reported to the IAEA. A data compilation and a statistical evaluation to assess agreement was then carried out by the IAEA. The ILC facilitated comparisons of the measurement results of TEPCO with those of the IAEA laboratories and with selected third-party laboratories from the IAEA ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity) [2]. The objective of the ILC was to assess TEPCO’s capability to undertake analyses relevant to its source monitoring to the required standard and to report high quality and comparable results.

The IAEA wishes to thank all the laboratories that took part in this interlaboratory comparison and the Japanese organisations that provided logistical support. The IAEA is also grateful to the Government of Monaco for its support.

1.2. OBJECTIVE

This publication reports the results of the third ILC to corroborate source monitoring related to the discharge of ALPS treated water at FDNPS.

1.3. SCOPE

This publication reports all aspects of the third ILC to corroborate source monitoring related to the discharge of ALPS treated water at FDNPS including: the ILC design; participating laboratories; the methods employed for sampling and for distribution of the samples between participating laboratories; the analytical methods used by each participating laboratory to determine activity concentrations of radionuclides in the samples; the methodology employed for the statistical evaluation of the results; and the results and conclusions.

1.4. STRUCTURE

This publication contains descriptions of the ILC design and participating laboratories (Section 2); the methods employed for the sampling of ALPS treated water and for distribution of the samples between participating laboratories (Section 3), the methods used by each participating laboratory to determine activity concentrations of radionuclides in the samples (Section 4) and the methodology employed for the statistical evaluation of the results (Section 5). The results of the ILCs are presented in Section 6 and conclusions in Section 7. Key results are also presented in charts in Appendices II and III.

2. DESIGN AND PARTICIPATING LABORATORIES

2.1. DESIGN

The ILC was comprised of quantitative comparisons of the results of analyses using statistical methods to assess any differences between TEPCO’s results and those of the IAEA laboratories and the participating third-party laboratories. Comparisons have been undertaken for all radionuclides that are included in TEPCO’s ALPS treated water source term and for which activity concentrations above detection limits were reported by at least two laboratories. The specific radionuclides in the source term – those that have been identified as requiring measurement and assessment within TEPCO’s source monitoring plan [3] – are listed in Table 1. It should be noted that the source term was augmented with an additional radionuclide, ^{113m}Cd, in August 2024 during the implementation of the ILC. TEPCO was required to analyse for all radionuclides in its source term. The IAEA analysed for as many radionuclides as possible, subject to the analytical capability and resources available and the third-party laboratories were encouraged to do likewise. The results were statistically evaluated according to the methodology described in Section 5.

TABLE 1. RADIONUCLIDES INCLUDED IN TEPCO’S ALPS TREATED WATER SOURCE TERM

^{3}H	^{90}Sr	^{129}I	^{155}Eu	^{241}Pu
^{14}C	^{90}Y	^{134}Cs	^{234}U	^{241}Am
^{54}Mn	^{99}Tc	^{137}Cs	^{238}U	^{244}Cm
^{55}Fe	^{106}Ru	^{144}Ce	^{237}Np	
^{60}Co	^{113m}Cd	^{147}Pm	^{238}Pu	
^{63}Ni	^{125}Sb	^{151}Sm	^{239}Pu	
^{79}Se	^{125m}Te	^{154}Eu	^{240}Pu	

Laboratories participating in the ILC were given freedom to determine activity concentrations of radionuclides by any appropriate method of their choice. Approximate activity concentrations of commonly detected radionuclides (^{60}Co, ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{129}I, ^{134}Cs, ^{137}Cs, ^{3}H, ^{14}C and ^{99}Tc) in ALPS treated water samples from the tanks at FDNPS have been published by TEPCO [4]. This data could be used by the participating laboratories to inform on selection of analytical methods. For other radionuclides, the authorised limits for discharge provided an upper bound when evaluating target detection limits [5].

Detailed instructions for this ILC were sent to all participating laboratories before shipment of the samples. Participating laboratories were requested to submit a single measurement result for each radionuclide analysed decay corrected to the sampling date (21 June 2024). A measurement result was assumed to be comprised of an activity concentration and associated uncertainty, or the detection limit, as appropriate, all expressed in Bq/L. Participating laboratories were asked to report additional information for each radionuclide as described in Appendix I.

2.2. PARTICIPATING LABORATORIES

The laboratories participating in analyses for this ILC are presented in Table 2.

IAEA analyses were undertaken at three participating IAEA Nuclear Sciences and Applications Laboratories:

- IAEA Marine Environment Laboratories, Radiometrics Laboratory (RML), Monaco;
- Terrestrial Environmental Radiochemistry Laboratory (TERC), Seibersdorf, Austria;
- Isotope Hydrology Laboratory (IHL), Vienna, Austria.

Third-party laboratories from China, Republic of Korea, Switzerland and the United States of America, members of the network of Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA) [2] with demonstrable competence in the methods required, were selected by the IAEA to participate in the ILC. ALMERA is a network comprising more than 200 member laboratories globally. It is coordinated jointly by RML and TERC and provides a platform for maintaining and developing capability on the determination of radionuclides in air, water, soil, sediment and vegetation that can be used for both routine and environmental emergency monitoring in the IAEA Member States.

The participation of each laboratory in specific analyses is presented in Table 3.

TABLE 2. LABORATORIES PARTICIPATING IN THE ILCS

Identifier	Laboratory
IAEA	IAEA Laboratories, Austria and Monaco
CIRP	China Institute for Radiation Protection, People’s Republic of China
KINS	Korea Institute of Nuclear Safety, Republic of Korea
LANL	Los Alamos National Laboratory, United States of America
SPIEZ	Spiez Laboratory (Labor Spiez), Switzerland
TEPCO	Tokyo Electric Power Company Holdings, Inc., Japan

TABLE 3. ALPS TREATED WATER SOURCE TERM RADIONUCLIDES ANALYSED BY PARTICIPATING LABORATORIES

Nuclide	IAEA	CIRP	KINS	LANL	SPIEZ	TEPCO
^{3}H	🟩	🟩	🟩		🟩	🟩
^{14}C	🟩	🟩	🟩			🟩
^{54}Mn	🟨	🟨	🟨	🟨	🟨	🟨
^{55}Fe	🟨	🟨	🟩		🟨	🟨
^{60}Co	🟩	🟩	🟩	🟩	🟩	🟩
^{63}Ni	🟩	🟩	🟩		🟩	🟨
^{79}Se						🟨
^{90}Sr[1]	🟩	🟩	🟩		🟩	🟩
^{99}Tc	🟩	🟩	🟩		🟩	🟩
^{106}Ru	🟨	🟨	🟨	🟨	🟨	🟨
^{113m}Cd[2]				🟨		🟨
^{125}Sb[3]	🟩	🟩	🟨	🟩	🟩	🟩
^{129}I	🟩	🟨	🟩	🟨	🟨	🟩
^{134}Cs	🟨	🟩	🟨	🟨	🟨	🟨
^{137}Cs	🟩	🟩	🟩	🟩	🟩	🟩
^{144}Ce	🟨	🟨	🟨	🟨	🟨	🟨
^{147}Pm						🟨
^{151}Sm						🟨
^{154}Eu	🟨	🟨	🟨	🟨	🟨	🟨
^{155}Eu	🟨	🟨	🟨	🟨	🟨	🟨
^{234}U	🟩	🟩	🟩	🟩	🟩	🟨
^{238}U	🟩	🟩	🟩	🟩	🟩	🟨
^{237}Np	🟨		🟨	🟩	🟩	🟨
^{238}Pu	🟨	🟨	🟨			🟨
^{239}Pu[4]	🟨	🟨	🟩	🟩	🟩	🟨
^{240}Pu[4]			🟩	🟩	🟩	🟨
^{241}Pu	🟨	🟨	🟩	🟩	🟩	🟨
^{241}Am	🟨		🟨	🟩	🟨	🟨
^{244}Cm					🟨	🟨

Notes: A green cell indicates that an activity concentration and associated standard uncertainty was reported, a yellow cell indicates that a detection limit was reported and a blank cell indicates that the analysis was not performed.

[1] ^{90}Y is included in TEPCO's source but it can be assumed to be in equilibrium with ^{90}Sr and present with an equal activity concentration. TEPCO reported a result for ^{90}Y derived from the measured activity concentration of ^{90}Sr on this basis.

[2] ^{113m}Cd was added to the source term in August 2024.

[3] ^{125m}Te is also included in TEPCO's source term but it can be assumed to be in equilibrium with ^{125}Sb and present with an activity concentration approximately equal to 0.23 times the activity concentration of ^{125}Sb after one year. TEPCO reported a result for ^{125m}Te derived from the measured activity concentration of ^{125}Sb on this basis.

[4] A combined value for ^{239,240}Pu was reported by IAEA and CIRP.

3. COLLECTION AND PREPARATION OF ALPS TREATED WATER SAMPLES

Samples for this ILC were collected on 21 June 2024 from the K4-C ALPS facility at FDNPS by TEPCO personnel with observation by IAEA staff members.

The K4-C tank group is one of three measurement and confirmation units in the ALPS discharge facility that are used to store each batch of water pending discharge. It is comprised of 10 individual 1,000 m^3 storage tanks that have been re-utilized for this purpose. To ensure the homogenisation of the ALPS treated water content prior to sampling, the tanks are interconnected and the contents are circulated by pumping. Additionally, the water in individual tanks is agitated. TEPCO has demonstrated the adequacy of its method for ensuring homogenisation of [6].

Before the samples for this ILC were collected, circulation and agitation was conducted by TEPCO for more than 144 hours to ensure inter sample homogeneity and, thus, suitability of the samples for the provision of comparable results of analyses. This is same method used by TEPCO to provide representative samples for source monitoring of each batch prior to discharge.

The samples were collected directly from a valve in one of interconnection pipes at K4-C. A volume of 10 L of ALPS treated water was collected for each participating laboratory: 2 x 5 L in plastic jerrycan containers. The sample containers were assigned and labelled beforehand and were immediately sealed with tamper proof tape under IAEA observation. The samples were not acidified or filtered prior to shipping.

The samples were prepared for shipping to each participating laboratory. They were received by the IAEA laboratories in Monaco and Austria, and by the participating ALMERA laboratories between July and August 2024.

4. ANALYTICAL METHODS

A general description of the analytical methods used for relevant analyses are described in this section. Specific variations are provided for each laboratory.

4.1. GENERAL PRE-TREATMENT OF THE ALPS TREATED WATER SAMPLE

Pre-treatment of the ALPS treated water samples performed by the IAEA laboratories was guided by the results of prior robustness testing with similar samples. This robustness testing was designed to examine the potential sources of variability in analyses of radionuclide activity concentrations of ALPS treated water samples due to sampling, temporary storage and transportation methods and is described in an earlier IAEA report [7]. The robustness testing indicated that while filtration at 0.45µm was a parameter that could potentially affect analyses for activity concentrations of some radionuclides, the results were not conclusive. Therefore, no predictable and, thus, preventable effect was recorded.

Therefore, at the IAEA laboratories, the pre-treatment of the samples to be analysed in this ILC followed standard protocol and best practice by separating an aliquot for volatile radionuclides (^{3}H, ^{14}C, ^{129}I), then acidifying the remaining volume to < pH 2 using nitric acid with a delay of 24 hours before analysis. This pre-treatment was designed to ensure that any particle reactive radionuclides (e.g. Pu/Th/U isotopes) present in the samples would be desorbed from the walls of the plastic sampling container and any particulates present to provide the most conservative analysis results.

Apart from the sub-sample analyzed for ^{129}I using HPGe gamma-ray spectrometry, LANL preserved the sample material by rinsing the two empty 5 L containers in which the ALPS treated water was received with 7.5 M nitric acid, which was then added to the samples.

LANL analysed whole sample gamma spectrometry without pre-treatment. All other analyses included pre-treatment comprised of full sample digestion by evaporation and addition of concentrated acids.

SPIEZ added 6 mL of concentrated HNO_3 per L of sample. CIRP adding acid to all sample material except that analysed for ^{3}H and ^{14}C.

KINS and TEPCO reported no pre-treatment of the ALPS treated water samples prior to analysis.

4.2. ^{3}H ANALYSIS

IAEA added aliquots of the samples directly into a Teflon-coated liquid scintillation counting (LSC) vials and mixed them with a scintillation cocktail. These were stored in a dark location for approximately 12 hours prior to measurement by LSC. Recovery was based on a metrologically traceable standard solution, using the same region of interest as the samples [8].

KINS, TEPCO, SPIEZ, and CIRP distilled the samples remove organic matter and interfering radionuclides while preparing samples for tritium analysis [8, 9]. After the distillation process, they mixed the samples with a scintillation cocktail (the sample-to-scintillator ratios used by the laboratories varied), followed by measurement using LSC after storing them in a dark location for approximately 12 hours. Unlike the other laboratories, the SPIEZ laboratory acidified the samples to be analysed (6mL conc. HNO_3 per L of sample). SPIEZ, KINS, and TEPCO calculated the counting efficiency of ^{3}H by using a quench curve.

4.3. ^{14}C ANALYSIS

All labs reporting results for ^{14}C, prepared the samples by creating the evolution of CO_2 followed by collection in an absorbing solution (e.g. Carbo-Sorb E) and mixed with a scintillation cocktail (e.g. PermaFluor E+). TEPCO utilized wet oxidation for the evolution of CO_2. KINS employed a pyrolizer to combust and trap oxidized samples [10, 11]. CIRP utilized wet oxidation, absorbed the CO_2 into NaOH solution and converted to $CaCO_3$ before mixing with the scintillation cocktail. All samples were measured by LSC following storage in a dark location for at least 12 hours. KINS, and TEPCO calculated the counting efficiency of ^{14}C using a quench curve.

The IAEA result foe ^{14}C was provided by subcontractor Laboratorio de Medidas de Baja Actividad de la Universidad del Pais Vasco UPV/EHU, accreditation number 350/LE560 under Entidad Nacional de Accreditacion..

4.4. GAMMA-EMITTING RADIONUCLIDE ANALYSES (^{54}Mn, ^{60}Co, ^{106}Ru, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu, ^{155}Eu)

The samples were analysed for the gamma-ray emitting radionuclides ^{54}Mn, ^{60}Co, ^{106}Ru, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu and ^{155}Eu included in TEPCO’s ALPS treated water source term by gamma-ray spectrometry using high purity germanium (HPGe) detectors by all participating laboratories. ^{106}Ru is determined by gamma-ray spectrometry via decay of its progeny ^{106}Rh.

In most cases, a single sample was prepared in a Marinelli beaker and counted without further treatment. The beaker volume varied from 1 L at IAEA, CIRP and KINS to 5 L at LANL, SPIEZ and TEPCO.

4.5. ^{55}Fe ANALYSIS

IAEA used a chemical separation with an anion exchange resin followed by purification by liquid-liquid extraction. Then the sample was mixed with a scintillation cocktail and measured by LSC.

CIRP, KINS and SPIEZ used an anion exchange resin for chemical separation of ^{55}Fe from the sample matrix followed by LSC [12].

TEPCO used the same chemical separation method but analysed the sample using a low-energy photon detector (Ge-LEPS) [13, 14].

4.6. ^{63}Ni ANALYSIS

All laboratories (IAEA, CIRP, KINS, SPIEZ and TEPCO) used a chemical separation with an anion exchange resin followed by a selective Ni extractive chromatographic resin to extract ^{63}Ni from the sample matrix. Measurements were conducted by LSC [12, 15].

4.7. ^{79}Se ANALYSIS

After selective removal of impurities and interfering nuclides by sedimentation and TEVA resin, TEPCO added hydroxylamine hydrochloride to precipitate Se. After filtering, dissolution and mixing with scintillation cocktails, ^{79}Se was measured by LSC [15].

4.8. ^{90}Sr ANALYSIS

For the analysis of ^{90}Sr, the laboratories employed various radiochemical methods and counting techniques for the removal of impurities and interfering radionuclides. Some laboratories directly calculated ^{90}Sr activity following chemical separation, while others determined it based on the progeny ^{90}Y.

The removal of impurities and interfering radionuclides can be achieved using multiple techniques. IAEA, SPIEZ and TEPCO used dicyclohexyl-18-crown-6-ether-based Sr resin and KINS used a cation exchange DGA resin [16, 17]. In addition, CIRP used P204 extraction resin for radiochemical separation.

IAEA and KINS measured ^{90}Sr by LSC, SPIEZ and CIRP used ^{90}Sr by low level gas proportional counting [18] while TEPCO used a plastic scintillation detector [19].

4.9. ^{99}Tc ANALYSIS

IAEA, CIRP, KINS and SPIEZ separated ^{99}Tc from the sample matrix using an extractive chromatographic resin [20]. IAEA and CIRP measured the sample by LSC, KINS used Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and SPIEZ used Sector Field Inductively Coupled Plasma Mass Spectrometry (sf-ICP-MS) [21].

TEPCO did not separate ^{99}Tc from the sample matrix and used a simple dilution (0.1M HNO_3) followed by analysis using ICP-MS [15].

4.10. ^{113m}Cd ANALYSIS

TEPCO performed chemical separation with an anion exchange resin followed, followed by HNO_3 recovery and counting by LSC [22].

LANL analysed for ^{113m}Cd by gamma-ray spectrometry using the method described in section 4.4.

4.11. ^{129}I ANALYSIS

The samples were analysed for ^{129}I by IAEA, CIRP, KINS, LANL and SPIEZ using the gamma-ray spectrometry method described in the previous section. TEPCO selected a different analytical method using ICP-MS after oxidation state adjustment for the determination of ^{129}I [23].

4.12. ALPHA-EMITTING RADIONUCLIDE ANALYSIS

At IAEA, CIRP, KINS and SPIEZ selective removal of impurities and interference radionuclides in the sample matrix was achieved using an extractive chromatographic resin, with recovery measured via the addition of suitable tracers. Sources for alpha-particle spectrometry were prepared by electrodeposition on stainless steel discs or via micro co-precipitation.

IAEA performed the analysis of ^{237}Np, ^{241}Am, ^{238}Pu, $^{239,\ 240}$Pu, ^{234}U and ^{238}U by alpha spectrometry.

The IAEA result for ^{241}Pu was provided by subcontractor Laboratorio de Medidas de Baja Actividad de la Universidad del Pais Vasco UPV/EHU, accreditation number 350/LE560 under

Entidad Nacional de Accreditacion. This laboratory performed the analysis by radiochemical separation followed by liquid scintillation counting.

CIRP performed the analysis of ^{238}Pu, $^{239,\,240}$Pu, ^{234}U and ^{238}U by alpha spectrometry.

KINS performed the analysis of ^{237}Np, ^{239}Pu, ^{240}Pu and ^{241}Pu by ICP-MS and ^{238}Pu, ^{234}U and ^{238}U by alpha spectrometry.

SPIEZ measured the separated actinides (^{241}Am, ^{244}Cm, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{234}U and ^{238}U) using sf-ICP-MS [24, 25].

LANL pre-treated samples using evaporation to remove tritium. Chemical separations including precipitations and anion exchange chromatography were performed and samples were measured using multi-ion counting (multi-static total evaporation) Thermal Ionization Mass Spectrometry (TIMS) for ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Pu; multi-dynamic TIMS for ^{234}U and ^{238}U; and multi-ion counting ICP-MS with standard bracketing for ^{241}Am.

TEPCO use gross alpha screening as a conservative estimate of the activity concentrations of individual alpha emitting radionuclides in the samples. Alkali and alkaline earth metal ions were separated via Fe(III) hydroxide co-precipitation. Precipitation was dissolved in HCl and Fe ions were removed by solvent extraction using 2,6-Dimethyl-4-heptanone. The aqueous phase was collected and dried into a solid. Gross alpha activity of the so-prepared sample was conducted on ZnS(Ag) plastic scintillation detector [26].

For ^{241}Pu CIRP treated the sample with nitric acid and analysed by LSC

KINS analysed for ^{241}Am by gamma-ray spectrometry using the method described in section 4.4.

4.13. OTHER RADIONUCLIDES IN TEPCO’S SOURCE TERM

For three radionuclides, ^{241}Pu, ^{147}Pm and ^{151}Sm, TEPCO reported results that have been calculated from measurements of the activity concentrations of other “reference” radionuclides ($C_{reference}$) in the source term and scaled using the ratio of the FDNPS reactor inventory estimates $I_{measureed}$ and $I_{reference}$ for the two radionuclides:

$$C_{calculated} = C_{reference}\frac{I_{calculated}}{I_{reference}}$$

^{241}Pu was calculated from measurements of ^{238}Pu, and both ^{147}Pm and ^{151}Sm from ^{154}Eu. The inventory activities and reference radionuclides are listed in TEPCO’s REIA [3]. The methodology to estimate the inventory, part of the characterisation of the ALPS treated water source that has been undertaken by TEPCO, is also described in detail the REIA. A summary is provided in an earlier IAEA report [27].

5. STATISTICAL EVALUATION OF THE RESULTS

The IAEA collected and evaluated the results reported by all laboratories participating in the ILC. The method used for the statistical evaluation was based on techniques that have been used for Key Comparisons by the International Bureau of Weights and Measures' (BIPM) Consultative Committee for Ionizing Radiation, Section II: Measurement of Radionuclides (CCRI(II)) for the past 10 years and, thus, adheres to best international practice.

For radionuclides for which measured activity concentrations above detection limits were reported by at least four laboratories, a comparison reference value x_{ref} was determined as a power-moderated mean of the combined results [28]:

$$x_{ref} = \sum_{i=1}^{N} w_i x_i$$

where $x_i$is the value reported by the laboratory i, N is the number of results reported and w_i is a normalized weighting factor.

Then, a ζ (zeta) score was calculated for each laboratory as follows.

$$\zeta = \frac{d_i}{u(d_i)}$$

where $d_i = x_i - x_{ref}$, the difference between the value reported by the laboratory x_i and the reference value x_{ref}, and $u(d_i)$ is the standard uncertainty associated with d_i.

Following the current ISO standard for statistical methods for use in proficiency testing [29], this ζ score is interpreted as follows:

A ζ score between -2 and 2 indicates that the reported result is accepted at a 95.4% confidence level;
A ζ score between 2 and 3 or between -2 and -3 is considered to give a "warning signal"
A ζ score greater than 3 or less than -3 indicates that the reported result is not accepted at a 99.7% confidence level, an "action signal"

A standard approach, according to the same ISO standard, is that an "action signal" can be taken as evidence that an anomaly requiring investigation has occurred.

For the purposes of this ILC, for ζ scores between -3 and 3, the corresponding result was evaluated as agreeing with the reference value at a 99.7% confidence level. (Therefore, results corresponding to "warning signals" were considered to agree with the reference value.) For ζ scores greater than 3 or less than -3, the corresponding result was evaluated as being discrepant at the same confidence level.

The measurement uncertainty of each result reported was further assessed to ensure that the analytical method selected by each laboratory for each analysis was fit for purpose. The precision of measurement results varies quite broadly depending on the analytical method employed, the radionuclide activity concentration and the sample matrix. However, the measurement uncertainty reported by any participating laboratory should not be excessive when compared to those for the same sample and radionuclide submitted by other laboratories. A precision check was undertaken by comparing each result reported to a multiple of $S_{50} =$

$S \times 0.67449$, a typical uncertainty for the dataset under consideration, a parameter calculated as part of the determination of the power-moderated mean. For the purposes of this ILC, a reported uncertainty of greater than five times S_{50} was considered to be statistically irrelevant.

The difference d_i between the value reported by the laboratory and the reference value and the combined uncertainty (of the reported value and the reference value) were assessed using a graphical method, the “PomPlot” [30, 31]. This is an intuitive graphical method, also used by the BIPM’s CCRI(II) for Key Comparisons, that provides a summary overview of the results reported by participating laboratories.

The PomPlot displays the difference of each reported result from the reference value on the horizontal axis and the standard uncertainty associated with each difference on the vertical axis. The red point indicates the reference value; and the green, blue and red solid lines represent ζ scores = ±1, ±2 and ±3, respectively. For both axes, the variables are expressed as multiples of S_{50}, the typical uncertainty for the dataset under consideration as defined above (Figure 1).

Values on the right-hand side of the graph correspond to results that are higher than the reference value while lower values are located on the left. When the reported uncertainty is low, the corresponding point is located high in the graph. The most accurate results should be located close to the top of the pyramid. Points outside of the ζ = ±3 lines are discrepant.

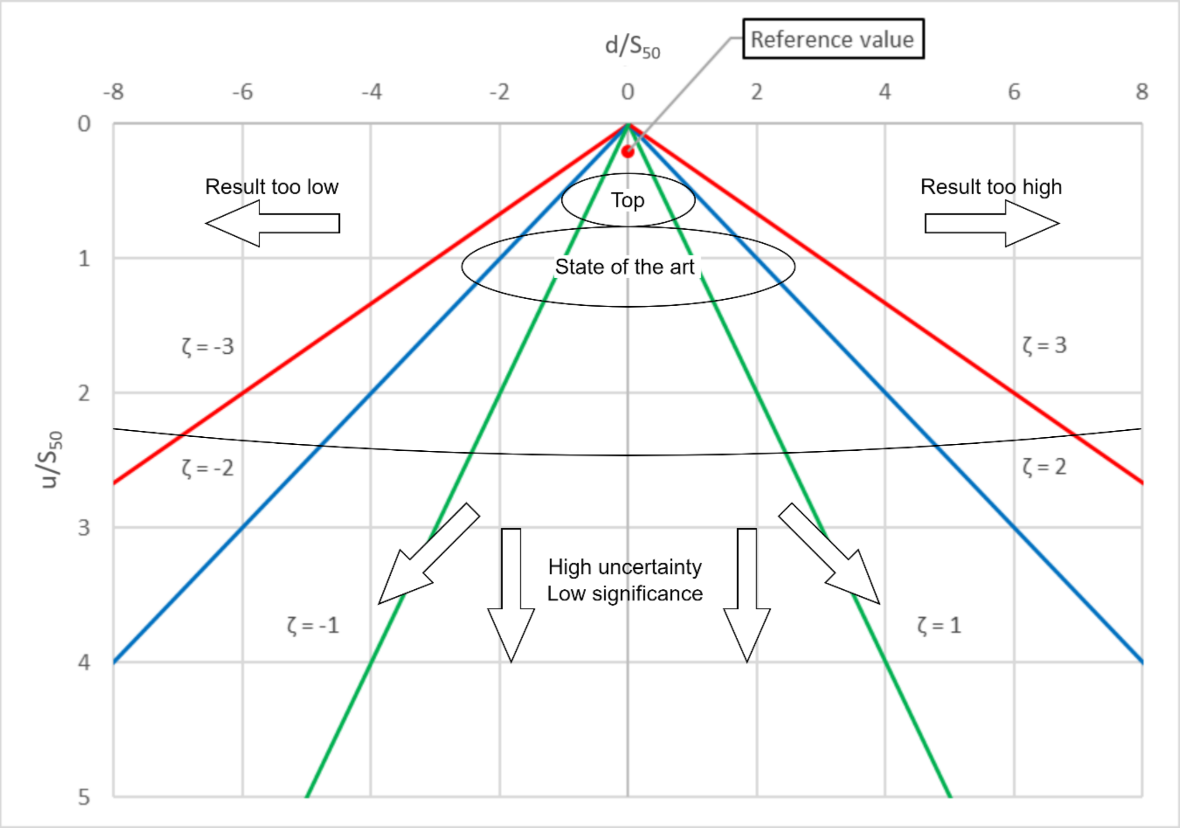

FIG. 1. Interpretation of a PomPlot (adapted from Pommé, 2006 [30]).

For radionuclides for which measured activity concentrations above detection limits were reported by two or three laboratories, then one or three zeta tests [1] were performed. The zeta $\zeta_{i,j}$ test is defined as:

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

where:

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

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

For radionuclides for which results could not be compared quantitatively (i.e. those for which activity concentrations above detection limits were reported by just one laboratory or none at all), the detection limits reported by participating laboratories were qualitatively compared to evaluate whether the analytical methods used by TEPCO were broadly equivalent and thus appropriate and fit for purpose.

Prior to publication, the compiled results for each laboratory were shared, separately, with all participating laboratories to ensure that their reported results were compiled accurately. Any discrepancies (significantly different results) identified by the statistical evaluation were carefully verified.

6. RESULTS

6.1. GENERAL

The results of the ILC are presented in this section. All reported results, with derived reference values where possible, are presented in section 6.2 (Table 4). ζ (Zeta) scores are presented in section 6.3 (Table 5) and PomPlots (Figures 2 to 11) in section 6.4. Charts showing the activity concentrations and detection limits reported are presented in Appendix III (Figures 13 to 27). Additional results for radionuclides not in TEPCO's ALPS treated water source term are presented in section 6.5.

6.1.1. Uncertainties, confidence intervals and rounding

In this report, each measurement result is stated in the format $\mathrm{x} \pm \mathrm{y}\ (z)$, where x is the activity concentration, y is the numerical value of the combined standard uncertainty, i.e. with a coverage factor of $k = 1$, and z is the detection limit of the analytical method used. In cases where an activity was not detected, the detection limit is reported in the format $< z$.

Reported activity concentrations are stated with a number of decimal places based on the first two significant figures of the associated uncertainty. Detection limits are also reported to two significant places. Both uncertainties and detection limits are also rounded up. Activity concentrations are rounded up or down according to normal rules for rounding [32].

6.1.2. Reference date

All activity concentrations for were reported at a reference date of 21 June 2024.

6.2. RESULTS REPORTED

TABLE 4. ACTIVITY CONCENTRATIONS (Bq/L) IN K4-C ALPS TREATED WATER SAMPLES

Nuclide	IAEA	CIRP	KINS	LANL	SPIEZ	TEPCO	Reference value
^{3}H	218400 ± 3000 (8.0)	208000 ± 6000 (1.6)	207000 ± 4000 (2.5)	-	228000 ± 5600 (3.5)	198200 ± 6000 (17)	212100 ± 5100
^{14}C	8.87 ± 0.71 (0.15)	10.7 ± 0.6 (0.29)	12.4 ± 2.1 (3.7)	-	-	11.54 ± 0.95 (1.6)	10.62 ± 0.73
^{54}Mn	<0.024	<0.039	<0.15	<0.018	<0.011	<0.026	-
^{55}Fe	<0.16	<0.25	0.267 ± 0.11 (0.24)	-	<0.21	<16	-
^{60}Co	0.38 ± 0.021 (0.023)	0.401 ± 0.026 (0.044)	0.373 ± 0.049 (0.15)	0.39 ± 0.02 (0.017)	0.37 ± 0.02 (0.01)	0.437 ± 0.041 (0.027)	0.387 ± 0.011
^{63}Ni	3.68 ± 0.11 (0.22)	4.09 ± 0.28 (0.15)	3.53 ± 0.34 (0.29)	-	3.8 ± 0.2 (0.07)	<8.1	3.75 ± 0.11
^{79}Se	-	-	-	-	-	<0.98	-
^{90}Sr[1]	1.219 ± 0.067 (0.056)	1.072 ± 0.07 (0.025)	1.35 ± 0.14 (0.21)	-	1.27 ± 0.061 (0.006)	1.205 ± 0.025 (0.035)	1.210 ± 0.040
^{99}Tc	0.662 ± 0.012 (0.062)	0.717 ± 0.074 (0.2)	0.492 ± 0.011 (0.096)	-	0.51 ± 0.04 (0.0009)	0.730 ± 0.081 (0.11)	0.612 ± 0.051
^{106}Ru	<0.28	<0.37	<1.4	<0.16	<0.093	<0.22	-
^{113m}Cd	-	-	-	<32	-	<0.077	-
^{125}Sb	0.204 ± 0.025 (0.065)	0.133 ± 0.021 (0.056)	<0.44	0.21 ± 0.01 (0.036)	0.23 ± 0.02 (0.04)	0.234 ± 0.038 (0.091)	0.201 ± 0.019
^{129}I	0.253 ± 0.016 (0.052)	<6.1	0.304 ± 0.062 (0.2)	<7.2	<0.36	0.289 ± 0.035 (0.060)	-
^{134}Cs	<0.028	<0.041	<0.13	<0.019	<0.013	<0.034	-
^{137}Cs	0.201 ± 0.013 (0.023)	0.316 ± 0.021 (0.043)	0.184 ± 0.024 (0.11)	0.188 ± 0.005 (0.016)	0.19 ± 0.02 (0.01)	0.222 ± 0.023 (0.027)	0.216 ± 0.021
^{144}Ce	<0.47	<0.51	<1.4	<0.075	<0.12	<0.38	-
^{147}Pm	-	-	-	-	-	<0.33	-
^{151}Sm	-	-	-	-	-	<0.013	-

TABLE 4. ACTIVITY CONCENTRATIONS (Bq/L) IN K4-C ALPS TREATED WATER SAMPLES (CONTINUED)

Nuclide	IAEA	CIRP	KINS	LANL	SPIEZ	TEPCO	Reference value
^{154}Eu	<0.089	<0.26	<0.38	<0.018	<0.028	<0.074	-
^{155}Eu	<0.13	<0.35	<0.5	<0.033	<0.051	<0.21	-
^{234}U	0.00079 ± 0.00017 (0.00025)	0.00236 ± 0.00017 (0.00011)	0.00078 ± 0.00019 (0.00028)	0.0007620 ± 0.0000045 (0.0000019)	0.00075 ± 0.000025 (0.000004)	<0.029	0.00108 ± 0.00032
^{238}U	0.00111 ± 0.00018 (0.00025)	0.00179 ± 0.00014 (0.00004)	0.00054 ± 0.00018 (0.00032)	0.000694 ± 0.00000094 (0.00000025)	0.0007 ± 0.000019 (0.0000022)	<0.029	0.00096 ± 0.00023
^{237}Np	<0.00043	-	<0.00000025	0.0000484 ± 0.00000027	0.000012 ± 0.0000031	<0.029	-
^{238}Pu	<0.00050	<0.00016	<0.00068	-	-	<0.029	-
^{239}Pu[3]	<0.00026	<0.00019	0.0000409 ± 0.000003 (0.0000017)	0.0000492 ± 0.00000024 (0.000000016)	0.000037 ± 0.0000028 (0.0000054)	<0.029	-
^{240}Pu[3]			0.0000566 ± 0.0000061 (0.0000058)	0.0000727 ± 0.0000005 (0.000000041)	0.000071 ± 0.0000067 (0.000016)	<0.029	-
^{241}Pu	<1.2	<0.019	0.00733 ± 0.00063 (0.0072)	0.007 ± 0.00005 (0.000011)	0.0063 ± 0.002 (0.0033)	<0.79	-
^{241}Am	<0.00010	-	<0.00079	0.0000602 ± 0.000003 (0.0000017)	<0.00017	<0.029	-
^{244}Cm	-	-	-	-	<0.013	<0.029	-

Notes: [1] A value of 1.2 Bq/L for ^{90}Y, derived from the measured activity concentration of ^{90}Sr, was reported by TEPCO.

[2] A detection limit of 0.034 Bq/L for ^{125m}Te derived from the measured activity concentration of ^{125}Sb, was reported by TEPCO.

[3] A combined value for ^{239,240}Pu was reported by IAEA and CIRP.

6.3. ζ (ZETA) SCORES

TABLE 5. ZETA SCORES FOR K4-C ALPS TREATED WATER SAMPLES

Nuclide	IAEA	CIRP	KINS	LANL	SPIEZ	TEPCO
^{3}H	1.14	-0.59	-0.86	-	2.37	-2.00
^{14}C	-1.49	-0.17	0.86	-	-	0.83
^{55}Fe	DL	DL	Note 1	-	DL	DL
^{60}Co	-0.42	0.57	-0.31	0.14	-0.98	1.27
^{63}Ni	-0.69	1.33	-0.71	-	0.29	DL
^{90}Sr	0.14	-2.03	1.11	-	0.98	-0.12
^{99}Tc	0.98	1.34	-2.35	-	-1.73	1.41
^{125}Sb	0.12	-2.79	DL	0.48	1.24	0.91
^{129}I	Note 2	DL	Note 2	DL	DL	Note 2
^{137}Cs	-0.67	**3.74**	-1.12	-1.35	-0.99	0.21
^{234}U	-0.87	**3.72**	-0.86	-1.02	-1.05	DL
^{238}U	0.55	**3.31**	-1.58	-1.19	-1.17	DL
^{237}Np	DL	-	DL	Note 3	Note 3	DL
^{239}Pu	DL	DL	Note 4	Note 4	Note 4	DL
^{240}Pu	DL	DL	Note 5	Note 5	Note 5	DL
^{241}Pu	DL	DL	Note 6	Note 6	Note 6	DL
^{241}Am	DL	-	DL	Note 1	DL	DL

Notes:

Evaluation criteria (99.7% confidence level): ζ score between -3 and 3: corresponding results evaluated as agreeing; ζ score greater than 3 or less than -3: corresponding results evaluated as discrepant.

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

Note 2: Values of -0.80, -0.94 and 0.21 for $\zeta_{\text{IAEA, KINS}}$, $\zeta_{\text{IAEA, TEPCO}}$ and $\zeta_{\text{KINS, TEPCO}}$, respectively, for ^{129}I.

Note 3: Value of **11.7** for $\zeta_{\text{LANL, SPIEZ}}$, for ^{237}Np.

Note 4: Values of -2.78, 0.95 and **4.42** for $\zeta_{\text{KINS, LANL}}$, $\zeta_{\text{KINS, SPIEZ}}$ and $\zeta_{\text{LANL, SPIEZ}}$, respectively, for ^{239}Pu.

Note 5: Values of -2.64, -1.59 and 0.25 for $\zeta_{\text{KINS, LANL}}$, $\zeta_{\text{KINS, SPIEZ}}$ and $\zeta_{\text{LANL, SPIEZ}}$, respectively, for ^{240}Pu.

Note 6: Values of 0.53, 0.51 and 0.36 for $\zeta_{\text{KINS, LANL}}$, $\zeta_{\text{KINS, SPIEZ}}$ and $\zeta_{\text{LANL, SPIEZ}}$, respectively, for ^{241}Pu.

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

6.4. POMPLOTS

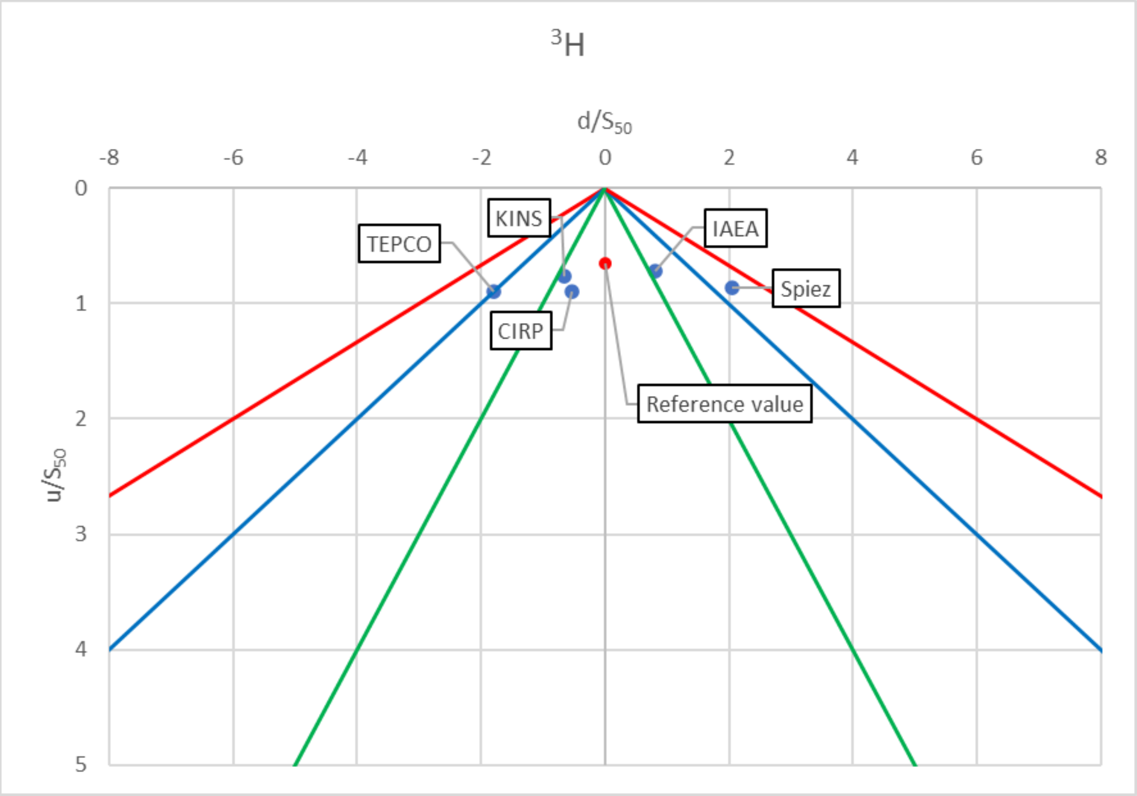

FIG. 2. PomPlot for ^{3}H results.

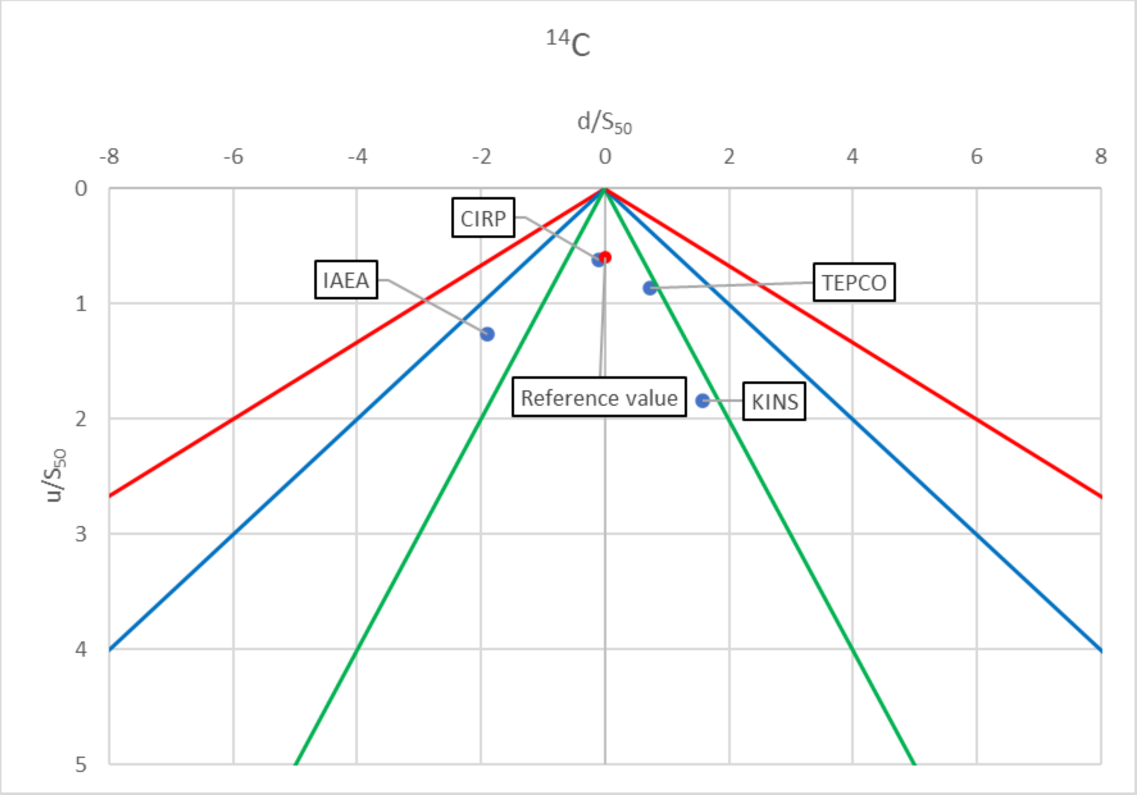

FIG. 3. PomPlot for ^{14}C results.

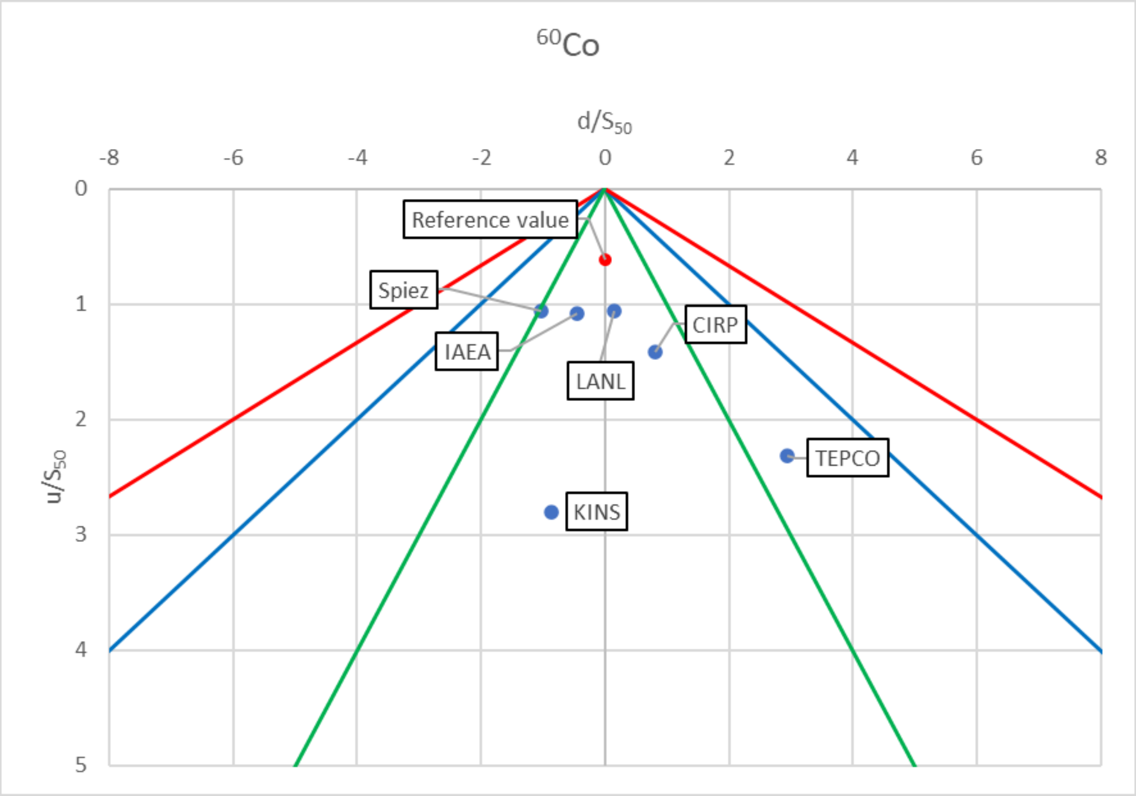

FIG. 4. PomPlot for ^{60}Co results.

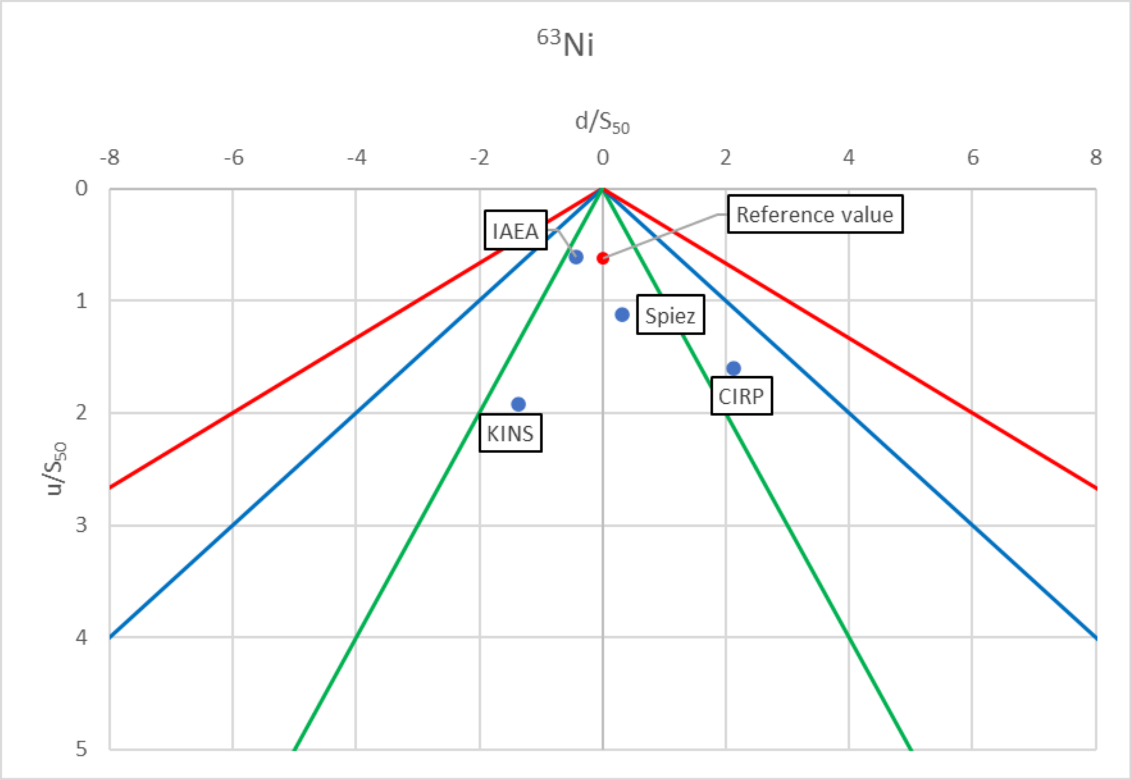

FIG. 5. PomPlot for ^{63}Ni results.

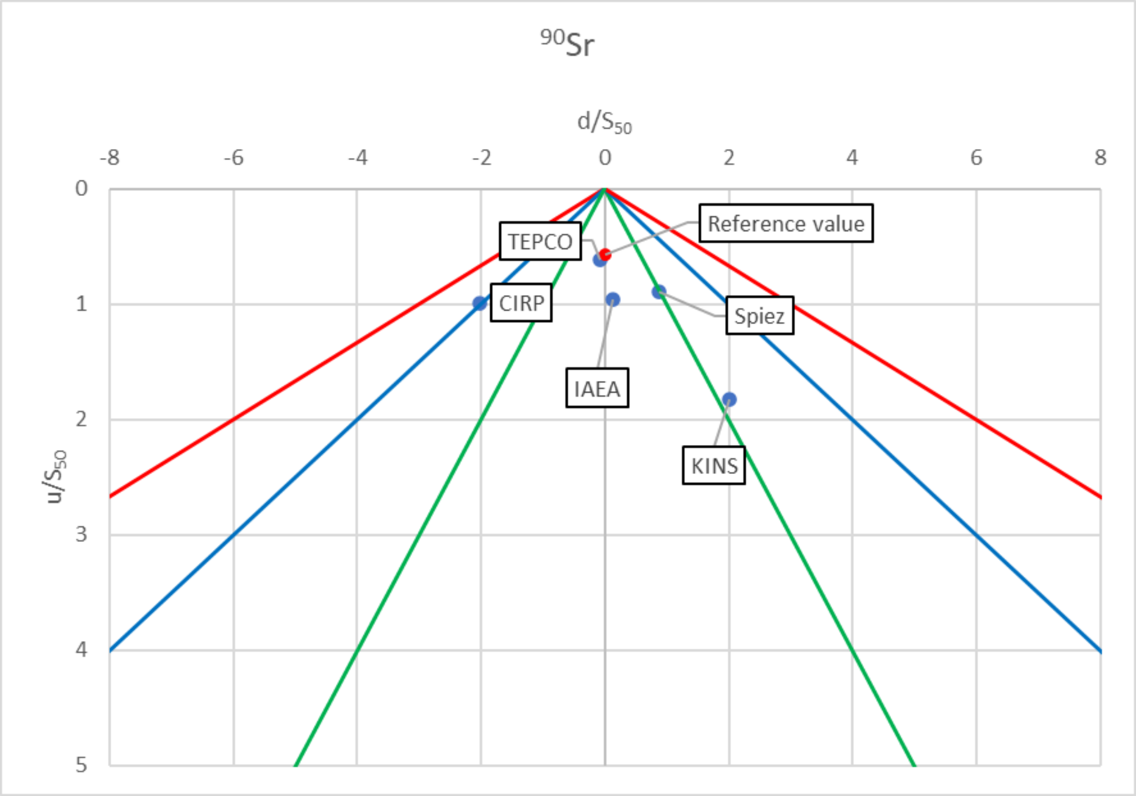

FIG. 6. PomPlot for ^{90}Sr results.

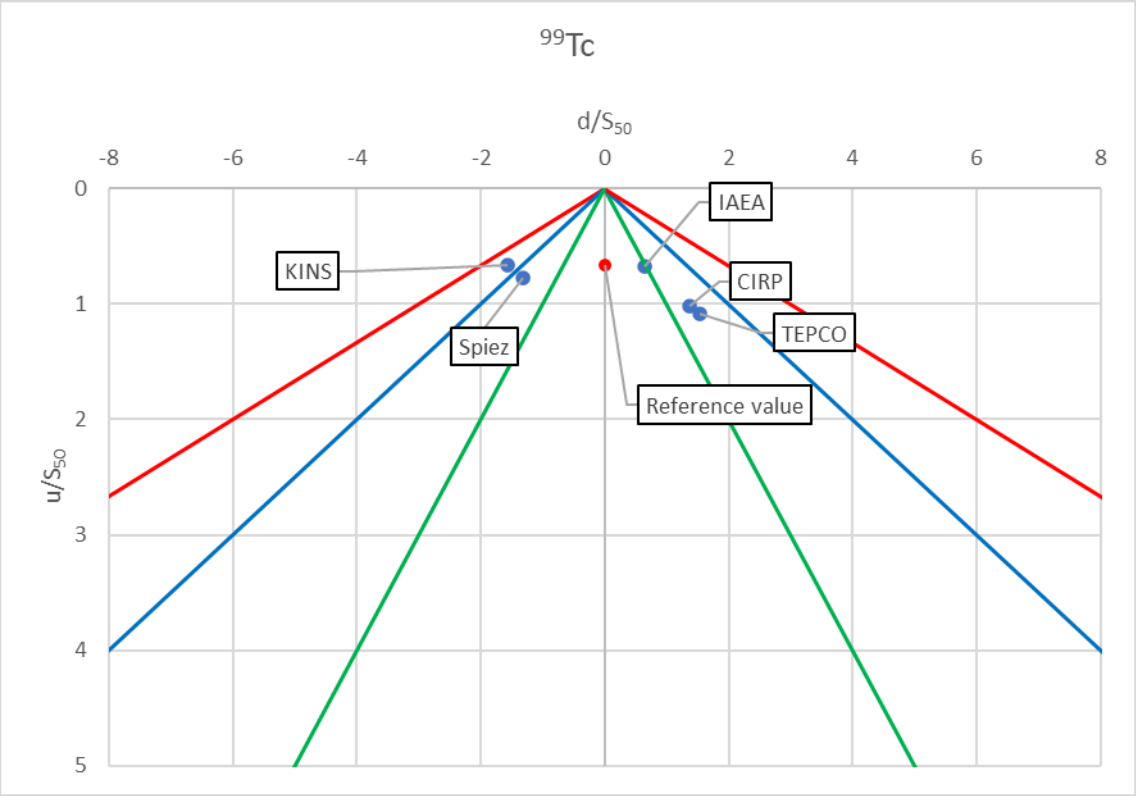

FIG. 7. PomPlot for ^{99}Tc results.

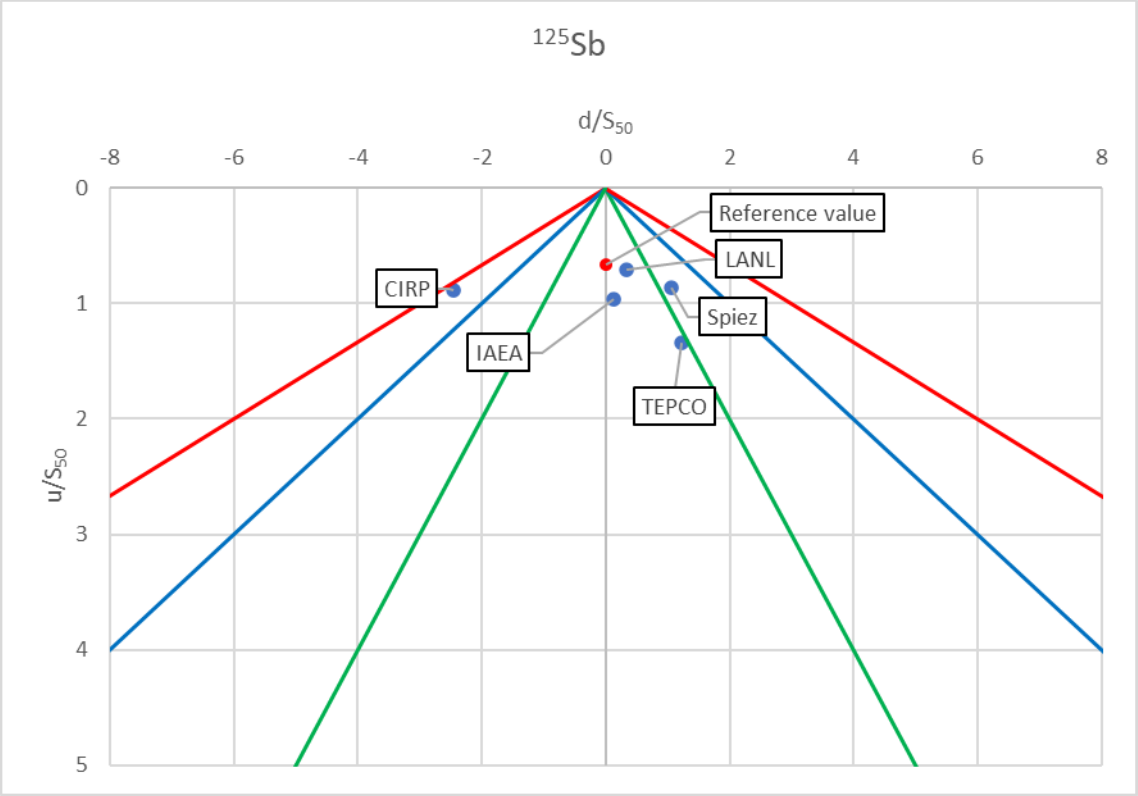

FIG. 8. PomPlot for ^{125}Sb results.

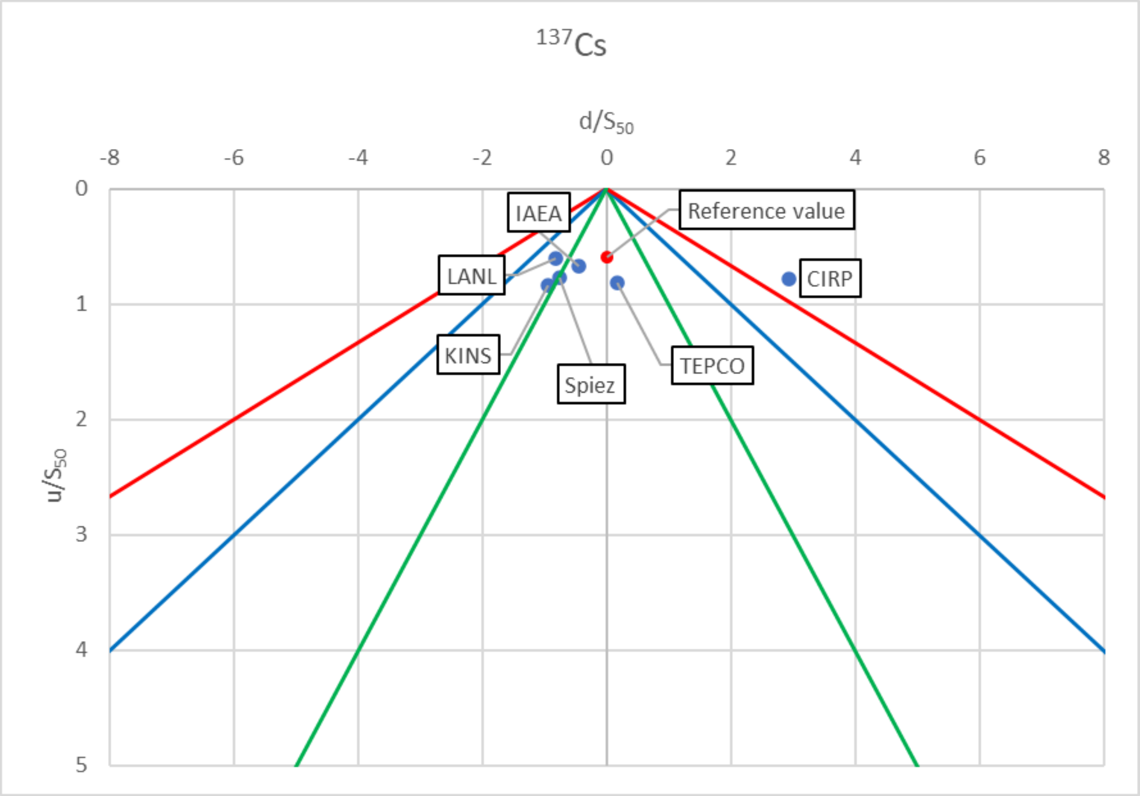

FIG. 9. PomPlot for ^{137}Cs results.

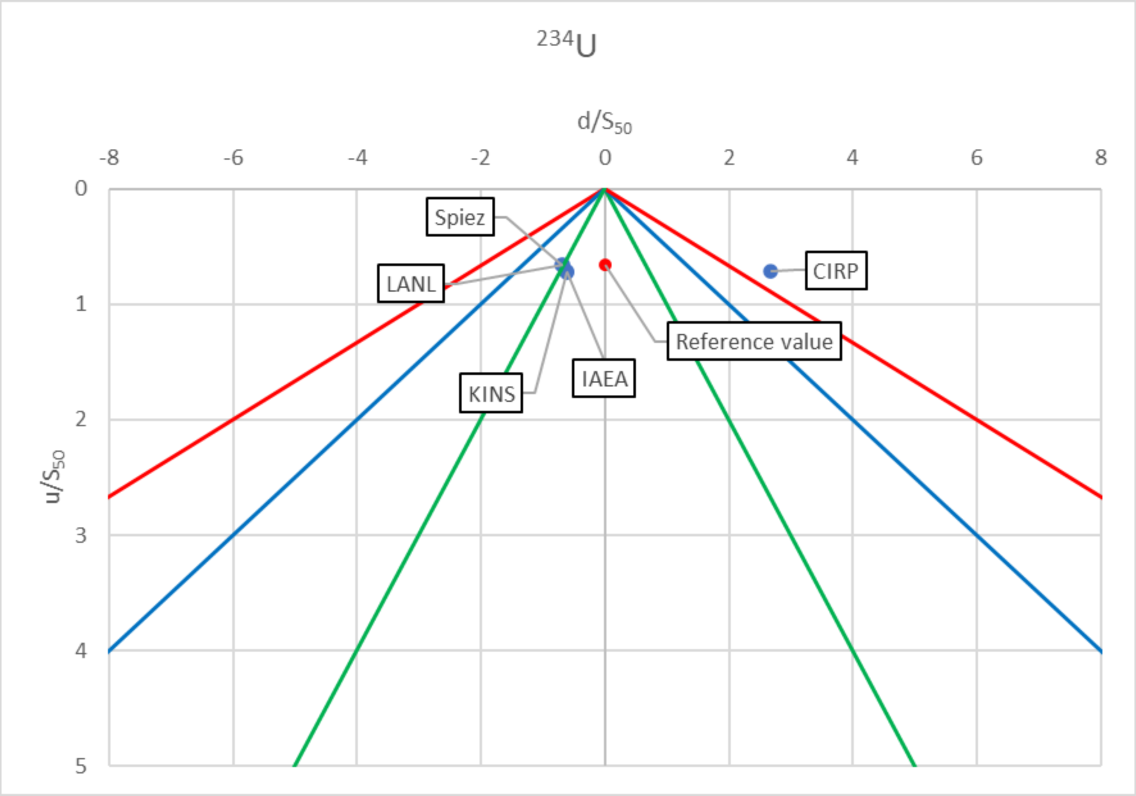

FIG. 10. PomPlot for ^{234}U results.

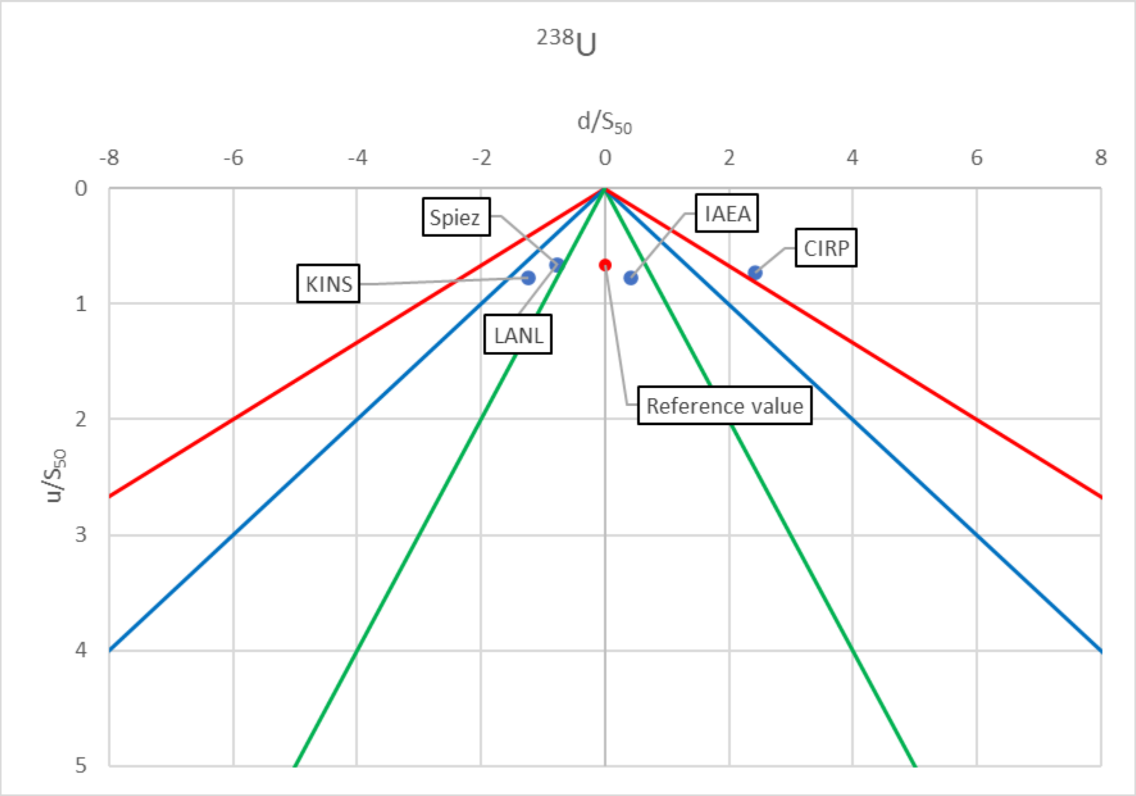

FIG. 11. PomPlot for ^{238}U results.

6.5. ADDITIONAL RESULTS REPORTED

Several participating laboratories analysed the samples for radionuclides in addition to those included in TEPCO's ALPS treated water source term.

KINS analysed for ^{94}Nb,^{133}Ba, ^{146}Pm, ^{152}Eu by gamma-ray spectrometry; ^{233}U, ^{235}U and ^{236}U by alpha spectrometry; and ^{242}Pu by ICP-MS. The results were all below the detection limits of 0.44 or less, Bq/L and 0.00000049 Bq/L for gamma, alpha and ICP-MS respectively. KINS also screened the samples for gross alpha and gross beta, reporting results of <0.69 and 3.7± 0.26 Bq/L respectively.

LANL analysed for ^{94}Nb and ^{133}Ba by gamma-ray spectrometry (< 0.016 Bq/L); ^{233}U (0.000136 ± 0.000031 Bq/L), ^{235}U (0.000032020 ± 0.000000061 Bq/L) and ^{236}U (0.000002030 ± 0.000000028 Bq/L) by multi-dynamic TIMS; ^{242}Pu (0.0000002450 ± 0.0000000018 Bq/L) by multi-ion counting (multi-static total evaporation) TIMS and ^{230}Th (0.00000117 ± 0.00000020) and ^{232}Th (0.000000333 ± 0.000000025) by multi-static ICP-MS.

SPIEZ analysed for ^{40}K, ^{58}Co, ^{59}Fe, ^{65}Zn, ^{96}Nb, ^{103}Ru, ^{110m}Ag, ^{124}Sb, ^{133}Ba, ^{141}Ce, ^{144}Pr, ^{152}Eu, ^{210}Pb and ^{228}Ac by gamma-ray spectrometry. Apart from ^{40}K (0.66 ± 0.33 Bq/L), all reported activity concentrations were below detection limits ranging from 0.012 Bq/L (^{58}Co) to 0.806 Bq/L (^{144}Pr). SPIEZ also analysed ^{235}U (0.00003200 ± 0.00000088 Bq/L), ^{236}U (0.000002000 ± 0.000000094 Bq/L), ^{244}Pu (<0.0000000010 Bq/L) and ^{245}Cm (<0.000018 Bq/L) by sf-ICP-MS.

TEPCO analysed for ^{58}Co, ^{59}Fe, ^{65}Zn ^{86}Rb, ^{91}Y, ^{95}Nb, ^{103}Ru, ^{110m}Ag, ^{115m}Cd, ^{123}Sn, ^{126}Sn, ^{124}Sb, ^{123m}Te, ^{127}Te, 129Te, ^{129m}Te, ^{136}Cs, ^{140}Ba, ^{141}Ce, ^{146}Pm, ^{148}Pm, ^{148m}Pm, ^{152}Eu, ^{153}Gd and ^{160}Tb, by gamma-ray spectrometry. All reported activity concentrations were below detection limits ranging from 0.025 Bq/L (^{110m}Ag) to 2.7 Bq/L (^{91}Y). TEPCO also analysed for ^{89}Sr using a plastic scintillation detector, reporting a detection limit of 0.069 Bq/L. TEPCO reported estimates of an upper bound for activity concentrations of ^{243}Am, ^{242}Cm and ^{243}Cm from gross alpha screening (< 0.029 Bq/L) and calculated activity concentrations of ^{103m}Rh, ^{106}Rh, ^{119m}Sn, ^{127m}Te, ^{135}Cs, ^{137m}Ba, ^{144}Pr, ^{144m}Pr and ^{242m}Am either by assuming equilibrium with appropriate measured radionuclides or using the inventory ration approach as described for ^{241}Pu, ^{147}Pm and ^{151}Sm in section 4.13.

7. CONCLUSION

This ILC has been implemented to check the capability of TEPCO to undertake analyses relevant to source monitoring with respect to discharges of ALPS treated water to the required standard and to report high quality and comparable results. Samples collected from the K4-C tank group at FDNPS in June 2024 have been analysed for radionuclide activity concentrations by TEPCO, the IAEA laboratories and selected third-party laboratories from the IAEA ALMERA network.

7.1. QUANTITATIVE INTERCOMPARISON OF REPORTED ACTIVITY CONCENTRATIONS

The main focus of the ILC was on the 31 radionuclides included in TEPCO's ALPS treated water source term [3]. For 24 of these radionuclides (^{3}H, ^{14}C, ^{54}Mn, ^{55}Fe, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{106}Ru, ^{125}Sb, ^{129}I, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu, ^{155}Eu, ^{234}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am), results were reported by at least two participating laboratories in addition to TEPCO. Results for ^{90}Y and ^{125m}Te, which are also included in TEPCO's source term, can be derived from the results for ^{90}Sr and ^{125}Sb, respectively, assuming equilibrium. The exceptions were ^{79}Se, ^{147}Pm and ^{151}Sm for which only TEPCO reported results and ^{113m}Cd and ^{244}Cm for which only TEPCO and LANL and SPIEZ reported results.

Activity concentrations above detection limits were reported by at least two laboratories for 14 radionuclides: ^{3}H, ^{14}C, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{125}Sb, ^{129}I, ^{137}Cs, ^{234}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu. The reported results for these radionuclides have been subjected to strict statistical tests as described in section 5. Out of a total of 68 reported results that were assessed using these statistical tests, over 92% (63), were evaluated as agreeing with a high level of confidence (99.7%). This high degree of comparability of the results provides evidence of the high level of competence of all participating laboratories.

The exceptions were the following results where zeta scores (ζ) indicated that values were significantly different from the respective reference value (Section 6.3):

- ζ scores of 3.74 for the activity concentration of ^{137}Cs reported by CIRP.
- ζ scores of 3.72 for the activity concentration of ^{234}U reported by CIRP.
- ζ scores of 3.31 for the activity concentration of ^{238}U reported by CIRP.

Two further zeta tests for directly compared results (for radionuclides for which measured activity concentrations above detection limits were reported by two or three laboratories) also indicated statistically significant differences for the activity concentrations of ^{237}Np and ^{239}Pu reported by LANL and SPIEZ (Section 6.3).

This small number of inconsistencies was to be expected having applied these strict statistical tests. Even in high performing laboratories, run by knowledgeable and experienced staff, anomalous results may sometimes be obtained. Furthermore, ALPS treated water constitutes a complex, highly processed sample matrix that has been subjected to a series of treatments at FDNPS to lower the radionuclide content, including co-precipitation, physical filtration and adsorption. It has been shown to be homogenous at a macro-level (within the tank group at FDNPS) [6] but at a micro-level this is more uncertain and inter-sample heterogeneity was reported by some participating laboratories performing repeat analyses. Additionally, radionuclides, other than ^{3}H, were present in the samples at relatively low levels and the participating laboratories were required to use a range of often complex analytical techniques, both factors potentially resulting in generally higher uncertainties.

For radionuclides for which results could be intercompared quantitatively with a reference value (^{3}H, ^{14}C, ^{60}Co, ^{90}Sr, ^{99}Tc, ^{125}Sb, ^{137}Cs), TEPCO's results were in all cases in agreement with a 99.7 % level of confidence (in fact, ζ scores were between -2 and 2 in all cases). A PomPlot showing this data is presented in Figure 12. As this chart demonstrates, the standard uncertainties reported by TEPCO were also judged to be appropriate and informative.

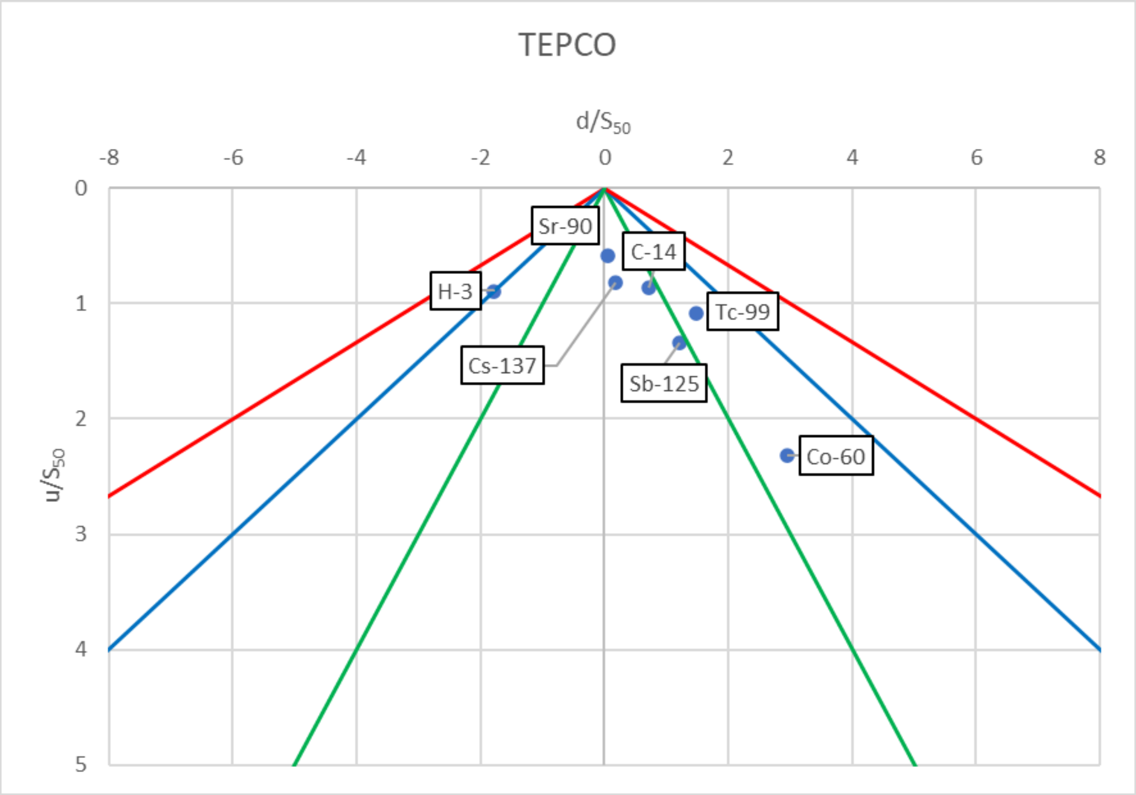

FIG. 12. PomPlot for TEPCO's results.

For ^{129}I, measured activity concentrations were reported by IAEA and KINS as well as TEPCO. The zeta tests for these directly compared results also indicated that TEPCO's result was in good agreement with the data reported by the other laboratories (in fact, both ζ scores comparing TEPCO to the other two laboratories were between -2 and 2).

7.2. ASSESSMENT OF REPORTED DETECTION LIMITS

For other radionuclides, the detection levels reported by participating laboratories were compared qualitatively. TEPCO's detection limits were in most cases comparable to those reported by the other participating laboratories. Exceptions were ^{55}Fe and ^{63}Ni and the actinides (^{234}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu and ^{241}Am) for which the detection limits were generally significantly higher than those reported by the other participating laboratories.

A comparison of all the detection limits reported by TEPCO with respective regulatory limits for all radionuclides in its source term showed that for each radionuclide analysed the reported detection limit was less than 1% of the respective regulatory limit in all cases. This demonstrates that the methods selected are appropriate and fit for purpose.

TEPCO assesses actinides included in its source term using a screening approach based on gross alpha counting. The chemical recovery of this method has recently been undergone an additional verification to ensure that it is robust for additional targeted radionuclides. The gross alpha result – in this case a detection limit of 0.029 Bq/L – is taken as an upper bound of the activity concentration of each individual actinide included in the source term. Contaminated and ALPS treated water samples collected at FDNPS have been characterised for alpha-emitting radionuclides using radionuclide specific analytical methods in the past. The resulting activity concentrations were <1% of the regulatory limit in all cases. As the screening approach is sufficiently conservative, is implemented to a high quality, and a procedure is in place describing steps to be taken for effective follow up if a pre-stated action limit is exceeded, this approach is considered to be fit for purpose.

7.3. ADDITIONAL RADIONUCLIDES

Some participating ALMERA laboratories analysed and reported results for radionuclides in addition to those included in TEPCO's source term. The results from this broad collective analytical capacity of the participating laboratories identified the presence of no additional radionuclides above detection limits, with the exception of very low levels of some isotopes of ^{230}Th, ^{232}Th, ^{233}U, ^{235}U, ^{236}U, and ^{242}Pu, that were detected by LANL using ICP-MS and TIMS, and ^{235}U, ^{236}U, ^{244}Pu and ^{245}Cm that were detected by SPIEZ using sf-ICP-MS.

7.4. KEY FINDINGS

The accurate results obtained in this ILC demonstrate a high degree of proficiency on the part of TEPCO. This should provide confidence in TEPCO's capability for conducting reliable and high-quality source monitoring related to the discharge of ALPS treated water.

TEPCO's sample collection procedures follow the appropriate methodological standards required to obtain representative samples. This is demonstrated by the generally high degree of comparability in the results reported by all participating laboratories.

The detection limits reported by TEPCO indicate that analytical methods selected were appropriate and fit for purpose. The reported detection limits were less than 1% of the respective regulatory limits for all radionuclides included in TEPCO's source term.

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APPENDIX I: INSTRUCTIONS SENT TO PARTICIPATING LABORATORIES

Participating laboratories were encouraged, subject to the analytical methods at hand, to analyse the K4-C samples for as many radionuclides as possible in TEPCO's ALPS treated water source term – those requiring measurement and assessment as part of its source monitoring plan.

Participating laboratories requested to submit a single measurement result for each radionuclide analysed, decay corrected to the sampling date (21 June 2024). A measurement result was assumed to be comprised of an activity concentration and associated uncertainty, or the detection limit, as appropriate, all expressed in Bq/L.

They were asked to report the following additional information for each radionuclide for which results were to be reported, in addition to the activity concentration and uncertainty:

(a) A short description of the analytical method applied for the sample analysis.
(b) The type of calibration applied.
(c) The software used for the counting/spectrometric systems.
(d) The nuclear data used (e.g., half-life, energy, intensity of gamma emission) in the determination of the results. The use of DDEP (Decay Data Evaluation Project, http://www.lnhb.fr/nuclear-data/) data was encouraged, subject to availability.
(e) The decision threshold.
(f) The uncertainty budget with components including the following, as relevant:
    - Statistical counting uncertainty, including any background subtraction.
    - Uncertainty of the detector efficiency, including the uncertainty of the calibration source, as applicable.
    - Uncertainty of chemical yields, as applicable.
    - Uncertainty of mass measurements.
    - Uncertainty of corrections applied, for instance for true coincidence summing or for decay correction.
    - Measurement repeatability uncertainty.

For analyses comprising repetitions, the laboratories were requested to inform IAEA on how the measurement result reported was determined (e.g., an average of the values from each repetition or a single value (from a single selected repetition). For values derived from an average, information was requested on how the uncertainty was estimated (e.g., weighted average, maximum value etc.).

The IAEA recommended samples to be prepared gravimetrically to minimise uncertainties. The value of the density of the sample used to convert from gravimetric to volumetric activity concentrations (i.e., from Bq/kg to Bq/L) was also requested to be reported, as necessary.

APPENDIX II: CHARTS OF ACTIVITY CONCENTRATIONS

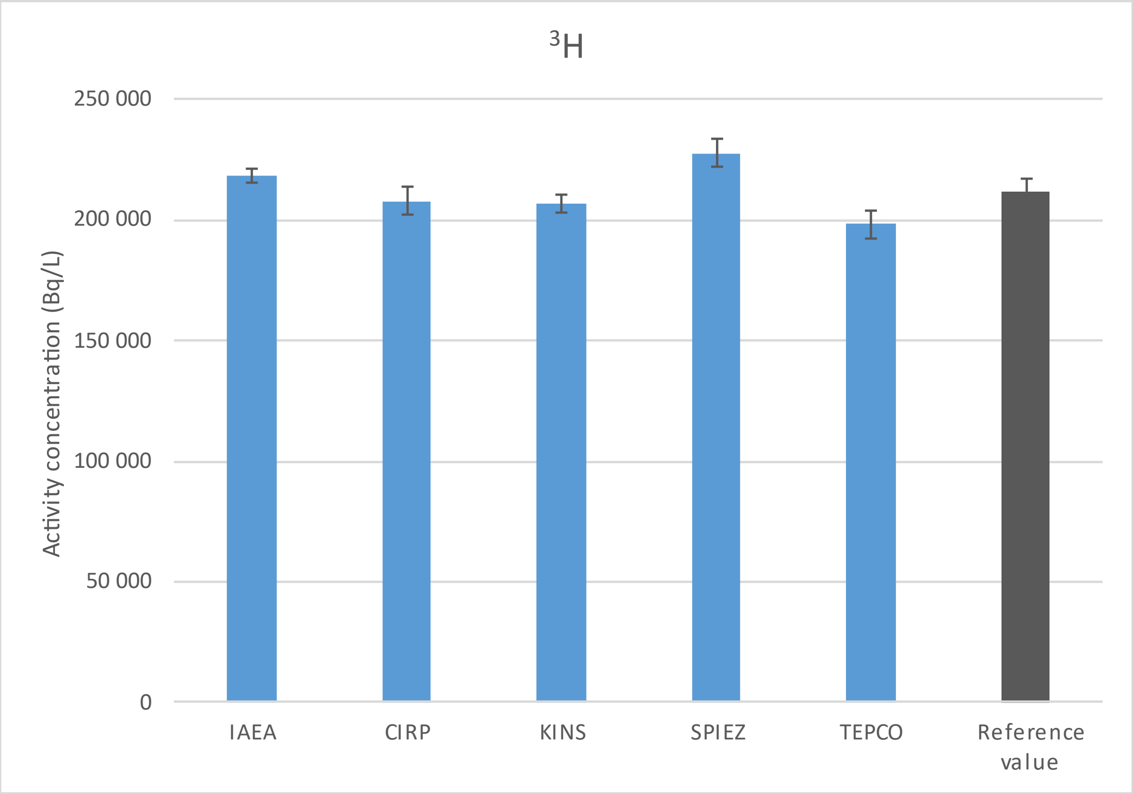

FIG. 13. Activity concentrations and detection limits for ^{3}H for the samples from the K4-C tank group.

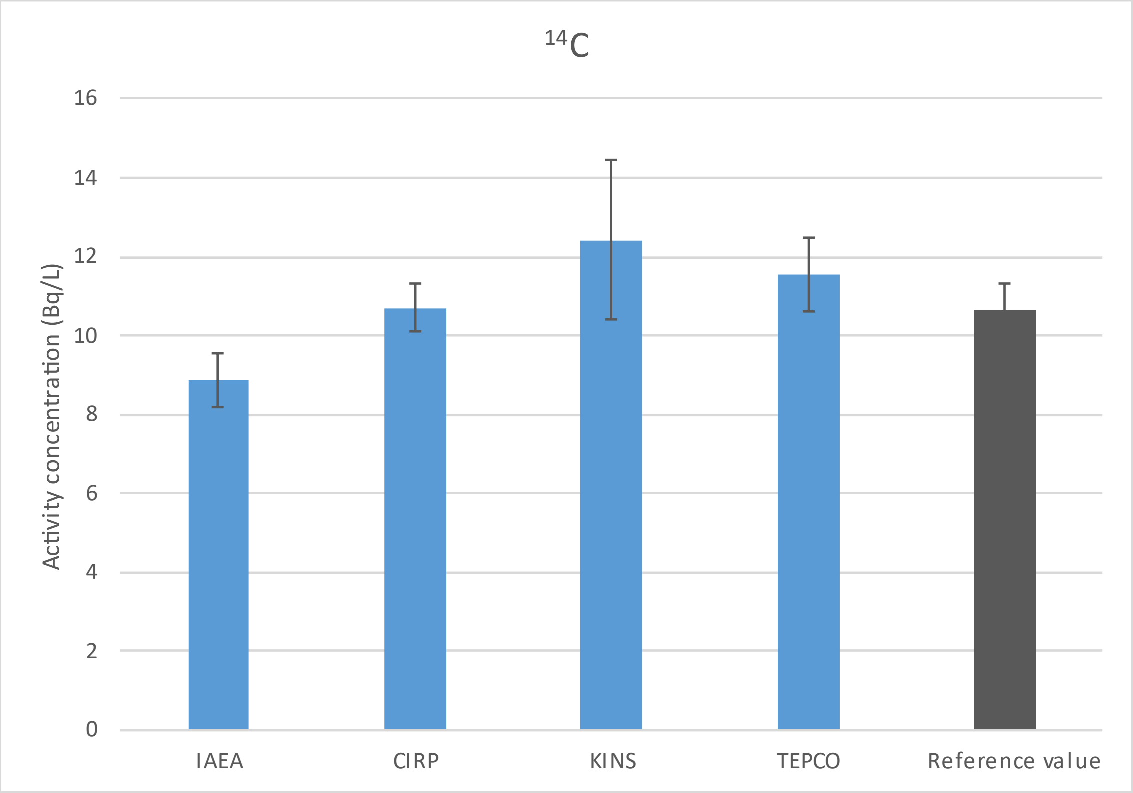

FIG. 14. Activity concentrations and detection limits for ^{14}C for the samples from the K4-C tank group.

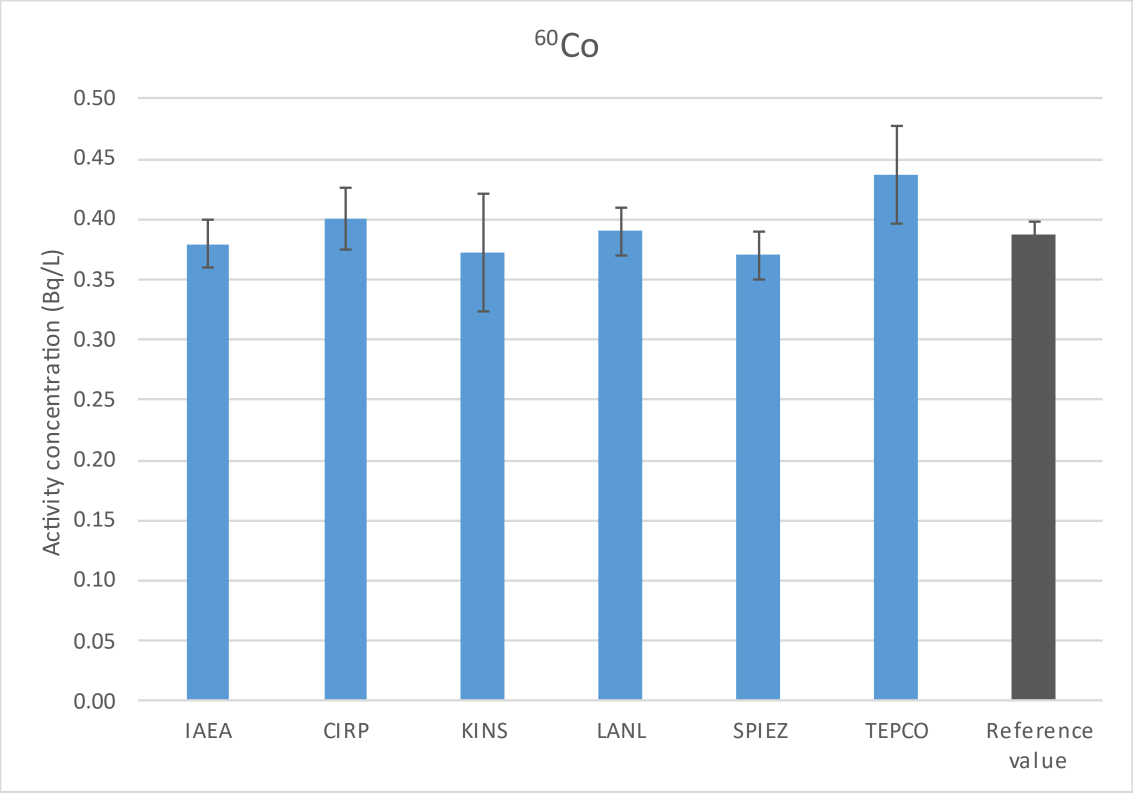

FIG. 15. *Activity concentrations and detection limits for ^{60}Co for the samples from the K4-C tank group.*

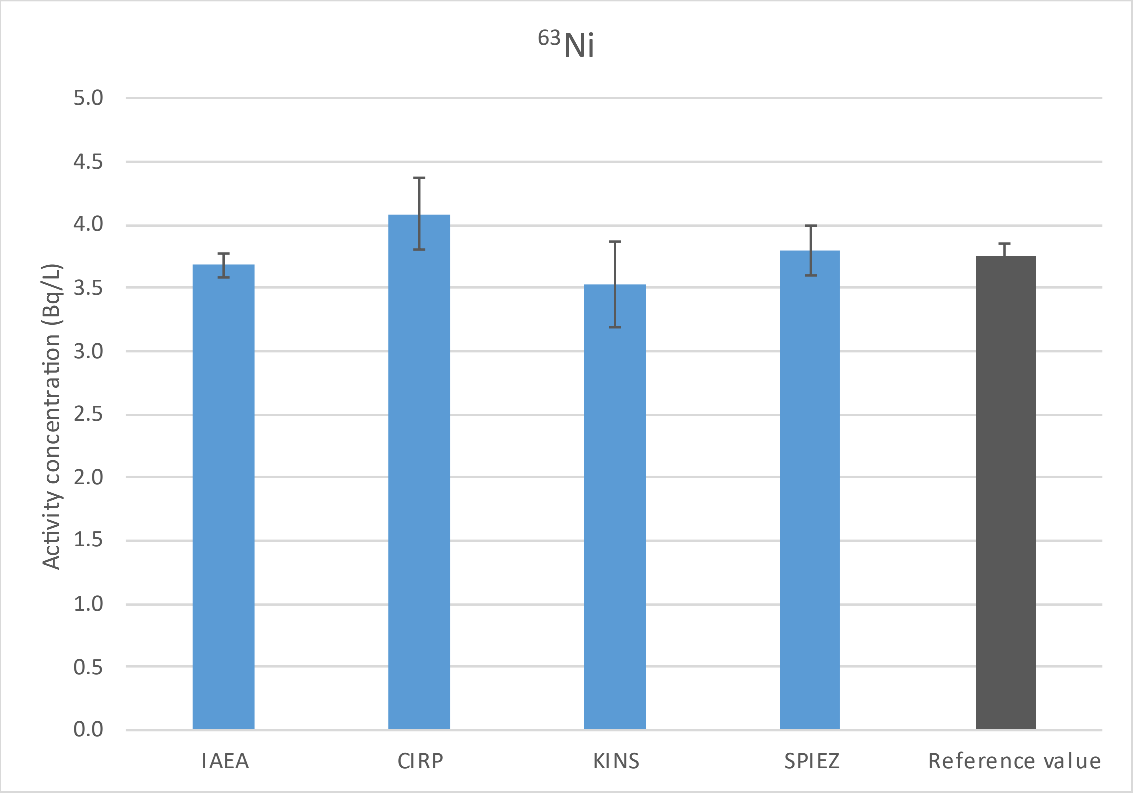

FIG. 16. *Activity concentrations and detection limits for ^{63}Ni for the samples from the K4-C tank group.*

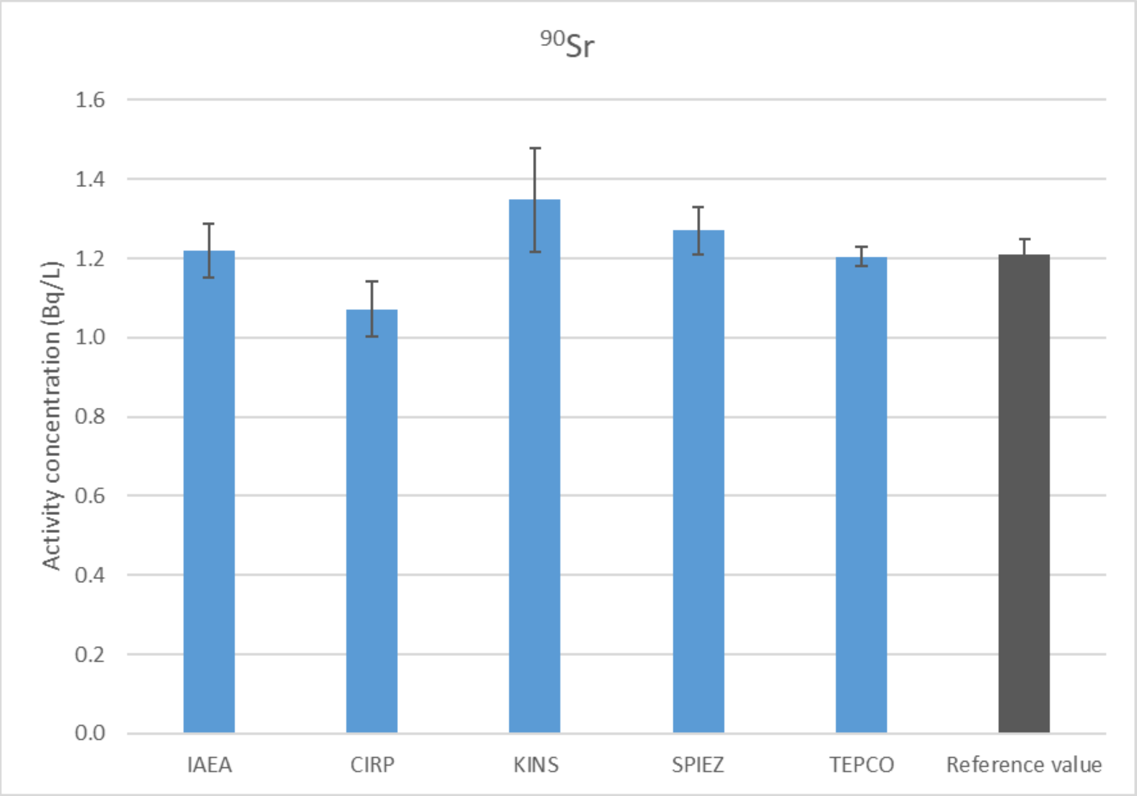

FIG. 17. Activity concentrations and detection limits for ^{90}Sr for the samples from the K4-C tank group.

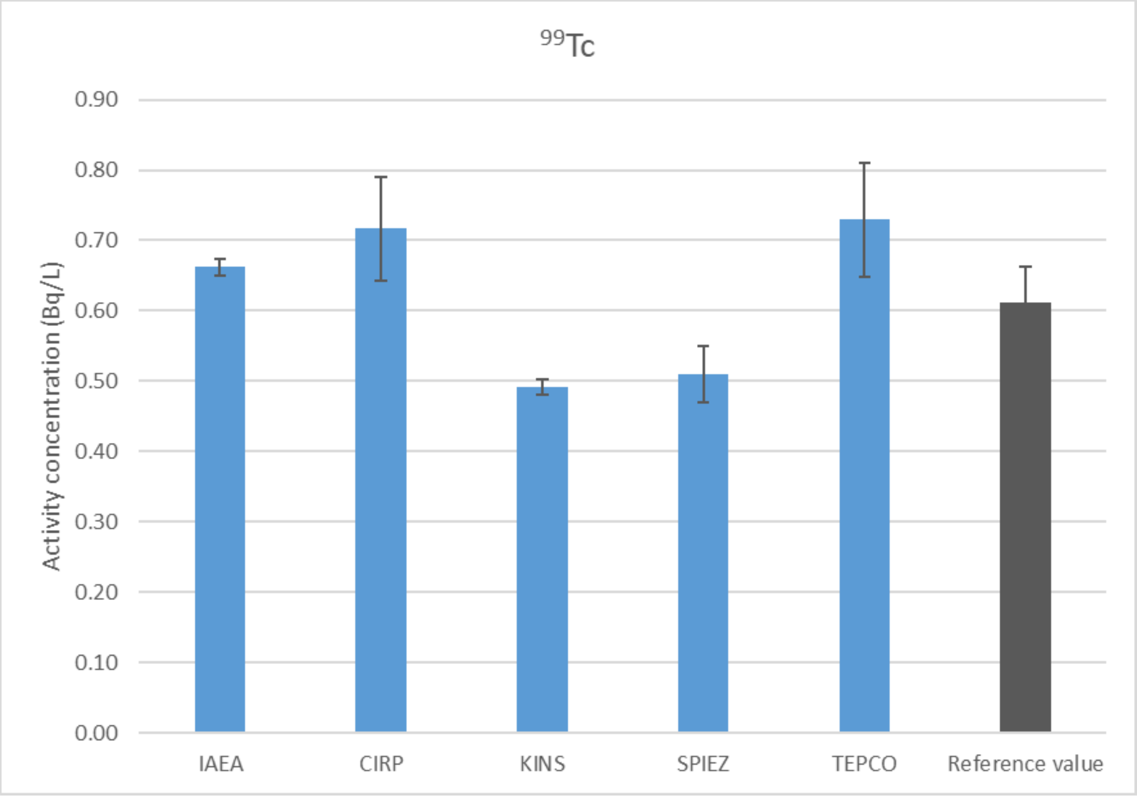

FIG. 18. Activity concentrations and detection limits for ^{99}Tc for the samples from the K4-C tank group.

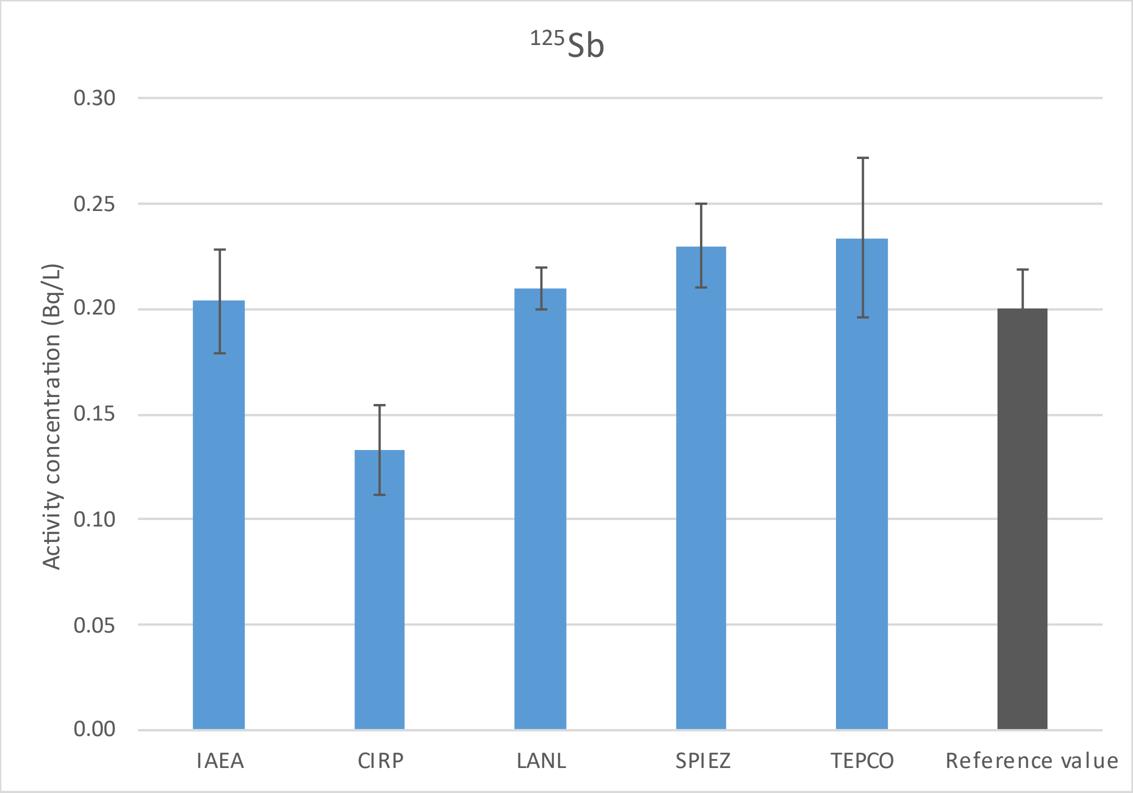

FIG. 19. Detection limits for ^{125}Sb for the samples from the K4-C tank group.

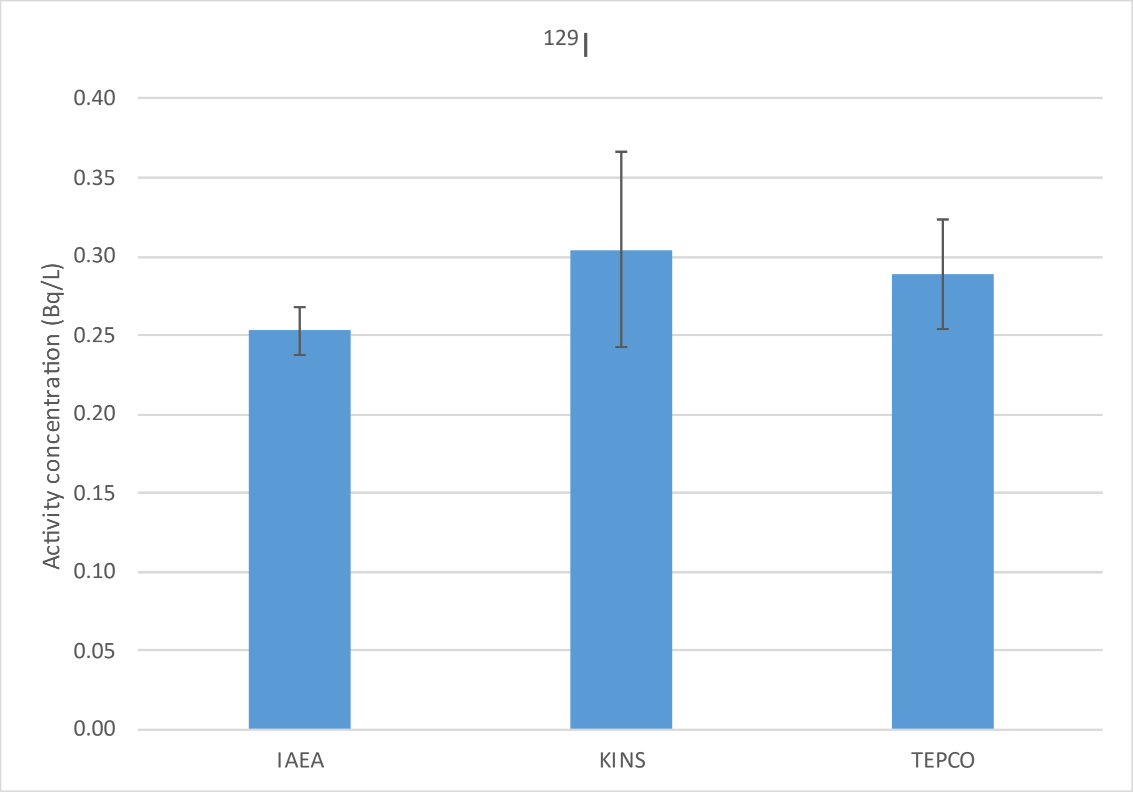

FIG. 20. Activity concentrations and detection limits for ^{129}I for the samples from the K4-C tank group.

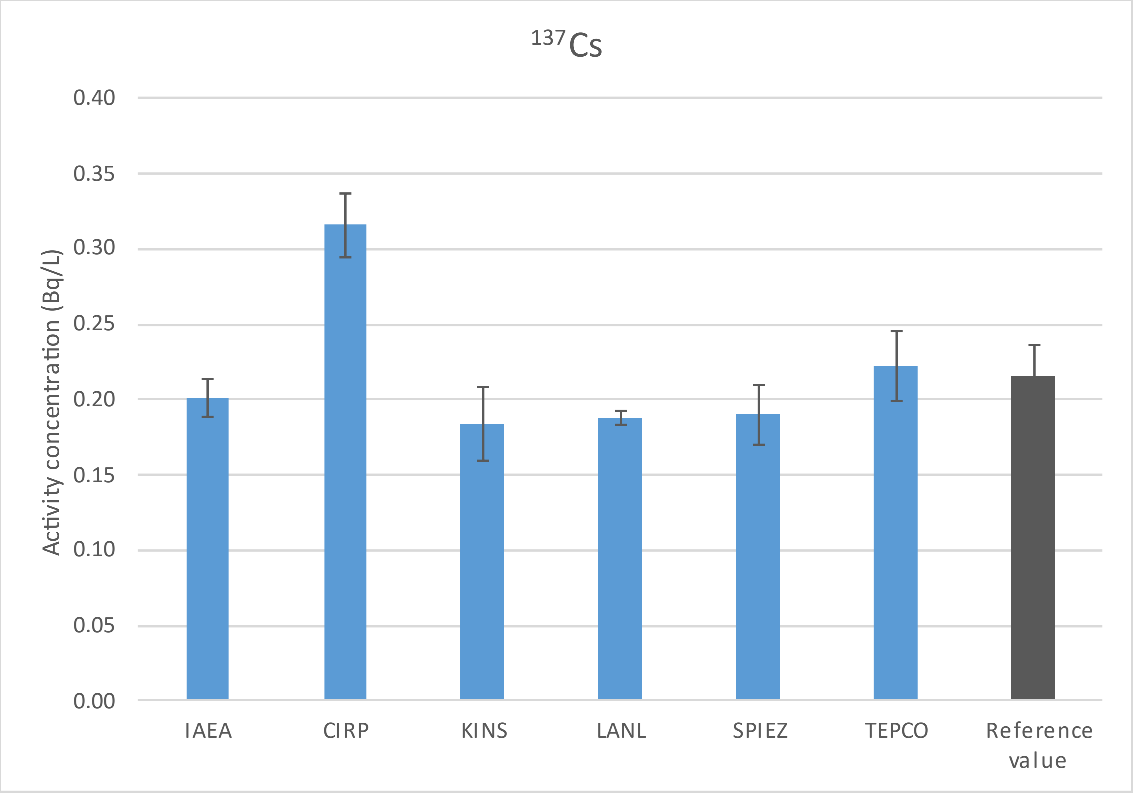

FIG. 21. *Activity concentrations and detection limits for* ^{137}*Cs for the samples from the K4-C tank group.*

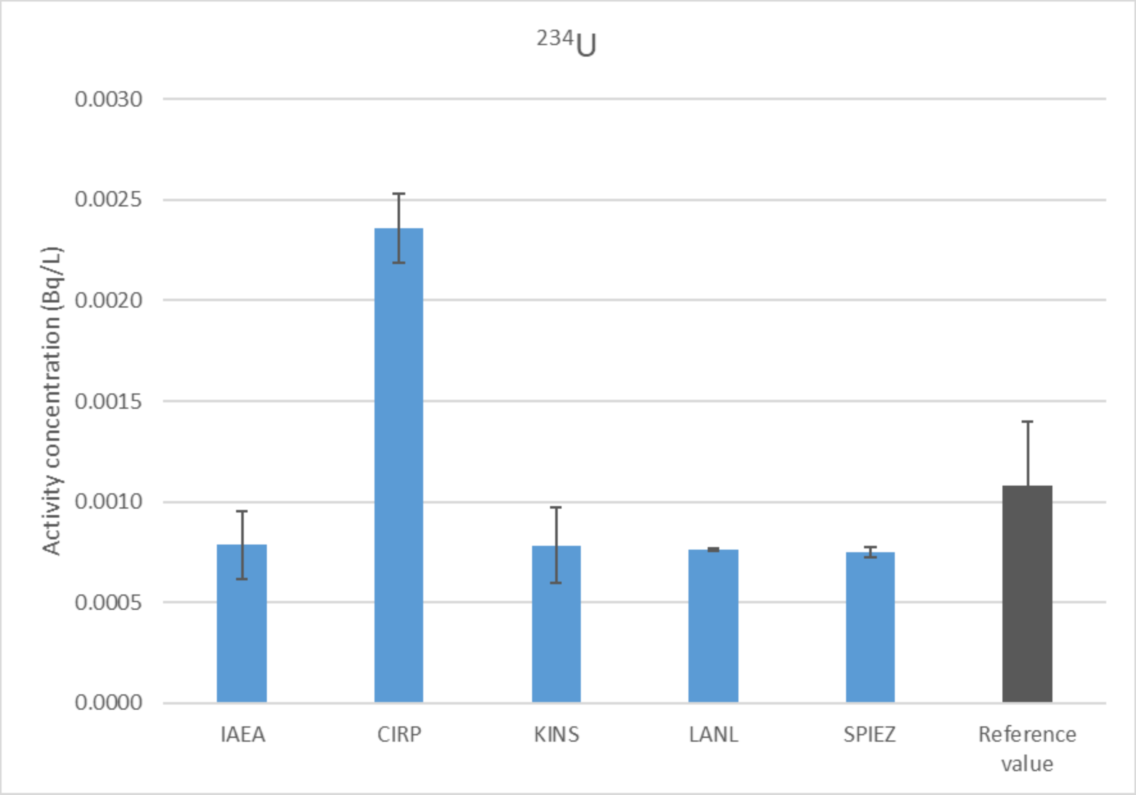

FIG. 22. *Activity concentrations and detection limits for* ^{234}*U for the samples from the K4-C tank group.*

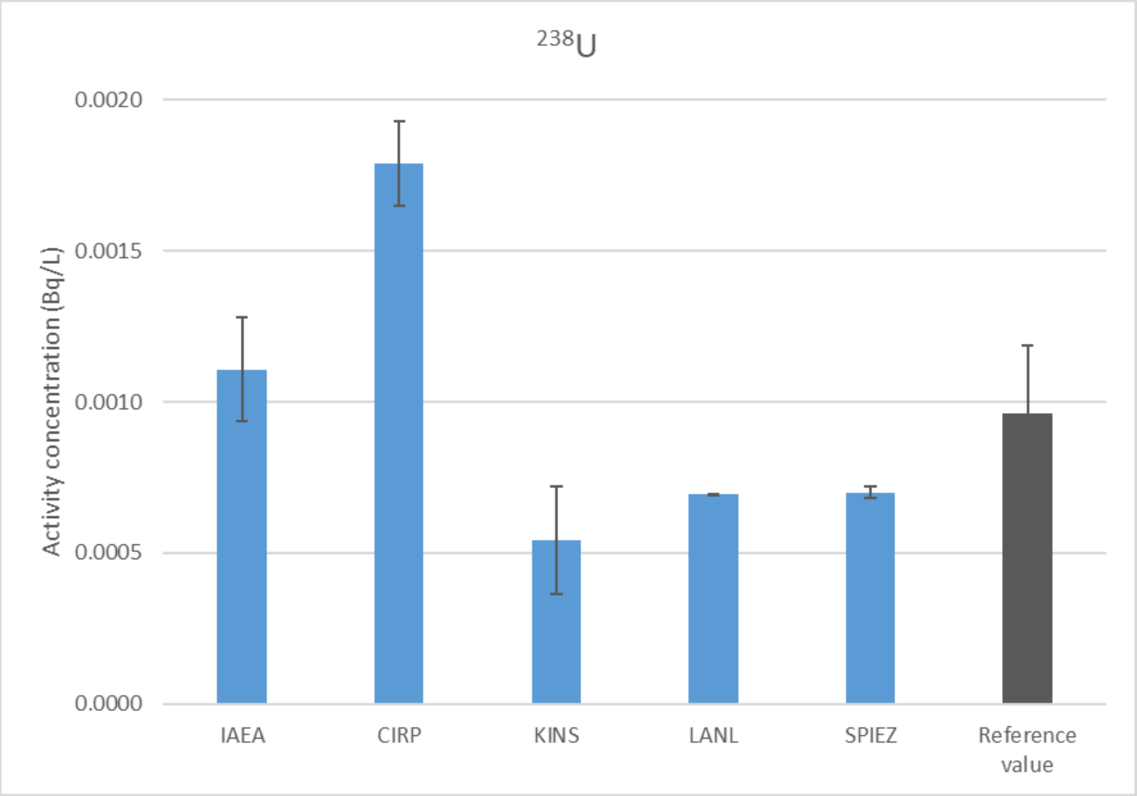

FIG. 23. Activity concentrations and detection limits for ^{238}U for the samples from the K4-C tank group.

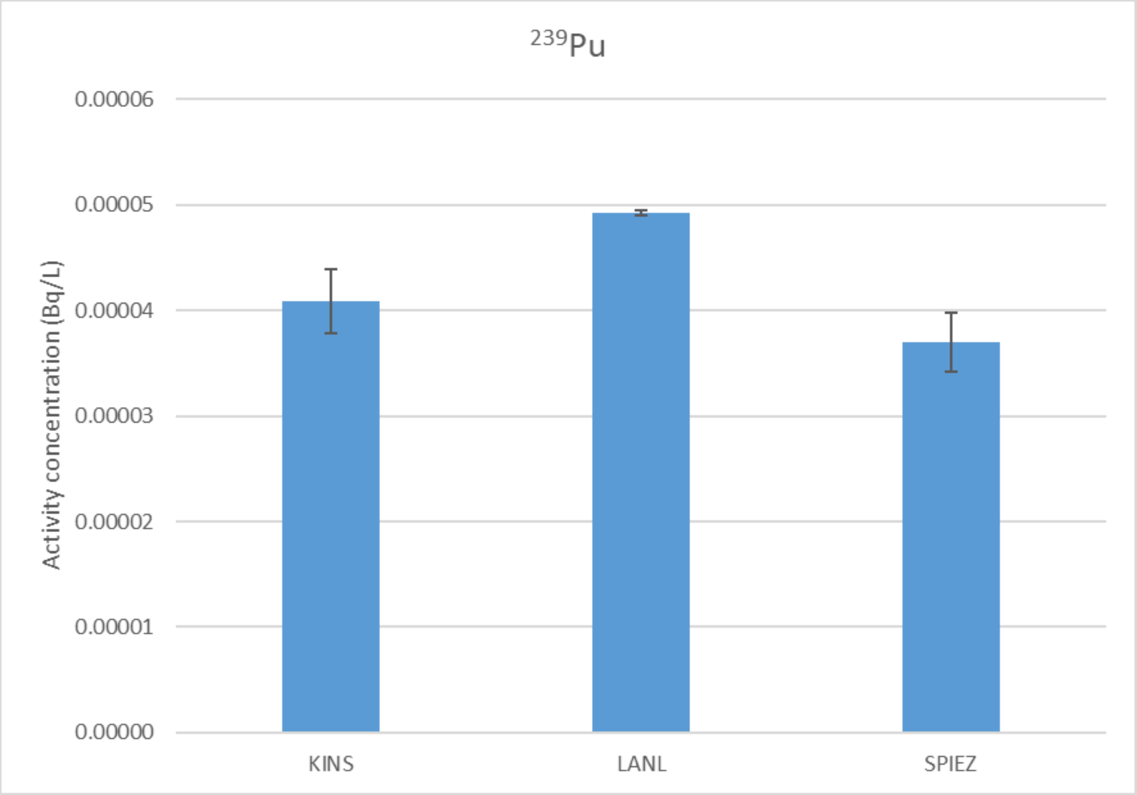

FIG. 24. Activity concentrations and detection limits for ^{239}Pu for the samples from the K4-C tank group.

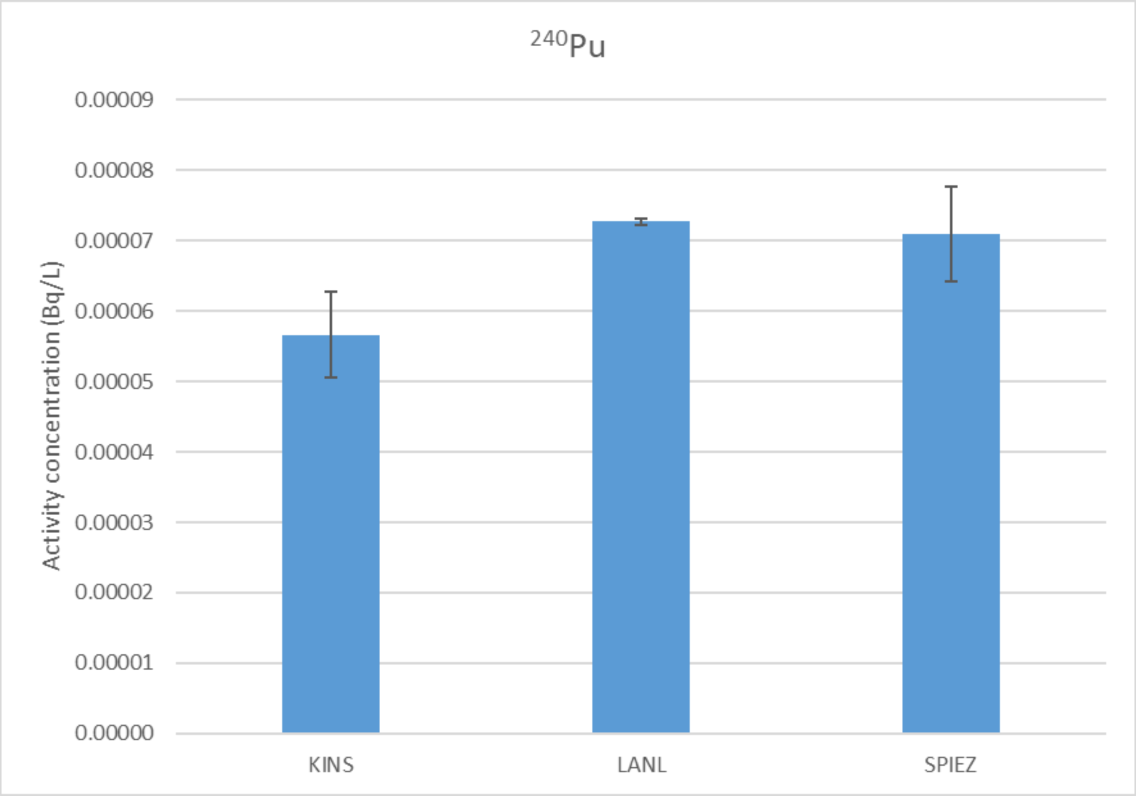

FIG. 25. Activity concentrations and detection limits for ^{240}Pu for the samples from the K4-C tank group.

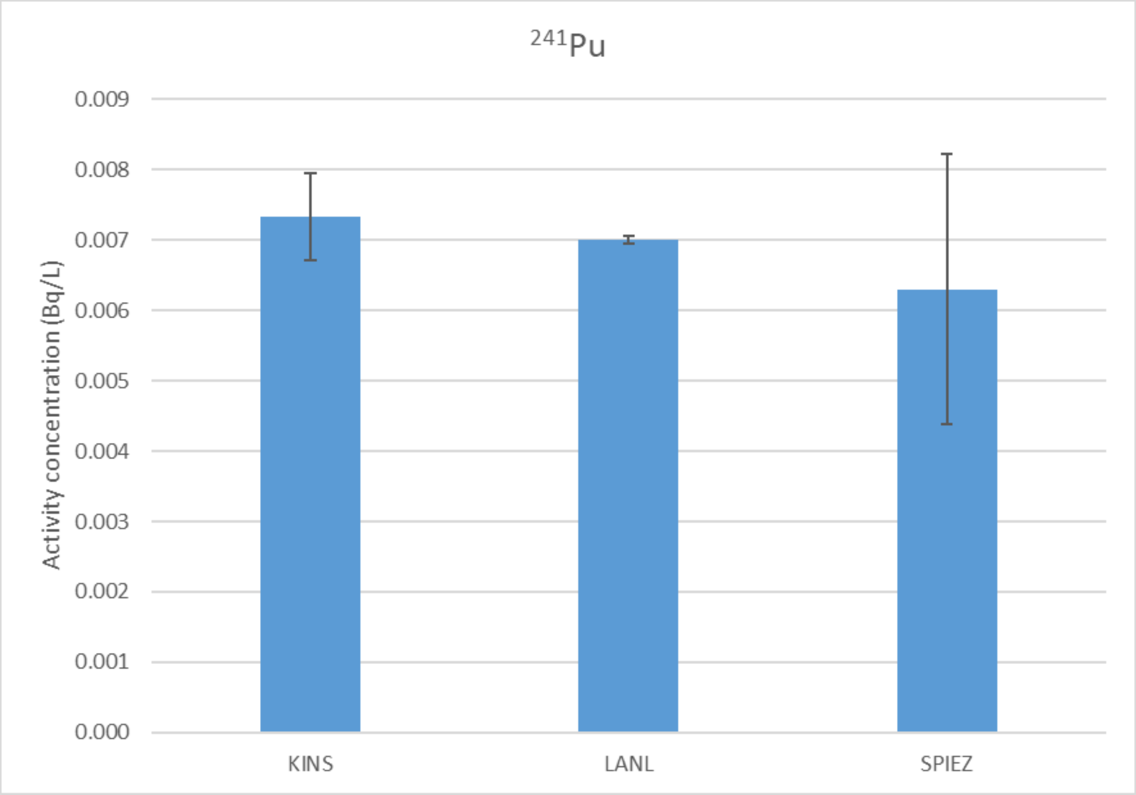

FIG. 26. Activity concentrations and detection limits for ^{241}Pu for the samples from the K4-C tank group.

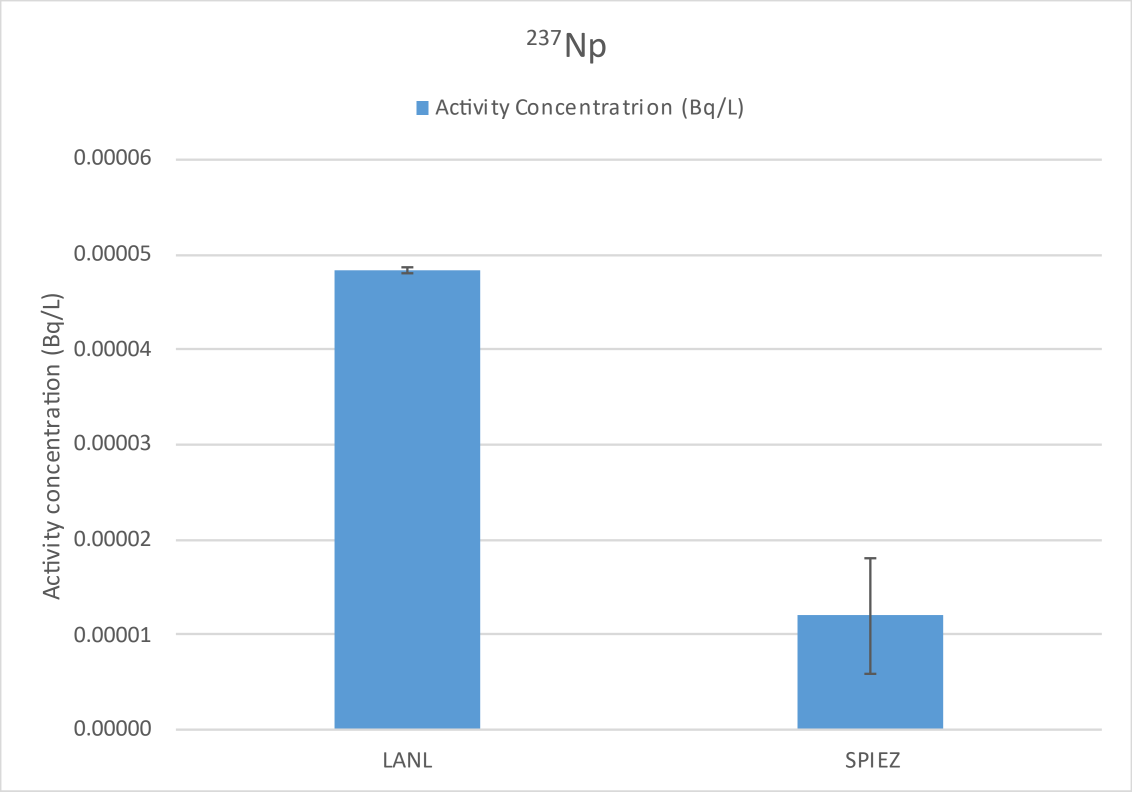

FIG. 27. Activity concentrations and detection limits for ^{237}*Np for the samples from the K4-C tank group.*

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