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

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

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<center>**EXECUTIVE SUMMARY**</center>

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 international 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 approximately 30 years.

Consistent with relevant IAEA international 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 independent sampling, data corroboration, and analysis. Further, the IAEA's independent sampling, data corroboration, and analysis 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, data corroboration, and analysis 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 first ILC under the IAEA's ALPS safety review, the ALPS treated water samples were taken in March 2022 from the K4-B tank group at FDNPS. The water contained in the K4-B tank group is expected to be the first batch of ALPS treated water that will be discharged, only once TEPCO receives all regulatory approvals from NRA. The focus of the analysis efforts for this ILC were on the radionuclides from the source term (Table 3) 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. Participating laboratories were also encouraged to analyse for additional radionuclides beyond the source term.

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 195 member laboratories globally that is coordinated jointly by RML and TERC. It 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:

- Spiez Laboratory (LS – Labor Spiez), Switzerland
- Institut de Radioprotection et de Sûreté Nucléaire (IRSN), France
- Los Alamos National Laboratory (LANL), United States of America
- Korea Institute of Nuclear Safety (KINS), Republic of Korea

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 demonstrated a high level of accuracy in their measurements and technical competence.
- 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.
- Neither the IAEA, nor the participating third-party laboratories, detected any additional radionuclides (i.e., radionuclides beyond what is included in the source term) at significant levels.

The IAEA notes that these findings provide confidence in TEPCO's capability for undertaking accurate and precise measurements 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.

Additional ILCs will be conducted in the future for ALPS treated water, as well as environmental samples, and occupational radiation protection[1]. The results from the first ILC for environmental samples, which were taken in November 2022, will be available later in 2023; these first samples will serve as a baseline for future environmental monitoring. Future ILCs, conducted after water discharges start in 2023, will allow for an assessment of any changes in the levels of relevant radionuclides in the marine environment, relative to the baseline. Furthermore, the corroboration of environmental monitoring complements a separate project – NA3/38 Marine Monitoring: Confidence Building and Data Quality Assurance – addressing the quality of data from marine monitoring undertaken in Japan following the accident at FDNPS. Through project NA3/38, which has been implemented since 2014, the IAEA is assisting the Government of Japan in ensuring that sea area monitoring carried out under the regularly updated Comprehensive Radiation Monitoring Plan is comprehensive, credible and transparent and is helping to build confidence of the stakeholders in the accuracy and quality of the marine monitoring data. Within project NA3/38, the IAEA has organized a series of ILCs and PTs to test the sampling and analytical performance of the Japanese laboratories for the analysis of radionuclides in seawater, sediment, and fisheries samples. Data from this project are available online at:

https://www.iaea.org/about/organizational-structure/department-of-nuclear-sciences-and-applications/division-of-iaea-marine-environment-laboratories/marine-monitoring-confidence-building-and-data-quality-assurance.

The results from the first ILC for occupational radiation protection will be available later in 2023. This first ILC will focus on external dosimetry, whereas future ILCs for occupational radiation protection will focus on internal dosimetry. ILCs for occupational radiation protection will be conducted between the IAEA's Radiation Safety Technical Services Laboratory and the individual monitoring service used by TEPCO for FDNPS workers.

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[1] The IAEA is currently in discussions with other laboratories to participate in future ILCs.

CONTENTS

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 IAEA corroboration of source and environmental monitoring related to discharges of ALPS treated water from FDNPS is comprised of three distinct elements (see also Figure 1):

1. Review of sampling and analytical methods for source and environmental monitoring related to ALPS treated water at FDNPS used by TEPCO and relevant Japanese authorities.
2. Corroboration of source monitoring undertaken by TEPCO, including a comprehensive radiological characterization of ALPS treated water samples.
3. Corroboration of environmental monitoring undertaken by TEPCO and relevant Japanese authorities.
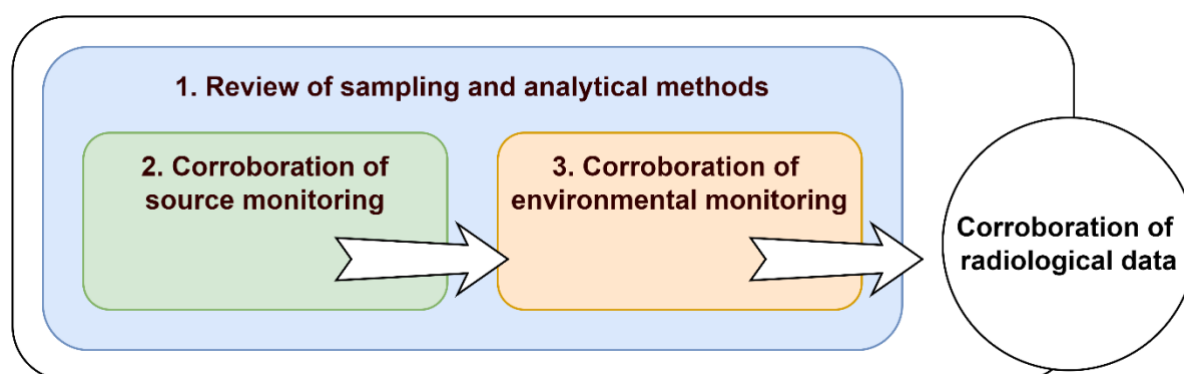
FIG. 1: A schematic overview of the elements of the corroboration being undertaken by the IAEA laboratories and the links between these elements.

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 first ILC to corroborate source monitoring.

According to its design stage radiological environmental impact assessment (REIA) [3], TEPCO has selected a "batch discharge" methodology. Safety Guide RS-G-1.8 para 5.18 [4]

states: "In the case of batch discharges, the material for discharge is adequately characterized by the volume of the batch and the radionuclide composition of a sample taken at the reservoir from the homogenized batch prior to discharge".

In a description of the ALPS discharge facility, also included in its REIA, TEPCO has provided details of its source monitoring plan. Prior to discharge, ALPS treated water will be transferred from tanks at the FDNPS site into measurement and confirmation facilities – three operating in parallel – into which volumes of ALPS treated water will be transferred. Circulation and agitation will be applied in each measurement and confirmation facility to ensure homogeneity of the ALPS treated water prior to collecting representative samples and performing confirmatory measurements to ensure that the sum of ratios of the legally required activity concentrations of radionuclides other than ^3H is less than one. Each volume of ALPS treated water transferred to each measurement and confirmation facility is a 'batch' (according to RS-G-1.8 para 5.18 [4]). The total radionuclide content of all batches discharged per annum defines the source which is compared to authorized limits on discharges (in Bq/year).

This ILC, based on samples collected from the K4-B ALPS measurement and confirmation facility in March 2022 with IAEA observation, 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 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.

1.2. OBJECTIVE

This publication reports the results of the first 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 first 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

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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. The results of the ILCs are also presented in charts in Annex II.

2. DESIGN AND PARTICIPATING LABORATORIES

2.1. DESIGN

The ILC was comprised of two components:

1. 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 current design stage 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. The results were statistically evaluated according to the methodology described in section II-C. Recognising that all laboratories are subject to resource constraints and that it is implausible to expect any single laboratory other than TEPCO to have the capability to analyse for all radionuclides, the IAEA encouraged participating laboratories to analyse for as many radionuclides as possible, subject to the analytical capability and resources available.

2. A qualitative interpretation of activity concentrations or detection limits reported by any participating laboratory for radionuclides in addition to those included in TEPCO's ALPS treated water source term for which the samples have been analysed. The IAEA laboratories analysed the ALPS treated water samples as comprehensively as possible for additional radionuclides for which they have analytical capability and asked all participating laboratories to do likewise. Of particular interest were high yield activation and fission products, and actinides that might reasonably be expected to be present in cooling water from a nuclear facility such as FDNPS. For guidance, all participating laboratories were provided with data including the results of past sampling and analysis activities at various points within the ALPS processing stream that have been undertaken by TEPCO and by other resources related to radionuclide source terms in other nuclear facilities and decommissioning activities. These results were not statistically evaluated. Rather, this component of the ILC constituted a check for any possible additional radionuclides in the ALPS treated water samples and allowed activity concentrations and detection limits reported to be evaluated as information values, with reference to relevant authorised discharge limits as appropriate.

This ILC constituted a comprehensive radiological characterization of samples of ALPS treated water. While it was not expected that any significant (or detectable) amounts of these radionuclides would be present in the samples analysed, the IAEA considered that this robust, independent, and comprehensive assessment of the radiological content of ALPS treated water samples, availing of the broad collective analytical capacity of all participating laboratories, was necessary, not least for enhancing transparency and promoting confidence in TEPCO's source monitoring.

It can also be noted that prior to the discharge of ALPS treated water to the sea, a pre-operational analysis should be carried out to identify, inter alia, "the inventories of radionuclides that would result in discharges during the operation of a facility or the conduct of an activity" (para. 5.20 of GSG-9) [5]. This comprehensive radiological characterization also provides evidence supporting that this aspect of the pre-operational analysis has been carried out adequately by TEPCO.

TABLE 1. RADIONUCLIDES INCLUDED IN TEPCO'S DESIGN STAGE ALPS TREATED WATER SOURCE TERM

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

Laboratories participating in the ILC were given freedom to determine activity concentrations of radionuclides by any appropriate method of their choice. Approximate activity concentrations (i.e., prior to homogenisation of ALPS treated water in the measurement and confirmation facility at FDNPS) of common radionuclides (^{60}Co, ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{129}I, ^{134}Cs, ^{137}Cs, ^{3}H, ^{14}C and ^{99}Tc) in the K4-B tanks from which the samples were collected has been published by TEPCO. 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 [6].

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 (24 March 2022). 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 France, 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 190 member laboratories globally that is coordinated jointly by RML and TERC. It 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 Tables 3 and 4.

TABLE 2. LABORATORIES PARTICIPATING IN THE ILCS

Identifier	Laboratory
IAEA	IAEA Laboratories, Austria and Monaco
LS	Spiez Laboratory (Labor Spiez), Switzerland
IRSN	Institut de Radioprotection et de Sûreté Nucléaire, France
LANL	Los Alamos National Laboratory, United States of America
KINS	Korea Institute of Nuclear Safety, Republic of Korea
TEPCO	Tokyo Electric Power Company Holdings, Inc., Japan

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

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

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] ^{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.

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

[4] A combined value for ^{243,244}Cm was reported by IRSN.

TABLE 4. ADDITIONAL RADIONUCLIDES ANALYSED BY PARTICIPATING LABORATORIES

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO
^{59}Fe					🟨	🟨
^{58}Co					🟨	🟨
^{65}Zn					🟨	🟨
^{86}Rb						🟨
^{89}Sr						🟨
^{90}Y[1]						🟩
^{91}Y						🟨
^{94}Nb	🟨	🟨	🟨	🟨		
^{95}Nb					🟨	🟨
^{103}Ru					🟨	🟨
^{103m}Rh						🟨
^{106}Rh						🟨
^{110m}Ag					🟨	🟨
^{113m}Cd				🟨		🟨
^{115m}Cd						🟨
^{119m}Sn						🟨
^{123}Sn						🟨
^{126}Sn	🟨					🟨
^{124}Sb					🟨	🟨
^{123m}Te						🟨
^{125m}Te[1]						🟩
^{127}Te						🟨
^{127m}Te						🟨
^{129}Te						🟨
^{129m}Te						🟨
^{135}Cs						🟩
^{136}Cs						🟨
^{133}Ba	🟨	🟨	🟨	🟨	🟨	
^{137m}Ba						🟩
^{140}Ba						🟨
^{141}Ce					🟨	🟨
^{144}Pr					🟨	🟨
^{144m}Pr						🟨
^{146}Pm	🟨		🟨			
^{148}Pm						🟨
^{148m}Pm						🟨
^{152}Eu	🟨	🟨	🟨	🟨	🟨	🟨
^{153}Gd						🟨
^{160}Tb						🟨
^{210}Pb		🟨			🟨	
^{228}Ac		🟨			🟨	
^{228}Th	🟨					
^{230}Th	🟨			🟩		
^{232}Th	🟨			🟩		

TABLE 4. ADDITIONAL RADIONUCLIDES ANALYSED BY PARTICIPATING LABORATORIES (CONTINUED)

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO
^{233}U		yellow	yellow	yellow		
^{235}U	yellow	green	yellow	green		
^{236}U		yellow	yellow	green		
^{236}Np				yellow		
^{242}Pu		yellow	yellow	green		
^{244}Pu				yellow		
^{242m}Am						yellow
^{243}Am		yellow				yellow
^{242}Cm		yellow				yellow
^{243}Cm						yellow
^{245}Cm					yellow	
Gross α	green	yellow	yellow		yellow	yellow
Gross β	green	green	green		green	
Total stable Sr				green		

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] TEPCO reported results for ^{90}Y and ^{125m}Te derived from the measured activity concentrations of ^{90}Sr and ^{125}Sb respectively.

3. COLLECTION AND PREPARATION OF ALPS TREATED WATER SAMPLES

Samples for this ILC were collected on 24 March 2022 from the K4-B ALPS facility at FDNPS, with observation by IAEA staff members.

The K4-B tank group is one of three measurement and confirmation units in the ALPS discharge facility that was under construction at FDNPS at that time. It is comprised of 10 individual 1,000 m³ storage tanks that have been re-utilized for this purpose. The tanks are interconnected and the contents are circulated by pumping. Additionally, the water in individual tanks is agitated Previously, TEPCO had performed a circulation and agitation experiment at K4-B to demonstrate the adequacy of its method for ensuring the homogenisation of the ALPS treated water content prior to sampling [7]. Once the discharge facility has been completed, this will become normal practice under TEPCO's source monitoring plan.

Prior to sampling for this ILC, circulation and agitation was conducted in the same way by TEPCO for 14 days to ensure inter sample homogeneity and, thus, suitability of the samples for the provision of comparable results of analyses.

The samples were collected directly from one of the interconnection pipes at K4-B. A volume of 25.3 L of ALPS treated water was collected for each participating laboratory: 5 x 5 L in plastic cubitainers and 3 x 100 mL in amber glass bottles. 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 received by the IAEA laboratories in Monaco and Austria and by the participating ALMERA laboratories between August and October 2022.

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 [8]. 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 sample 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 sample would be desorbed from the walls of the plastic sampling container and any particulates present to provide the most conservative analysis results.

IRSN analysed for tritium and gamma-emitting radionuclides without pre-concentration. All other analyses included pre-treatment comprised of full sample digestion by evaporation or addition of concentrated organic acids or radiochemical separation.

KINS utilised acidification for the pre-treatment of samples for gross alpha and gross beta only.

LANL analysed tritium and 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.

LS performed pre-treatment of acidification using concentrated nitric acid on the 5L cubitainers, and no pre-treatment for the samples in 100 mL amber glass bottles.

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

4.2. GAMMA-EMITTING RADIONUCLIDE ANALYSES (^{54}Mn, ^{60}Co, ^{106}Ru, ^{125}Sb, ^{129}I, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu, ^{155}Eu, ^{241}Am)

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. IRSN's method complied with ISO 10703:2021 [9].

In most cases, a single sample was prepared in a Marinelli beaker and counted without further treatment. The volumes of the beakers varied from 0.5 L (IRSN) through 1 L (IAEA, KINS and LS), to 5L (TEPCO). An exception was LANL: a 5L sample was evaporated to remove tritium

and then digested with nitric, hydrochloric, hydrofluoric, and perchloric acids to make a stock solution. Then, the sample was counted as a slurry in a 200 mL container.

Efficiency calibrations were determined from measurements of calibration sources prepared from traceable reference standards in most cases. A software-based technique to determine efficiency calibrations, Mirion's LabSOCS (Laboratory Sourceless Calibration Software), was additionally used in IAEA and LS. Correction for true coincidence summing was undertaken, when necessary, also using LabSOCS (IAEA, LS), EFFTRAN (IAEA, KINS) and GESPECOR (IRSN). TEPCO determined correction factors experimentally.

Spectral analysis was undertaken using a range of software including Mirion's Genie 2000 (IAEA, IRSN, KINS) and Apex Gamma (LS), Seiko EG&G's Gamma Station (TEPCO), and LANL's own PeakEasy. Nuclide data from DDEP/ Nucléide-Lara was used in all cases except Table of Isotopes, 6th Edition (TEPCO) and the Lund/LBNL Nuclear Data Search (LANL).

All participating laboratories reported that regular quality control procedures to control peak centroid, detector resolution, efficiency and background were undertaken.

The sample was analysed for ^{241}Am using the same method by KINS.

Detection limits for a number of gamma-emitting radionuclides not included in TEPCO's source term were determined using the same methods by participating laboratories.

4.3. ^{129}I ANALYSIS

The sample was analysed for ^{129}I by IAEA, IRSN, KINS and LS using the gamma-ray spectrometry method described in the previous section. Due to the volatility of iodine, LANL's sample preparation method involving evaporation was unsuitable. Therefore, for this radionuclide, a 200 mL untreated sample was counted and analysed as described above by LANL. TEPCO selected a different analytical method using ICP-MS after oxidation state adjustment for the determination of ^{129}I.

4.4. ^{3}H ANALYSIS

IAEA and IRSN added an aliquot of sample directly into a Teflon-coated LSC vial and mixed it with a scintillation cocktail. It was stored in a dark location for approximately 12 hours prior to measurement by liquid scintillation counting. Recovery was based on a metrologically traceable standard solution, using the same region of interest as the samples [10].

LS, KINS and TEPCO prepared the sample by removing organic matter and interfering radionuclides via distillation [10,11]. LANL used column-purification (Eichrom tritium column) for this purpose.

4.5. ^{14}C ANALYSIS

Samples were prepared by creating the evolution of CO_2 followed by collection in an absorbing solution (e.g. Carbosorb) and mixed with a scintillation cocktail (e.g. Permafluor E+). All samples were measured using liquid scintillation counting.

IRSN and TEPCO utilized wet oxidation for the evolution of CO_2; KINS employed a pyrolizer to combust and trap oxidized samples [12,13].

4.6. ^{90}Sr ANALYSIS

All laboratories followed the general procedure to selectively remove impurities and interfering nuclides using extractive chromatographic resin, and gravimetric recovery of Sr ions via strontium carbonate precipitation.

The removal of impurities and interfering radionuclides can be achieved using multiple techniques. IAEA, IRSN and LS used dicyclohexyl-18-crown-6-ether-based Sr resin column and KINS used cation exchange resin (Dowex 50W-X8) [14,15].

IAEA and KINS measured Sr-90 using liquid scintillation counting, LS used ^{90}Sr by low level gas proportional counting [16] while TEPCO used a plastic scintillation detector [17].

4.7. ^{55}Fe ANALYSIS

IAEA, LS, IRSN and KINS used an anion chromatography resin (e.g. 1-X4 Biorad) for chemical separation of ^{55}Fe from the sample matrix followed by liquid scintillation counting [18]. TEPCO selected a different analytical method using an anion chromatography resin for chemical separation, hydroxide precipitation for source preparation and a low-energy photon detector (Ge-LEPS) for analysis [19,20].

IAEA results were provided by subcontractor Laboratorio de Medidas de Baja Actividad de la Universidad del Pais Vasco, accreditation number 350/LE560 under Entidad Nacional de Accreditacion.

4.8. ^{63}Ni ANALYSIS

All laboratories used a chemical separation with an anion exchange resin (e.g. 1-X4 Biorad) followed by a selective Ni extractive chromatographic resin to extract ^{63}Ni from the sample matrix. Measurements were conducted by liquid scintillation counting [18,21].

IAEA results were provided by subcontractor Laboratorio de Medidas de Baja Actividad de la Universidad del Pais Vasco, accreditation number 350/LE560 under Entidad Nacional de Accreditacion.

4.9. ^{99}Tc ANALYSIS

IAEA, LS, IRSN and KINS separated ^{99}Tc from the sample matrix using an extractive chromatographic resin [22]. Analysis was then performed by ICP-MS. Spiez utilised sf-ICP-MS [23].

TEPCO did not separate ^{99}Tc from the sample matrix and used a simple dilution followed by analysis using ICP-MS [21].

IAEA results were provided by subcontractor Laboratorio de Medidas de Baja Actividad de la Universidad del Pais Vasco, accreditation number 350/LE560 under Entidad Nacional de Accreditacion.

4.10. ^{79}Se ANALYSIS

After selective removal of impurities and interfering nuclides by sedimentation and TEVA resin, hydroxylamine hydrochloride was added to precipitate Se. After filtering, dissolution and mixing with scintillation cocktails, ^{79}Se was measured using liquid scintillation counting [21].

4.11. GROSS ALPHA ANALYSIS

Gross alpha analysis is used to screen the total alpha emitting radionuclide content in the sample by either direct count, preconcentration (evaporation, co-precipitation) and measurement.

IAEA removed alkali and alkaline earth metal ions using a calcium phosphate co-precipitation. The precipitation was then dissolved in 2M H_3PO_4 and scintillation cocktail added. Measurement of gross alpha activity was performed using liquid scintillation counting [24,25].

LS utilised a direct counting method where filtered sample was mixed with Ultima Gold AB and measurements were performed using liquid scintillation counter.

KINS pre-concentrated the sample which was transferred onto a planchette and dried. Measurements were performed using alpha-beta counter [24].

IRSN performed gross alpha by evaporation of a known volume of water on a stainless-steel disk with the use of a ZnS scintillator detector [26,27].

At TEPCO, alkali and alkaline earth metal ions were removed 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 [28].

4.12. ALPHA-EMITTING RADIONUCLIDE ANALYSIS

Selective removal of impurities and interference radionuclides sample matrix using an extractive chromatographic resin, with recovery measured via addition of suitable tracers. Sources for alpha-particle spectrometry were prepared by electrodeposition on stainless steel discs or via micro co-precipitation.

LS measured the separated actinides (^{241}Am, ^{244}Cm, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{234}U and ^{238}U) using sf-ICP-MS [29,30].

IRSN followed ISO 13167:2015 using alpha spectrometry to measure ^{238}Pu, $^{239+240}$Pu, ^{241}Am and ^{243,244}Cm [31]. Additional radionuclides ^{242}Pu, ^{243}Am, and ^{242}Cm were measured using the same method. ^{233}U, ^{234}U, ^{238}U ^{235}U and ^{236}U were measured by ICP-MS isotopic dilution with an internal isotopic standard [32].

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

LANL pre-treated samples using evaporation to remove tritium. Chemical separations including precipitations and anion exchange chromatography were performed and samples were measured using either dynamic multi-ion counting or multi-static total evaporation TIMS analysis. Samples were analysed for ^{236}Np, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{244}Pu ^{233}U, ^{235}U, ^{236}U, ^{234}U and ^{238}U. Additionally, LANL measured ^{230}Th and ^{232}Th by multi-ion counting ICP-MS.

4.13. ADDITIONAL RADIONUCIDES 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 Table I-7 of TEPCO's design stage 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 of provided in an earlier IAEA report [33].

Results for additional radionuclides not included in the source term were determined using the same method and reported by TEPCO.

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. This method is described in detail in Appendix II.

In brief, for results that could be intercompared quantitatively (i.e. for each radionuclide for which activity concentrations above detection limits were reported by at least two laboratories) a comparison reference value x_{ref} was determined as a power-moderated mean of the combined results [34]:

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

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 [35], 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" [36, 37]. 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 2).

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 $\zeta = \pm 3$ lines are discrepant.
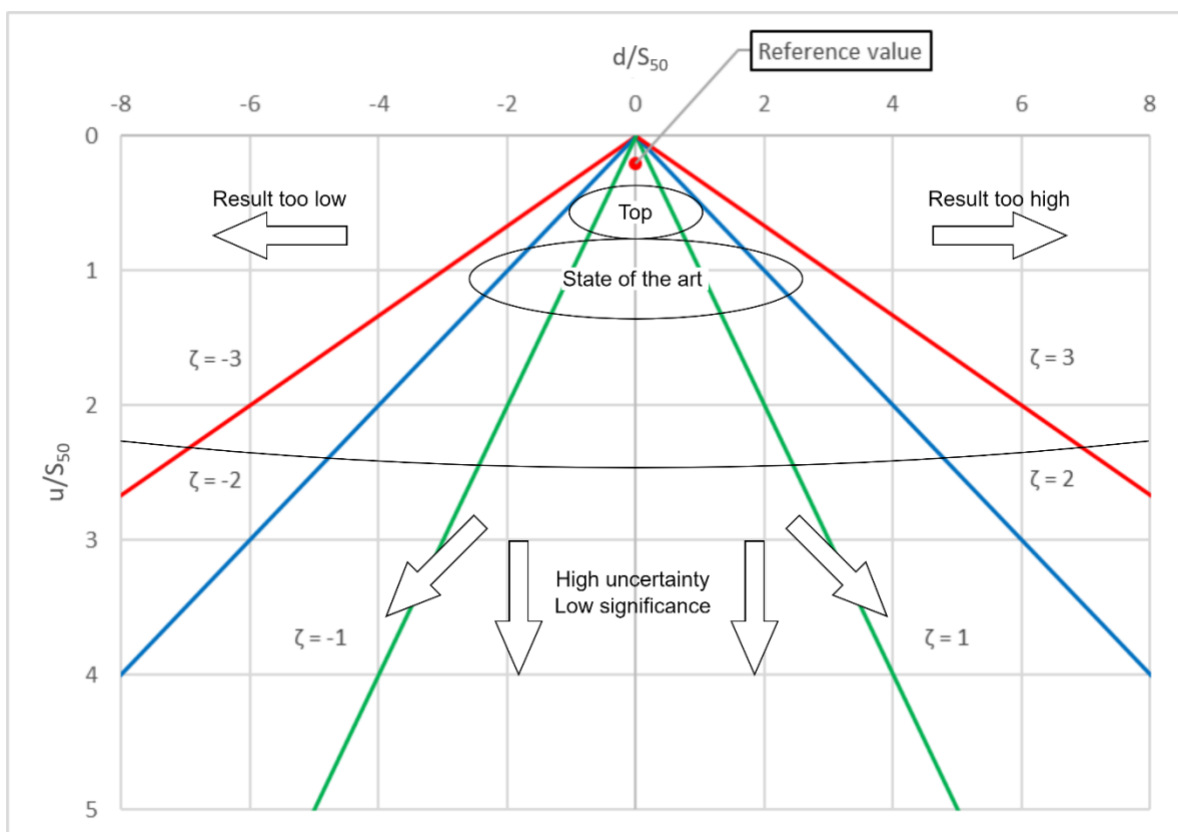

FIG. 2. Interpretation of a PomPlot (adapted from Pommé , 2006 [36]).

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), 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. ζ (Zeta) scores are presented in section 6.3 and PomPlots (Figures 3 to 14) in section 6.4. Charts showing the activity concentrations and detection limits reported are presented in Appendix III (Figures 17 to 41). 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 x $\pm$ y, where x is the activity concentration and y is the numerical value of the combined standard uncertainty, i.e. with a coverage factor of $k = 1$. In cases where an activity was not detected, the detection limit z of the analytical method used 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 [38].

6.1.2.   Reference date

All activity concentrations for were reported at a reference date of 24 March 2022.

6.2. RESULTS REPORTED

TABLE 5. ACTIVITY CONCENTRATIONS (Bq/L) IN K4-B ALPS TREATED WATER (SOURCE TERM RADIONUCLIDES)

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO	Reference value
^{3}H	151500 ± 1300	146000 ± 9000	160400 ± 2800	145800 ± 1500	165800 ± 7300	145700 ± 5200	152300 ± 3400
^{14}C	-	15.2 ± 1.1	13.25 ± 0.84	-	-	13.79 ± 0.96	14.01 ± 0.58
^{54}Mn	<0.048	<0.85	<0.17	<0.023	<0.042	<0.050	-
^{55}Fe	<0.24	<0.47	<0.098	-	<0.24	<19	-
^{60}Co	0.382 ± 0.020	0.373 ± 0.037	0.387 ± 0.044	0.361 ± 0.020	0.380 ± 0.010	0.373 ± 0.038	0.3764 ± 0.0081
^{63}Ni	2.799 ± 0.090	2.41 ± 0.15	2.018 ± 0.087	-	3.10 ± 0.20	<8.1	2.57 ± 0.24
^{79}Se	-	-	-	-	-	<0.85	-
^{90}Sr[1]	0.32 ± 0.14	0.41 ± 0.12	0.404 ± 0.019	-	0.430 ± 0.030	0.399 ± 0.020	0.405 ± 0.015
^{99}Tc	0.792 ± 0.017	0.63 ± 0.15	<0.0037	-	0.750 ± 0.060	0.74 ± 0.21	0.776 ± 0.025
^{106}Ru	<0.42	<5.9	<1.4	<0.097	<0.32	<0.42	-
^{125}Sb	0.1010 ± 0.0090	<1.3	<0.39	0.088 ± 0.010	0.150 ± 0.010	0.150 ± 0.038	0.119 ± 0.017
^{129}I	2.03 ± 0.16	1.87 ± 0.16	1.14 ± 0.12	<20	1.90 ± 0.30	2.126 ± 0.081	1.81 ± 0.19
^{134}Cs	0.0220 ± 0.0070	<0.47	<0.13	0.0210 ± 0.0050	0.0210 ± 0.0030	<0.058	0.0212 ± 0.0025
^{137}Cs	0.471 ± 0.025	0.466 ± 0.038	0.472 ± 0.037	0.4660 ± 0.0060	0.502 ± 0.031	0.517 ± 0.051	0.4705 ± 0.0071
^{144}Ce	<0.33	<4.4	<1.5	<0.051	<0.26	<0.60	-
^{147}Pm	-	-	-	-	-	<0.33	-
^{151}Sm	-	-	-	-	-	<0.013	-
^{154}Eu	<0.054	<0.56	<0.26	<0.049	<0.046	<0.072	-
^{155}Eu	<0.069	<0.62	<0.43	0.023 ± 0.012	<0.050	<0.19	-
^{234}U	<0.00068	<0.0013	0.00078 ± 0.00024	0.0007080 ± 0.0000026	0.000740 ± 0.000047	<0.031	0.0007103 ± 0.0000091
^{238}U	0.00068 ± 0.00022	<0.00095	0.00114 ± 0.00028	0.00064137 ± 0.00000093	0.000640 ± 0.000013	<0.031	0.000676 ± 0.000069
^{237}Np	-	<0.0041	<0.00000016	0.0000000337 ± 0.0000000013	<0.00000068	<0.031	-

TABLE 5. ACTIVITY CONCENTRATIONS (Bq/L) IN K4-B ALPS TREATED WATER (SOURCE TERM RADIONUCLIDES, CONTINUED)

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO	Reference value
^{238}Pu	<0.00043	<0.0030	<0.0014	-	-	<0.031	-
^{239}Pu	<0.00031^3	0.0042 ± 0.0020^3	<0.00000018	0.00006405 ± 0.00000037	<0.0000058	<0.031	-
^{240}Pu			<0.00000025	0.00005618 ± 0.00000049	<0.000020	<0.031	-
^{241}Pu	-	<0.062	<0.00000034	0.000203 ± 0.000032	<0.0072	<0.84	-
^{241}Am	0.0043 ± 0.0013	<0.017	<0.56	-	<0.00020	<0.031	-
^{244}Cm	-	<0.012^4	-	-	<0.0075	<0.031	-

Notes: [1] A value of 0.399 ± 0.020 for ^{90}Y, derived from the measured activity concentration of ^{90}Sr, was reported by TEPCO.
[2] A value of 0.0346 ± 0.0087 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 IRSN.
[4] A combined value for ^{243,244}Cm was reported by IRSN.

6.3.ζ (ZETA) SCORES

TABLE 6. ζ (ZETA) SCORES IN K4-B ALPS TREATED WATER

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO
^{3}H	-0.23	-0.73	2.03	-1.86	1.89	-1.23
^{14}C	-	1.35	-1.07	-	-	-0.27
^{60}Co	0.31	-0.09	0.25	-0.89	0.44	-0.10
^{63}Ni	0.94	-0.63	-2.29	-	1.91	-
^{90}Sr	-0.62	0.05	-0.09	-	0.89	-0.38
^{99}Tc	0.74	-1.01	-	-	-0.47	-0.20
^{125}Sb	-1.03	-	-	-1.76	1.79	0.89
^{129}I	1.01	0.28	-3.29	-	0.30	1.66
^{134}Cs	0.13	-	-	-0.05	-0.08	-
^{137}Cs	0.02	-0.12	0.05	-0.78	1.06	0.94
^{234}U	-	-	0.29	-0.27	0.65	-
^{238}U	0.00	-	1.73	-0.51	-0.53	-

Note: Evaluation criteria (99.7% confidence level): ζ score between -3 and 3: corresponding result evaluated as agreeing with the reference value; ζ score greater than 3 or less than -3: corresponding result evaluated as discrepant.

6.4.POMPLOTS
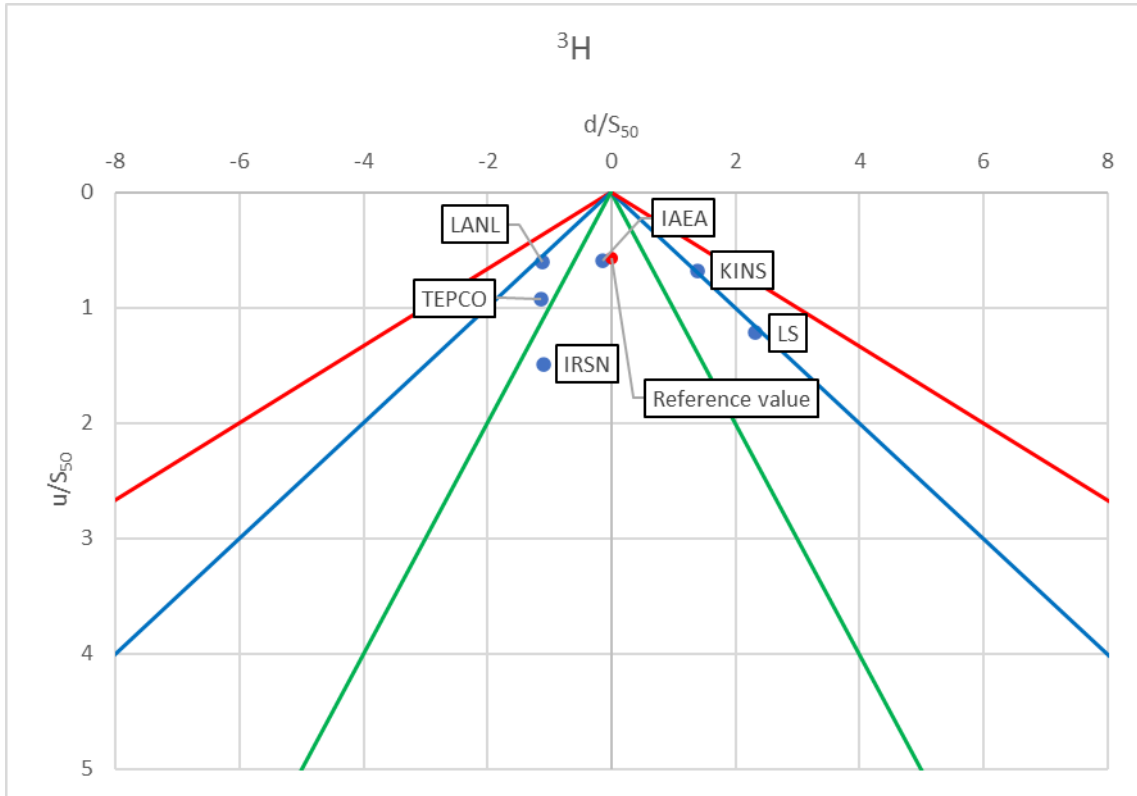
FIG. 3. PomPlot for ³H results.
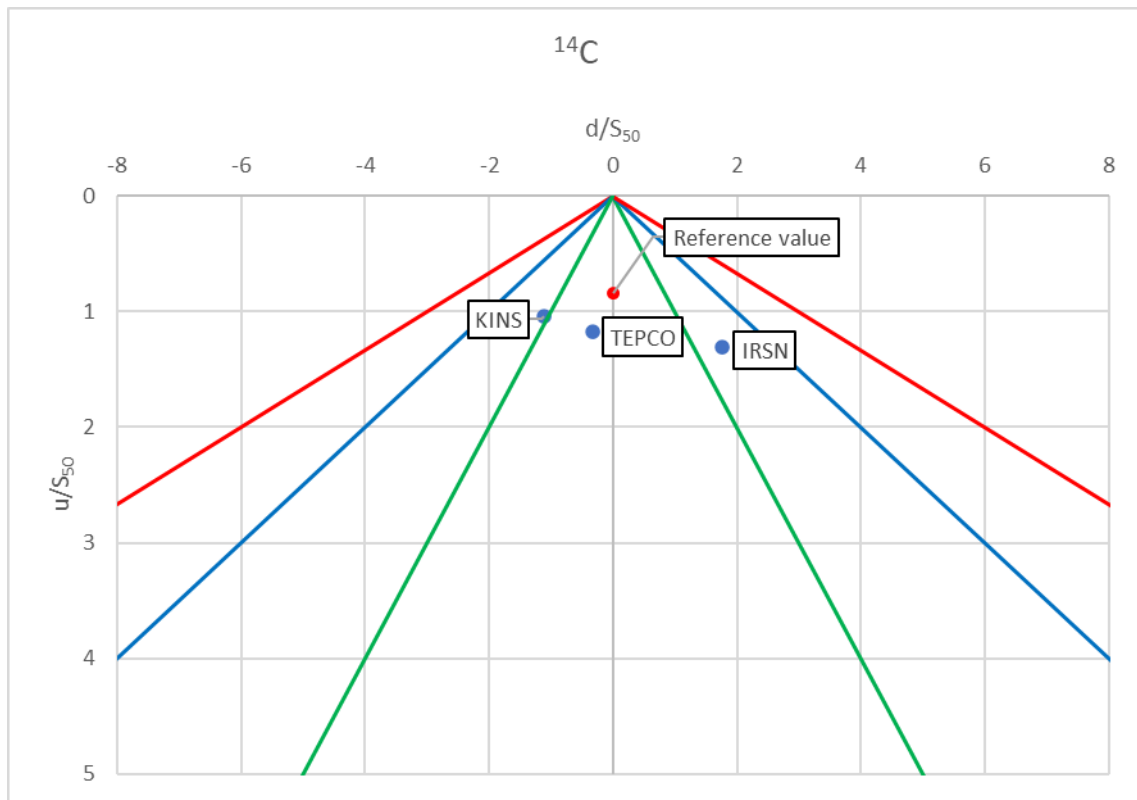
FIG. 4. PomPlot for ¹⁴C results.
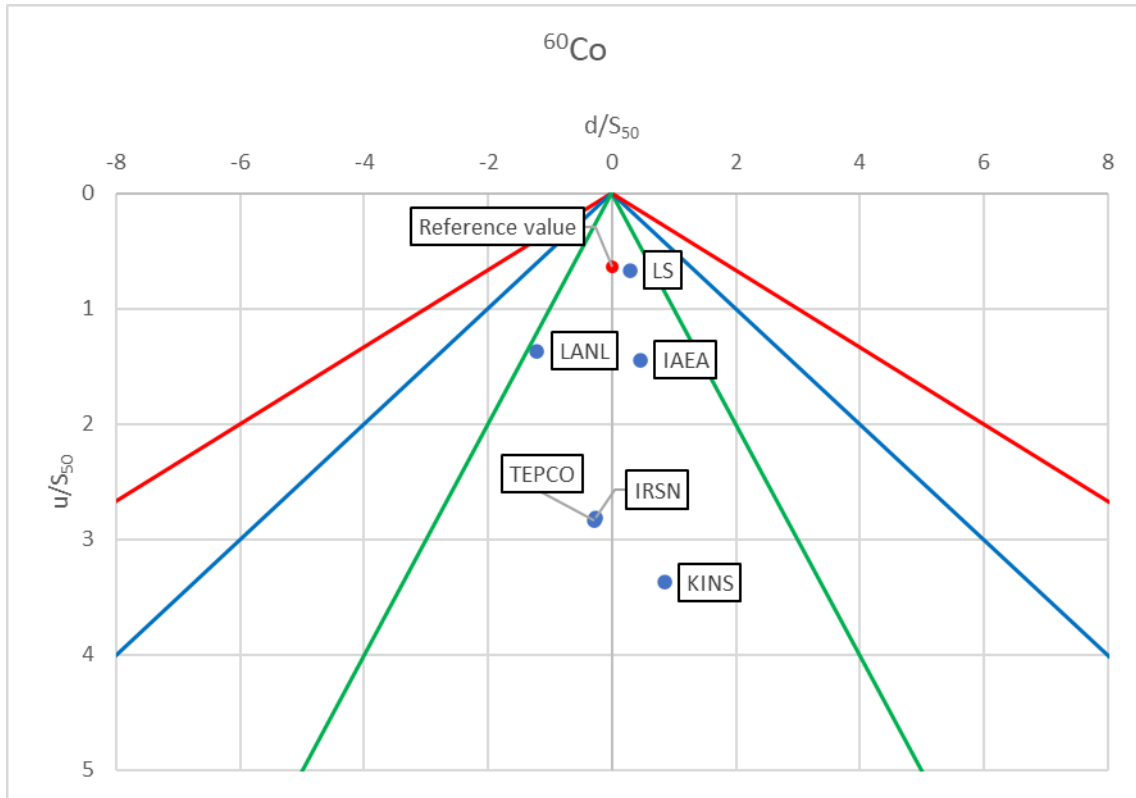
FIG. 5. PomPlot for ^{60}Co results.
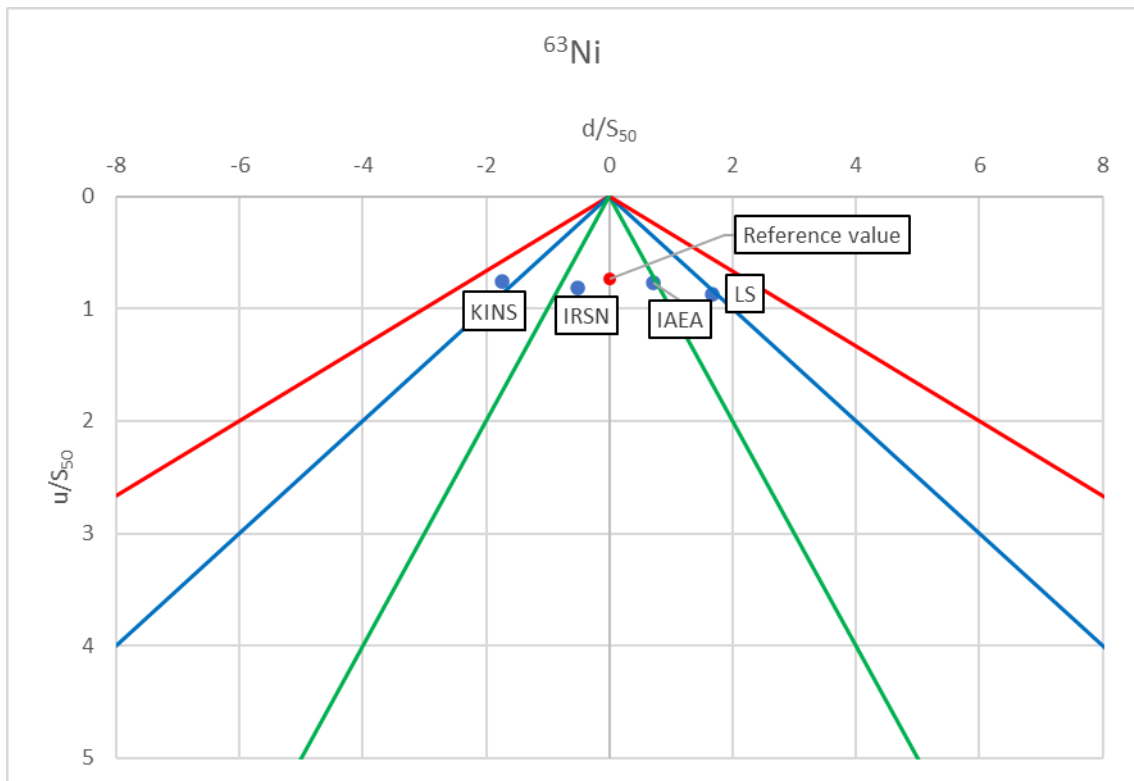
FIG. 6. PomPlot for ^{63}Ni results.
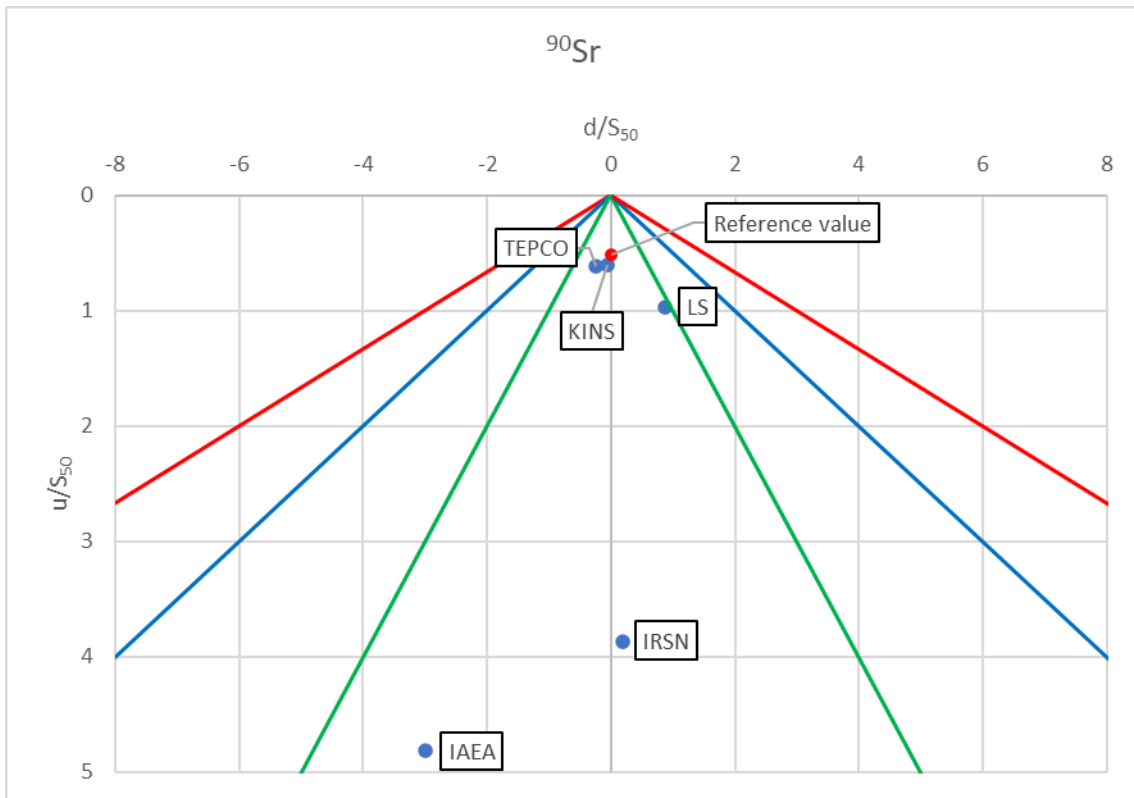
FIG. 7. PomPlot for ^{90}Sr results.
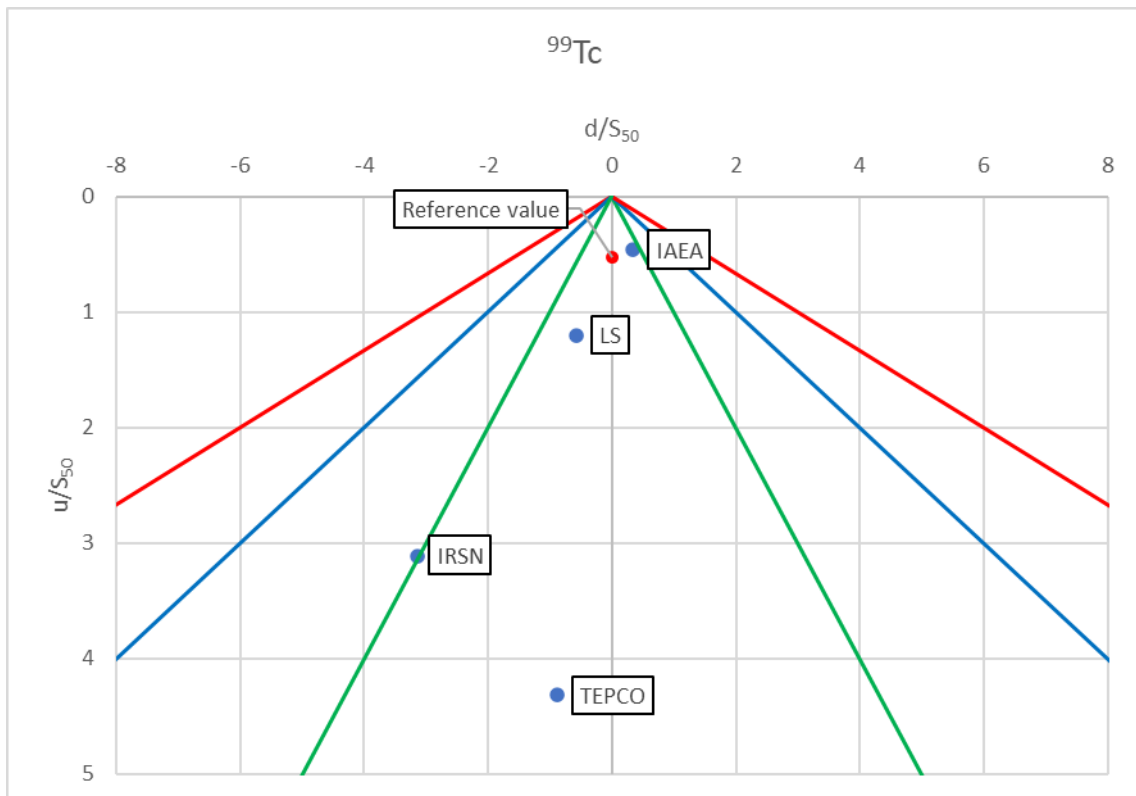
FIG. 8. PomPlot for ^{99}Tc results.
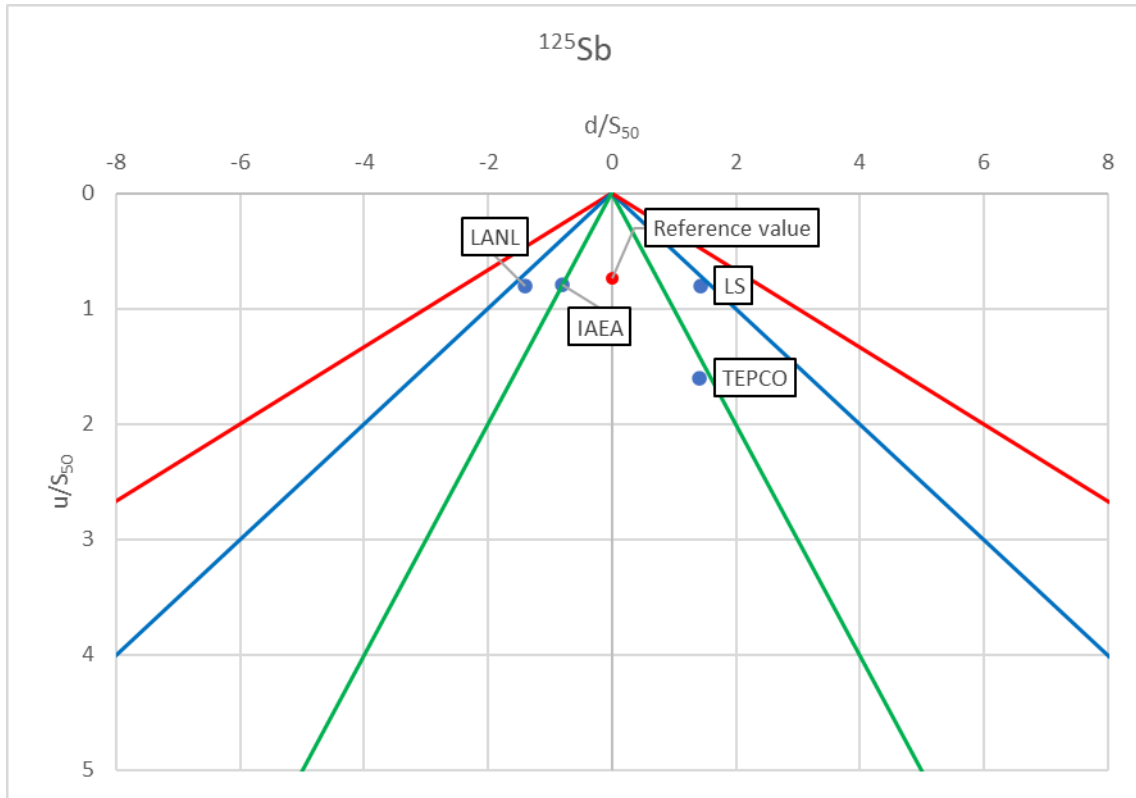
FIG. 9. PomPlot for ^{125}Sb results.
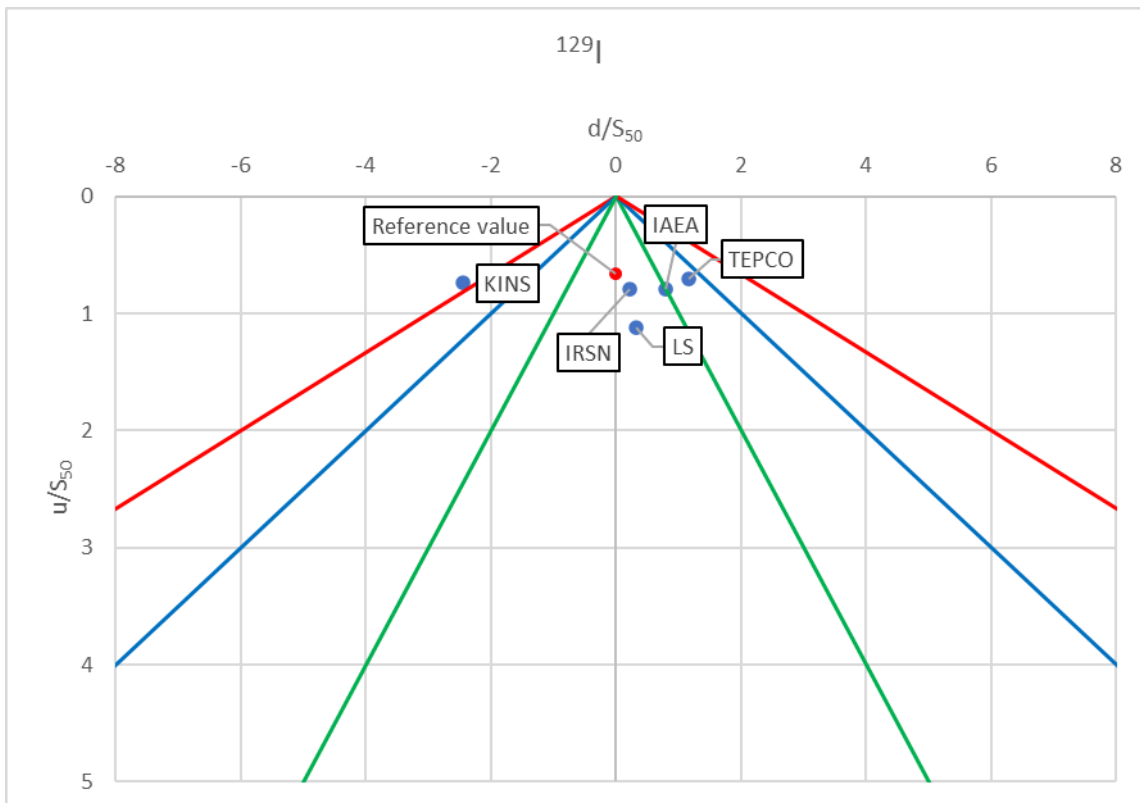
FIG. 10. PomPlot for ^{129}I results.

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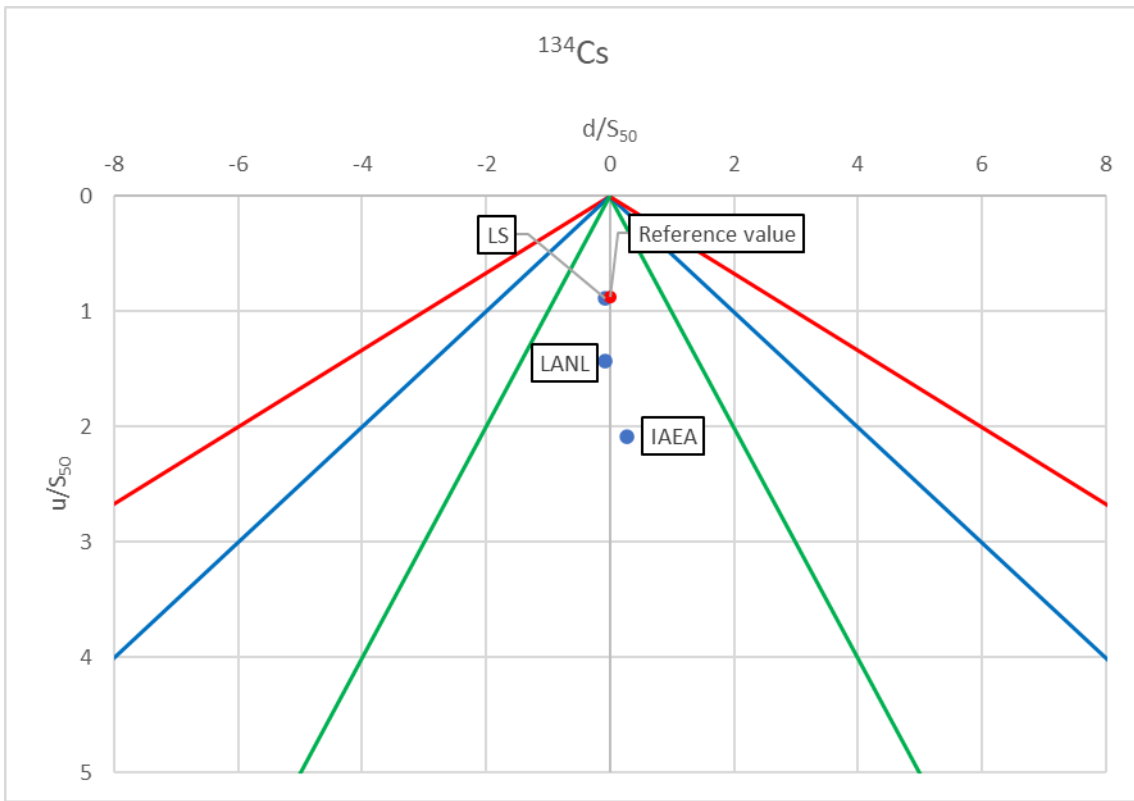
FIG. 11. PomPlot for ¹³⁴Cs results.
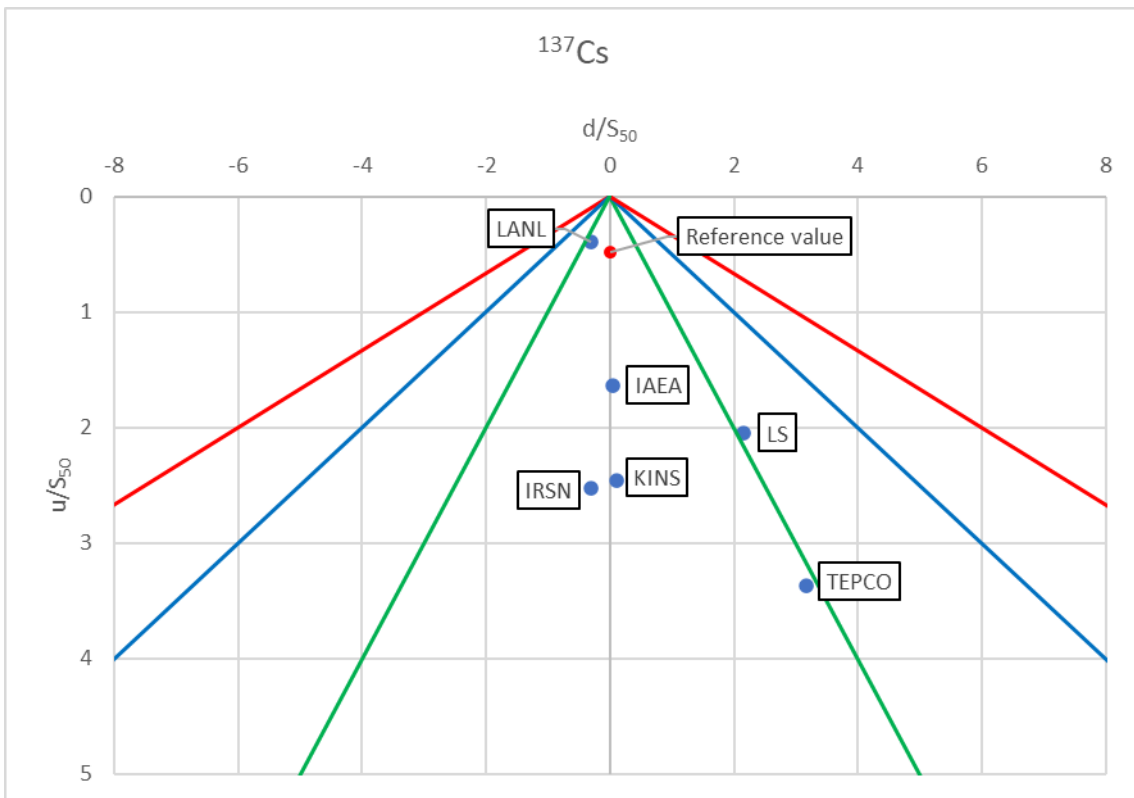
FIG. 12. PomPlot for ¹³⁷Cs results.
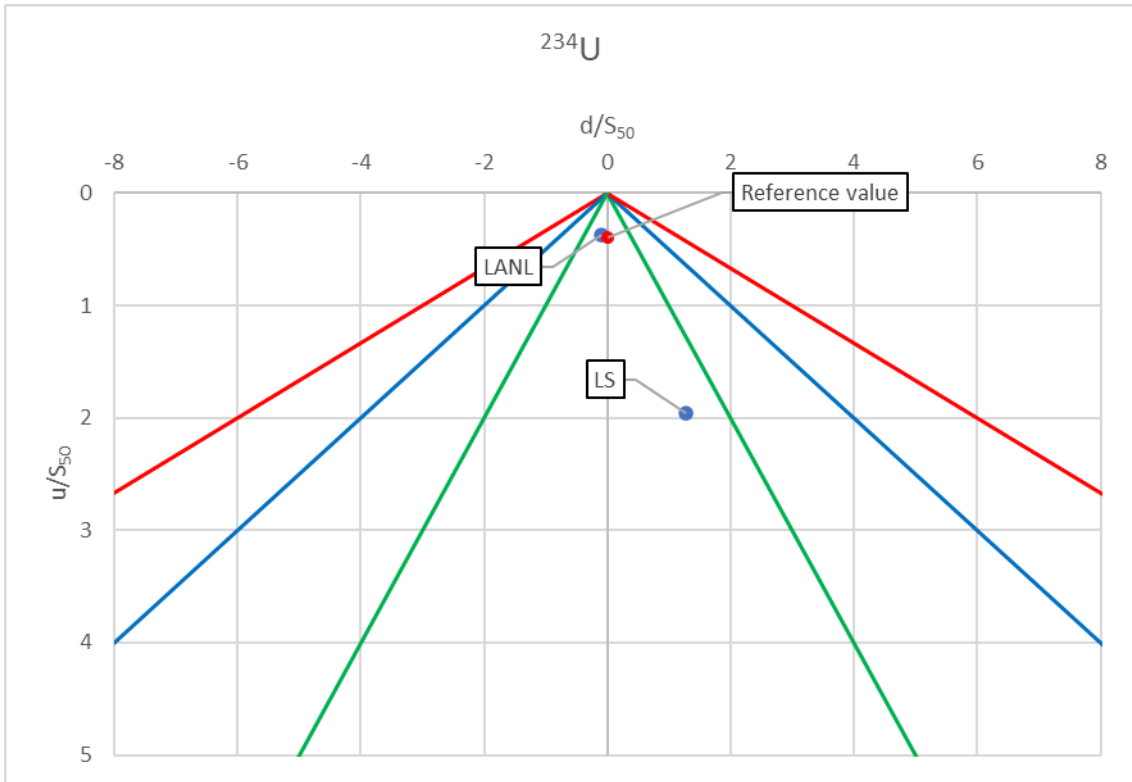
FIG. 13. PomPlot for ^{234}U results.
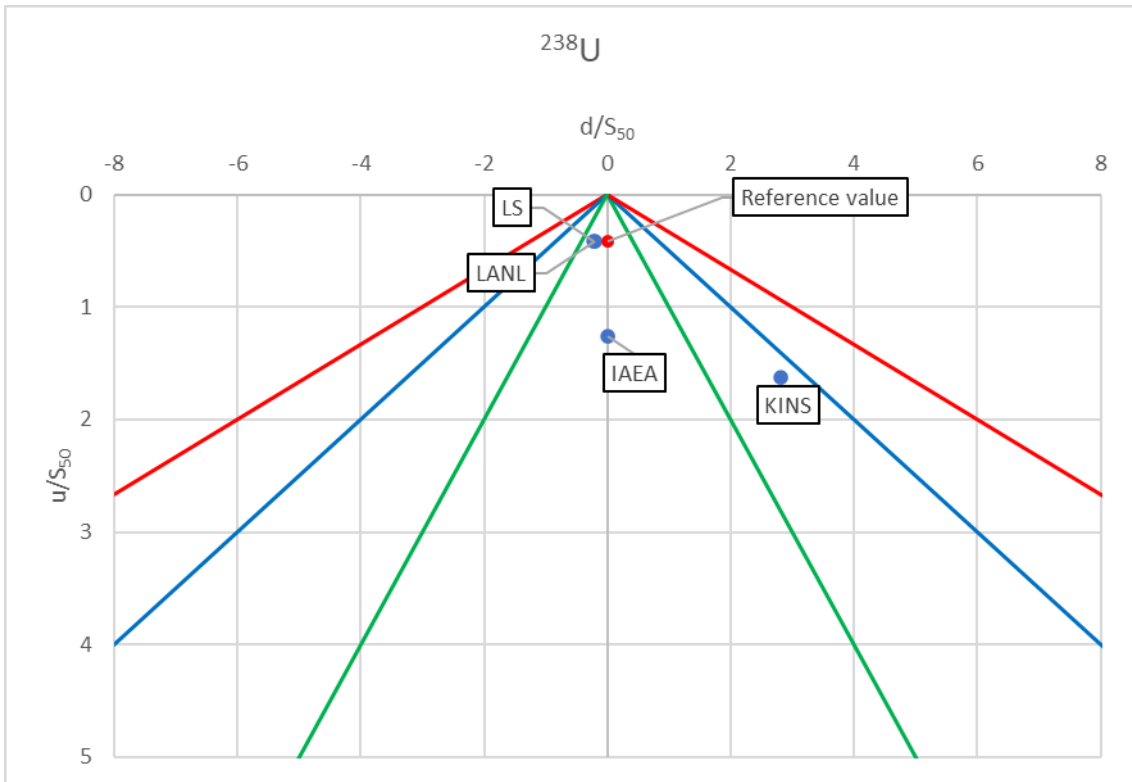
FIG. 14. PomPlot for ^{238}U results.

6.5. ADDITIONAL RESULTS REPORTED

TABLE 7. ACTIVITY CONCENTRATIONS (Bq/L) IN K4-B ALPS TREATED WATER (ADDITIONAL RADIONUCLIDES)

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO
^{59}Fe	-	-	-	-	<2.0	<2.6
^{58}Co	-	-	-	-	<0.26	<0.34
^{65}Zn	-	-	-	-	<0.12	<0.11
^{86}Rb	-	-	-	-	-	<5200
^{89}Sr	-	-	-	-	-	<3.2
^{90}Y	-	-	-	-	-	0.399 ± 0.020
^{91}Y	-	-	-	-	-	<180
^{94}Nb	<0.028	<0.38	<0.11	<0.011	-	-
^{95}Nb	-	-	-	-	<2.6	<0.68
^{103}Ru	-	-	-	-	<1.6	<4.4
^{103m}Rh	-	-	-	-	-	<4.3
^{106}Rh	-	-	-	-	-	<0.42
^{110m}Ag	-	-	-	-	<0.078	<0.057
^{113m}Cd	-	-	-	<27	-	<0.083
^{115m}Cd	-	-	-	-	-	<78
^{119m}Sn	-	-	-	-	-	<0.0032
^{123}Sn	-	-	-	-	-	<18
^{126}Sn	<0.062	-	-	-	-	<0.087
^{124}Sb	-	-	-	-	<0.38	<1.2
^{123m}Te	-	-	-	-	-	<0.20
^{125m}Te	-	-	-	-	-	0.0346 ± 0.0087
^{127}Te	-	-	-	-	-	<16
^{127m}Te	-	-	-	-	-	<16
^{129}Te	-	-	-	-	-	<85
^{129m}Te	-	-	-	-	-	<200

TABLE 7. ACTIVITY CONCENTRATIONS (Bq/L) IN K4-B ALPS TREATED WATER (ADDITIONAL RADIONUCLIDES, CONTINUED)

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO
^{135}Cs	-	-	-	-	-	0.00000339 ± 0.00000033
^{136}Cs	-	-	-	-	-	<29000
^{133}Ba	<0.085	<0.49	<0.16	<0.011	<0.040	-
^{137m}Ba	-	-	-	-	-	0.489 ± 0.048
^{140}Ba	-	-	-	-	-	<160000
^{141}Ce	-	-	-	-	<5.1	<24
^{144}Pr	-	-	-	-	<3.4	<0.60
^{144m}Pr	-	-	-	-	-	<0.0056
^{146}Pm	<0.041	-	<0.17	-	-	<0.048
^{148}Pm	-	-	-	-	-	<60000000000000
^{148m}Pm	-	-	-	-	-	<2.1
^{152}Eu	<0.073	<0.33	<0.36	<0.057	<0.050	<0.13
^{153}Gd	-	-	-	-	-	<0.40
^{160}Tb	-	-	-	-	-	<1.1
^{210}Pb	-	<3.9	-	-	<0.35	-
^{228}Ac	-	<1.6	-	-	<0.11	-
^{228}Th	<0.0083	-	-	-	-	-
^{230}Th	<0.00026	-	-	0.00000337 ± 0.00000031	-	-
^{232}Th	<0.00026	-	-	0.00000234 ± 0.00000016	-	-
^{233}U	-	<0.0021	<0.00048	<0.0000091	-	-
^{235}U	<0.00028	0.000032 ± 0.000002	<0.00048	0.000029839 ± 0.000000061	-	-
^{236}U	-	<0.000014	<0.00048	0.0000001025 ± 0.0000000087	-	-
^{236}Np	-	-	-	<0.000000087	-	-

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TABLE 7. ACTIVITY CONCENTRATIONS (Bq/L) IN K4-B ALPS TREATED WATER (ADDITIONAL RADIONUCLIDES, CONTINUED)

Nuclide	IAEA	IRSN	KINS	LANL	LS	TEPCO
^{242}Pu	-	<0.0050	<0.00000031	0.0000000186 ± 0.0000000021	-	-
^{244}Pu	-	-	-	<0.000000000018	-	-
^{242m}Am	-	-	-	-	-	<0.00021
^{243}Am	-	<0.020	-	-	-	<0.031
^{242}Cm	-	<0.034	-	-	-	<0.031
^{243}Cm	-	-	-	-	-	<0.031
^{245}Cm	-	-	-	-	<0.0000049	-
Gross α	0.020 ± 0.011	<0.23	<0.69	-	<0.20	<0.031
Gross β	0.716 ± 0.022	6.75 ± 0.43	1.799 ± 0.093	-	3.43 ± 0.32	-
Total stable Sr (ng/ml)	-	-	-	1.370 ± 0.030	-	-

7.   CONCLUSION

This ILC has been implemented to check the veracity of the source monitoring undertaken by TEPCO with respect to discharges of ALPS treated water. Samples collected from the K4-B tank group at FDNPS 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 radionuclides included in TEPCO's design stage ALPS treated water source term [3] (Table 5). 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.

Activity concentrations above detection limits were reported by at least two laboratories for 12 radionuclides: ^{3}H, ^{14}C, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{125}Sb, ^{129}I, ^{134}Cs, ^{137}Cs, ^{234}U and ^{238}U. The reported results for these radionuclides have been subjected to strict statistical tests following the same methods used at the top level of radionuclide metrology [34]. Out of a total of the 53 reported results that were assessed using these statistical tests, over 98% (52), were evaluated as agreeing with the respective reference value with a high level of confidence (99.7%). This provides evidence of the high level of competence of all participating laboratories.

The only exception was one "action signal" ζ score of -3.29 for a reported activity concentration of ^{129}I (Table 6).

A small number of inconsistencies – two "warning signals" were also identified (Table 6) – 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. In addition, ALPS treated water constitutes a 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) [7] but at a micro-level this is more uncertain, inter-sample heterogeneity was reported by some participating laboratories performing repeat analyses.

Inter-sample heterogeneity for Pu isotopes, ^{241}Am and to a lesser extent Th isotopes, was observed by some participating laboratories performing repeat analyses, possibly indicating a degree of low-level inhomogeneity for particle-reactive isotopes. The results were comparable between samples for U and Np isotopes. (It should be noted that the activities reported for these radionuclides were several orders of magnitude below the respective regulatory limits for discharge, so this low-level heterogeneity is largely academic.)

Other factors possibly contributing to the identification of a small number of inconsistencies were that the radionuclides, other than ^{3}H, are present in the samples at low levels and the dataset intercompared for each radionuclide was small ($N \leq 6$). Finally, the laboratories used a range of different analytical techniques, some of which are not routinely implemented.

The results for ^{234}U and ^{238}U determined by alpha spectrometry reported by IAEA and KINS were very close to the method detection limit with relative standard uncertainties of around of

30%. In fact, for ^{234}U IAEA reported a detection limit and the uncertainty reported by KINS was greater than five times S_{50}, the typical uncertainty determined from all ^{234}U results reported, determined to be statistically irrelevant and, therefore, does not appear on the respective PomPlot (Figure 13). In contrast, the results reported by LANL and LS determined by low level measurement techniques utilising TIMS and ICP-MS, respectively, were more precise and in good agreement. It can be noted that the presence of uranium isotopes at low levels in ALPS treated water is considered to be naturally occurring.

The maximum relative bias of the results reported for these radionuclides, with respect to the reference value, are presented in Table 8. With the exception of ^{234}U and ^{238}U (naturally occurring) and ^{129}I (which includes an "action signal" result as described above), the results agreed within 30% and in many cases (^{3}H, ^{60}Co, ^{134}Cs, ^{137}Cs) within 10%.

TABLE 8. MAXIMUM RELATIVE BIAS OF INTERCOMPARED RESULTS

Radionuclide	Maximum relative bias (%)
^{3}H	8.8
^{14}C	8.5
^{60}Co	4.1
^{63}Ni	22
^{90}Sr	21
^{99}Tc	19
^{125}Sb	26
^{129}I	37
^{134}Cs	3.7
^{137}Cs	9.8
^{234}U	9.3
^{238}U	68

For radionuclides for which its data could be intercompared quantitatively (^{3}H, ^{14}C, ^{60}Co, ^{90}Sr, ^{99}Tc, ^{125}Sb, ^{129}I, ^{137}Cs), TEPCO's results were in all cases in agreement with the respective reference value (in fact, $\zeta < 2$ in all cases). A PomPlot showing this data is presented in Figure 15. As this chart demonstrates, the standard uncertainties reported by TEPCO were also judged to be appropriate and informative.
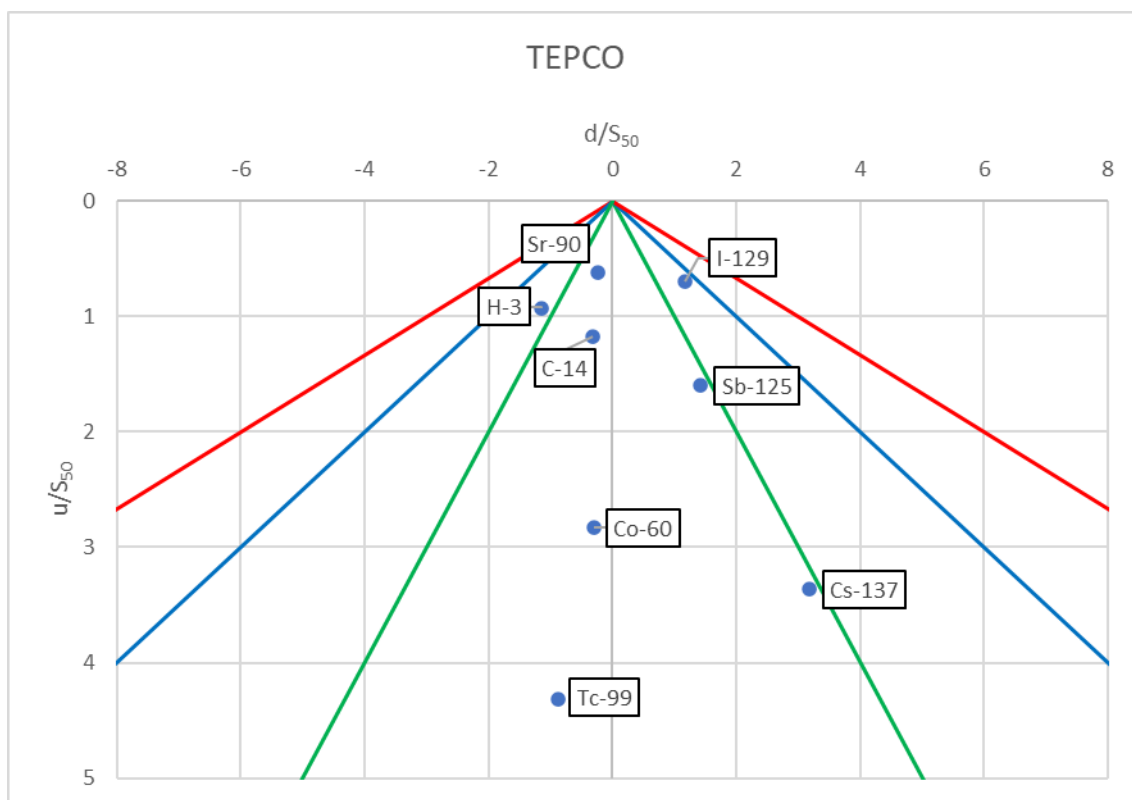
FIG. 15. PomPlot for TEPCO's results.

7.2. ASSESSMENT OF REPORTED DETECTION LIMITS

For other radionuclides, the detection levels reported by participating laboratories were compared qualitatively. Results were reported by at least two participating laboratories in addition to TEPCO for 12 radionuclides (^{54}Mn, ^{55}Fe, ^{106}Ru, ^{144}Ce, ^{154}Eu, ^{155}Eu, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am). In one case (^{244}Cm) just one participating laboratory in addition to TEPCO reported a result. For three radionuclides (^{79}Se, ^{147}Pm and ^{151}Sm) only TEPCO reported results.

A comparison of all of the detection limits reported by TEPCO with respective regulatory limits for all radionuclides in its source term is presented in Table 9. This shows 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'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, ^{241}Am and ^{244}Cm) for which the detection limits were significantly higher than those reported by some other participating laboratories.

TEPCO assesses actinides included in its source term using a screening approach based on gross alpha counting. The gross alpha result – in this case a detection limit of 0.031 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 [3, Table I-6]. As the screening approach is sufficiently conservative, is implemented to a high

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quality, and a procedure is in place describing steps to be taken for effective follow up in the event that a pre-stated action limit is exceeded, this approach is considered to be fit for purpose. It can be noted that the gross alpha values reported in this ILC by four additional laboratories, including the IAEA, all agree within an order of magnitude (Table 7).

It is known, however, that the chemical recovery of the gross alpha method used is determined from a single alpha-emitting radionuclide (^{241}Am). Investigation of the chemical recovery for other targeted radionuclides is recommended to further verify the robustness of this method.

7.3. ADDITIONAL RADIONUCLIDES

The participating ALMERA laboratories were encouraged to analyse and report results for radionuclides in addition to those included in TEPCO's source term that might reasonably be expected to be present in ALPS treated water. The IAEA laboratories did likewise. The results of this comprehensive radiological characterization carried out on the samples using the broad collective analytical capacity of the participating laboratories identified the presence of no additional radionuclides above detection limits, with the exception of low levels of some isotopes of ^{230}Th, ^{232}Th, ^{235}U, ^{236}U, and ^{242}Pu, that were detected by LANL using TIMS, and ^{235}U that was detected by IRSN using ICP-MS (Table 7).

TABLE 9. COMPARISON OF TEPCO'S DETECTION LIMITS WITH RESPECTIVE JAPANESE REGULATORY LIMITS FOR DISCHARGE

	Regulatory limit (RL) (Bq/L)	Detection limit (DL) (Bq/L)	DL relative to RL (%)
^{3}H	60000	210	0.35
^{14}C	2000	1.6	0.080
^{54}Mn	1000	0.047	0.0047
^{55}Fe	2000	19	0.94
^{60}Co	200	0.028	0.014
^{63}Ni	6000	8.1	0.13
^{79}Se	200	0.85	0.43
^{90}Sr	30	0.069	0.23
^{99}Tc	1000	0.43	0.043
^{106}Ru	100	0.42	0.41
^{125}Sb	800	0.10	0.013
^{129}I	9	0.026	0.29
^{134}Cs	60	0.057	0.10
^{137}Cs	90	0.036	0.040
^{144}Ce	200	0.59	0.30
^{147}Pm	3000	0.32	0.011
^{151}Sm	8000	0.012	0.00015
^{154}Eu	400	0.072	0.018
^{155}Eu	3000	0.19	0.0063
^{234}U	20	0.031	0.15
^{238}U	20	0.031	0.15
^{237}Np	9	0.031	0.34
^{238}Pu	4	0.031	0.76
^{239}Pu	4	0.031	0.76
^{240}Pu	4	0.031	0.76
^{241}Pu	200	0.84	0.42
^{241}Am	5	0.031	0.61
^{244}Cm	7	0.031	0.44

7.4. KEY FINDINGS

The results of this ILC demonstrate a high level of accuracy and competence on the part of TEPCO. This should provide confidence in TEPCO's capability for undertaking accurate and precise 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.

The comprehensive radiological characterization carried out on the samples using the broad collective analytical capacity of the IAEA and the ALMERA laboratories identified the presence of no additional radionuclides at significant levels.

APPENDIX I: INSTRUCTIONS SENT TO PARTICIPATING LABORATORIES

Participating laboratories were requested to submit a single measurement result for each radionuclide analysed, decay corrected to the sampling date (24 March 2022). 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 the following additional information for each radionuclide for which results will 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.
(e) The detection limit and 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 in which a number of repetitions were analysed, 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 deriving from an average, information was requested on how the uncertainty was estimated (e.g., weighted average, maximum value etc.).

The IAEA recommended that samples were prepared using gravimetric measurements 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: STATISTICAL EVALUATION METHODOLOGY

The method used for the statistical evaluation of results reported in this ILC is 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 [34].

For results that could be intercompared quantitatively (i.e. for each radionuclide for which activity concentrations above detection limits were reported by at least two laboratories) a comparison reference value x_{ref} was determined as a power-moderated mean of the combined results [34]:

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

$$\frac{1}{u^2(x_{ref})} = \sum_{i=1}^{N} \left[\left(\sqrt{u_i^2 + s^2} \right)^{\alpha} S^{2-\alpha} \right]^{-1}$$

$$w_i = u^2(x_{ref}) \left[\left(\sqrt{u_i^2 + s^2} \right)^{\alpha} S^{2-\alpha} \right]^{-1}$$

where x_i is the result reported by laboratory i, $u(x_{ref})$ is the standard uncertainty of the reference value and w_i is the normalized weighting factor from the power-moderated mean equation. S is a typical uncertainty for the dataset under consideration, calculated from the variance associated with the arithmetic mean or the Mandel-Paule mean, whichever is higher, and s is a parameter used to increase the reported variances by a common amount and thus to moderate the weighting. α is a measure of the reliability of the uncertainties reported (0< α <2, $\alpha = 2 - 3/N$ by default); and N is the number of measurement values reported.

A ζ (zeta) score was then determined for each laboratory:

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

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. The standard uncertainty $u(d_i)$ was determined from the generally valid expression for any weighted mean:

$$u^2(d_i) = (1 - 2w_i)u_i^2 + u^2(x_{ref})$$

where u_i and $u(x_{ref})$ are the standard uncertainties associated with x_i and x_{ref}, respectively, and w_i is the normalized weighting factor from the power moderated mean equation.

This definition of the ζ score is valid when the reference value is calculated as a consensus of the results reported by the participating laboratories, as is the case for this ILC[2]. The uncertainty takes the correlation between individual results and the reference value into account.

This ζ score is interpreted as follows [35]:

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"; and

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 radionuclides submitted by other laboratories. A precision check can be 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. A reported uncertainty of greater than five times S_{50} was considered to be statistically irrelevant.

An example for the results reported for ^3H in this ILC is now provided. The reported results and derived parameters are presented in Table 10.

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[2] It is analogous to the ζ score for reference values calculated independently from the results reported by the participating laboratories (without weighting factors):

$$\zeta = \frac{y_i - y_{ref}}{\sqrt{u^2(x_i) + u^2(x_{ref})}}$$

and where $\zeta = 2E_n$, the E_n number calculated from the respective expanded uncertainties ($k = 2$) by:

$$E_n = \frac{y_i - y_{ref}}{\sqrt{U^2(x_i) + U^2(x_{ref})}}$$

TABLE 10. EXAMPLE RESULTS FOR ^{3}H

Laboratory	x_i (Bq/L)	u_i (Bq/L)	w_i	d_i	$u(d_i)$	ζ
IAEA	151543.6	1290.0	0.21	-789.5	3469.4	-0.23
IRSN	146000.0	9000.0	0.10	-6333.2	8704.2	-0.73
KINS	160366.2	2724.6	0.20	8033.0	3950.0	*2.03*
LANL	145800.0	1500.0	0.21	-6533.2	3518.7	-1.86
LS	165778.0	7216.0	0.12	13444.8	7097.8	1.89
TEPCO	145700.0	5100.5	0.16	-6633.2	5386.5	-1.23

x_{ref}	$u(x_{ref})$	S_{50}
152333.2	3329.3	5821.5

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, the "PomPlot" [36, 37], an intuitive graphical method, also used by the BIPM's CCRI(II) for Key Comparisons, providing an 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. The results for the ^{3}H example from this ILC are shown in Figure 16.
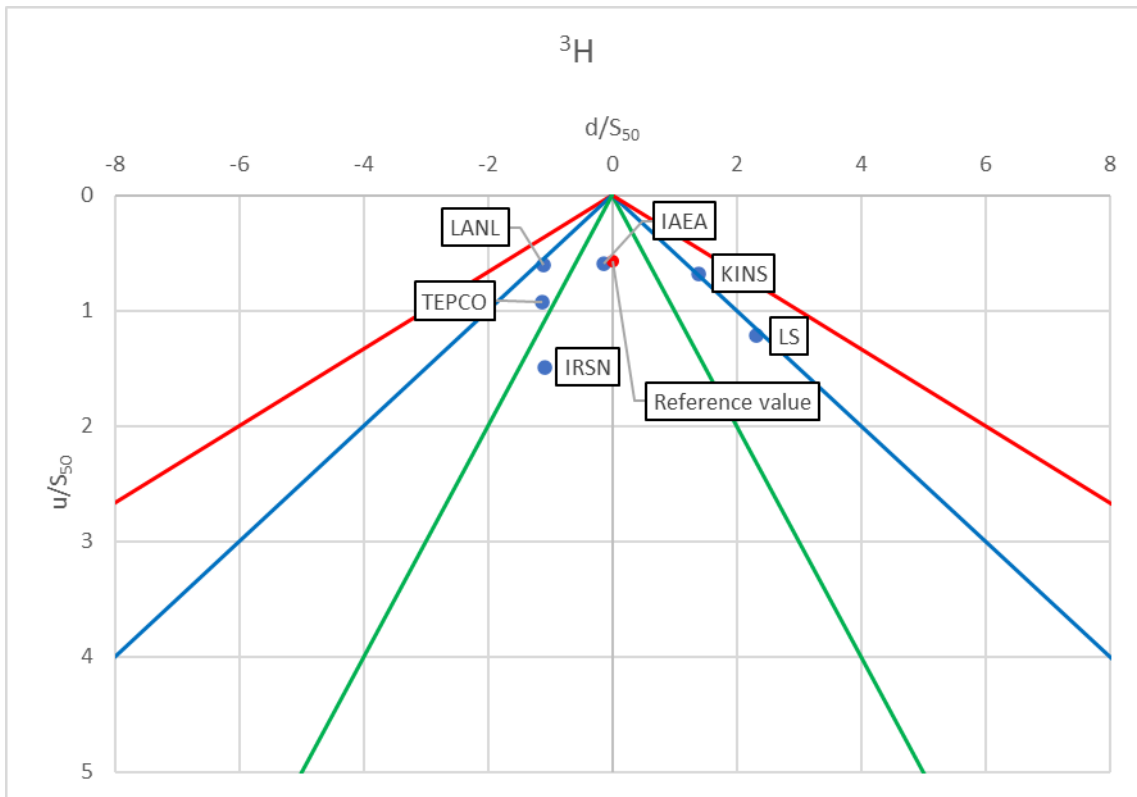
FIG. 16. Example PomPlot for ^{3}H results.
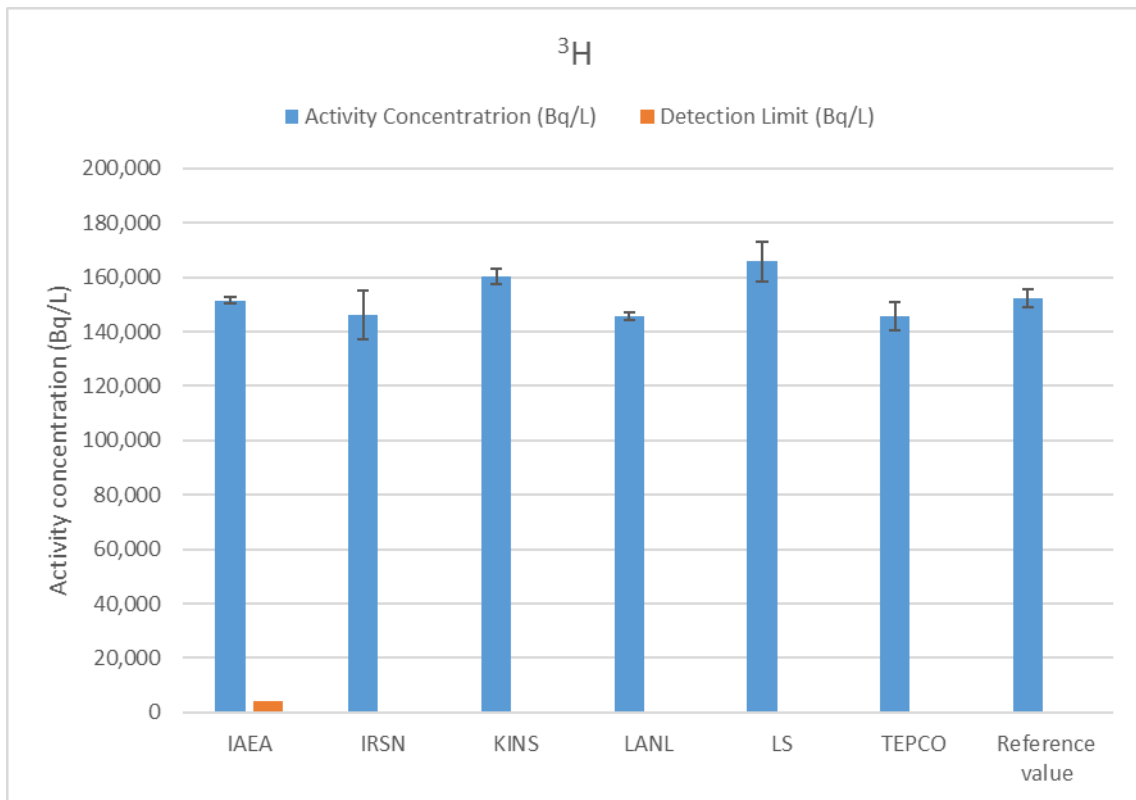
FIG. 17. Activity concentrations and detection limits for ^3H.
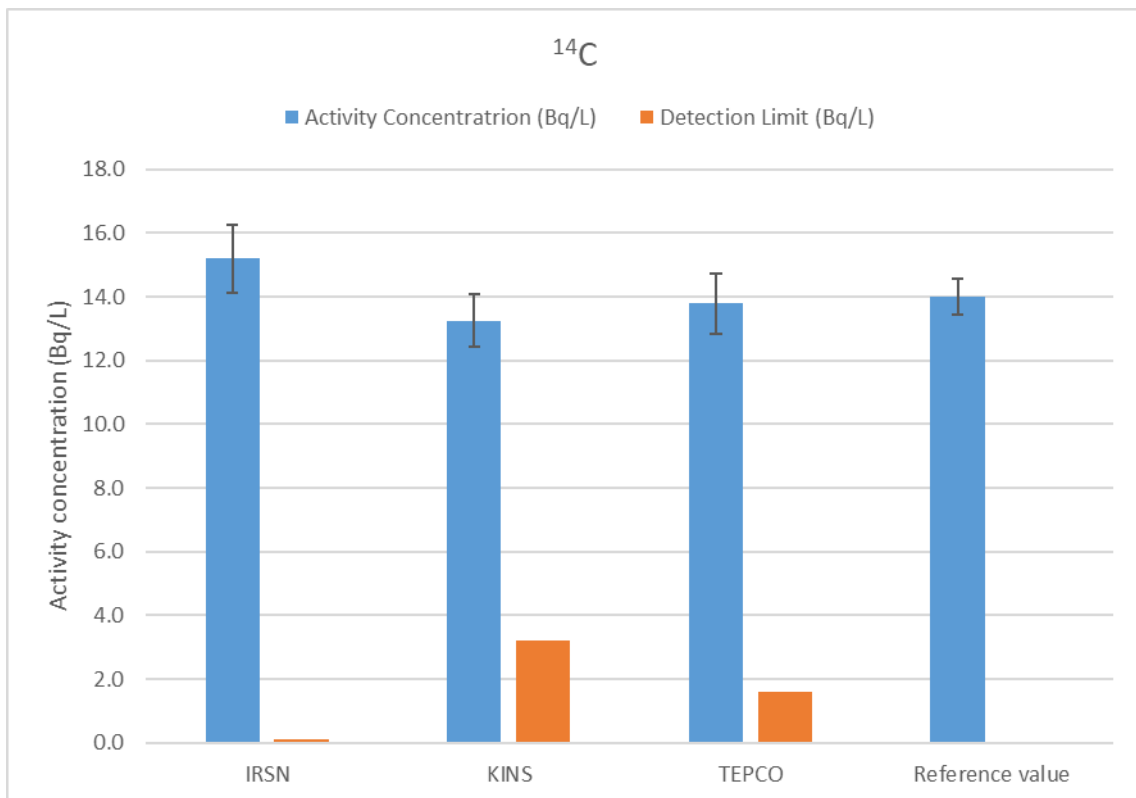
FIG. 18. Activity concentrations and detection limits for ^{14}C.
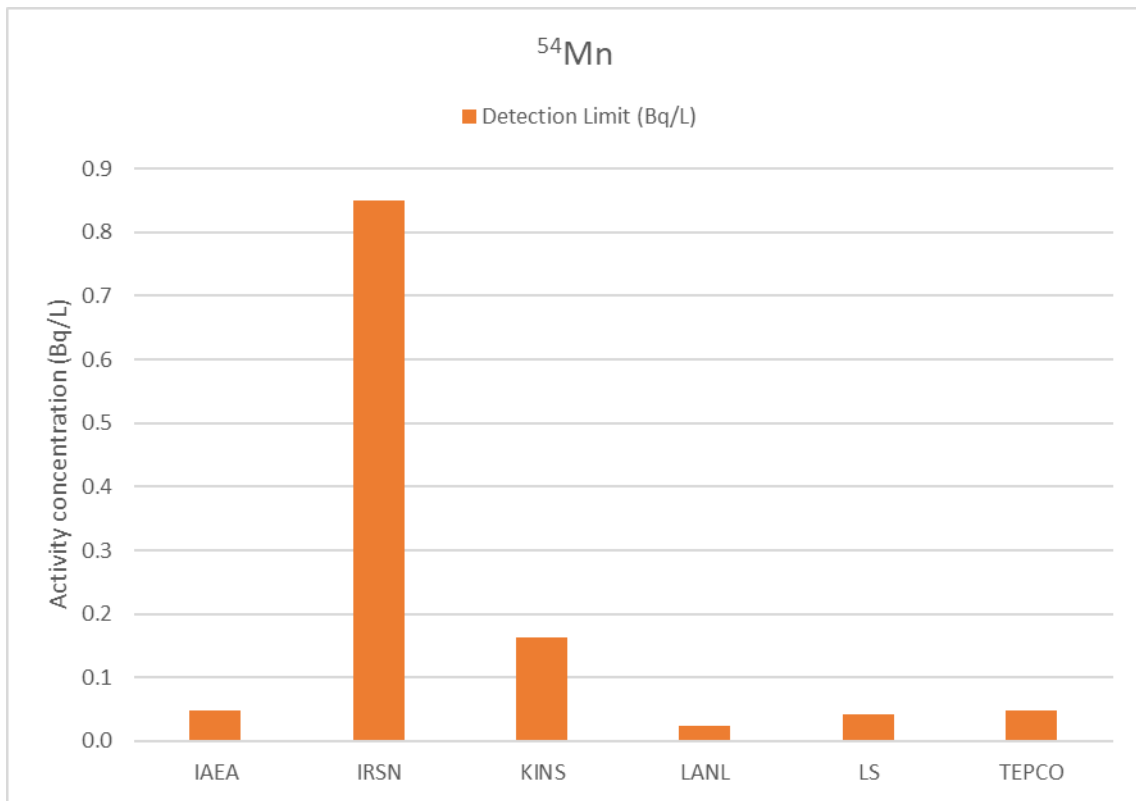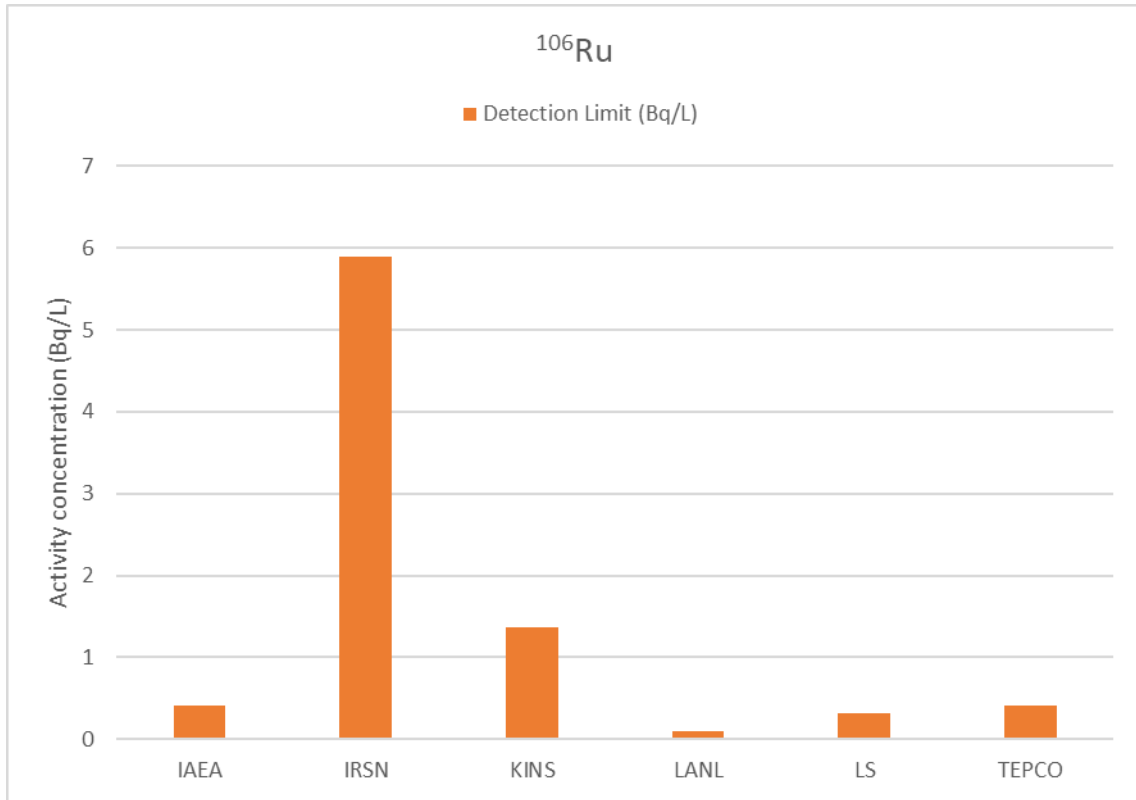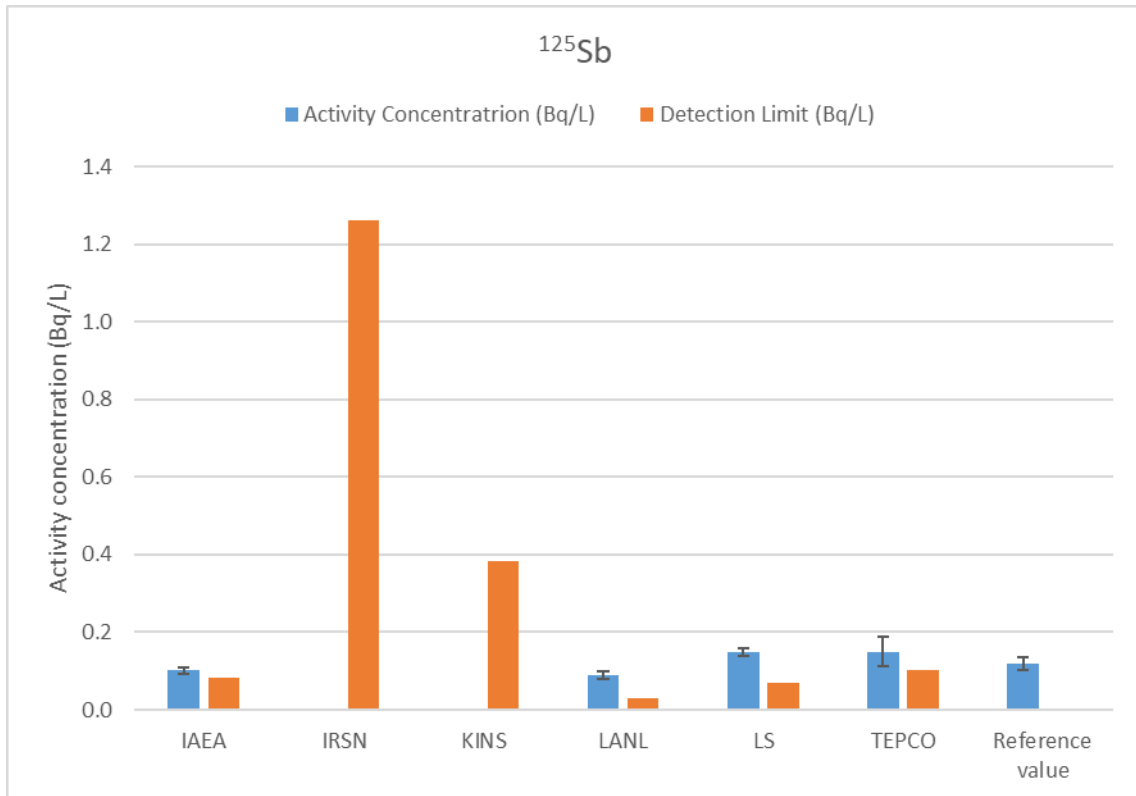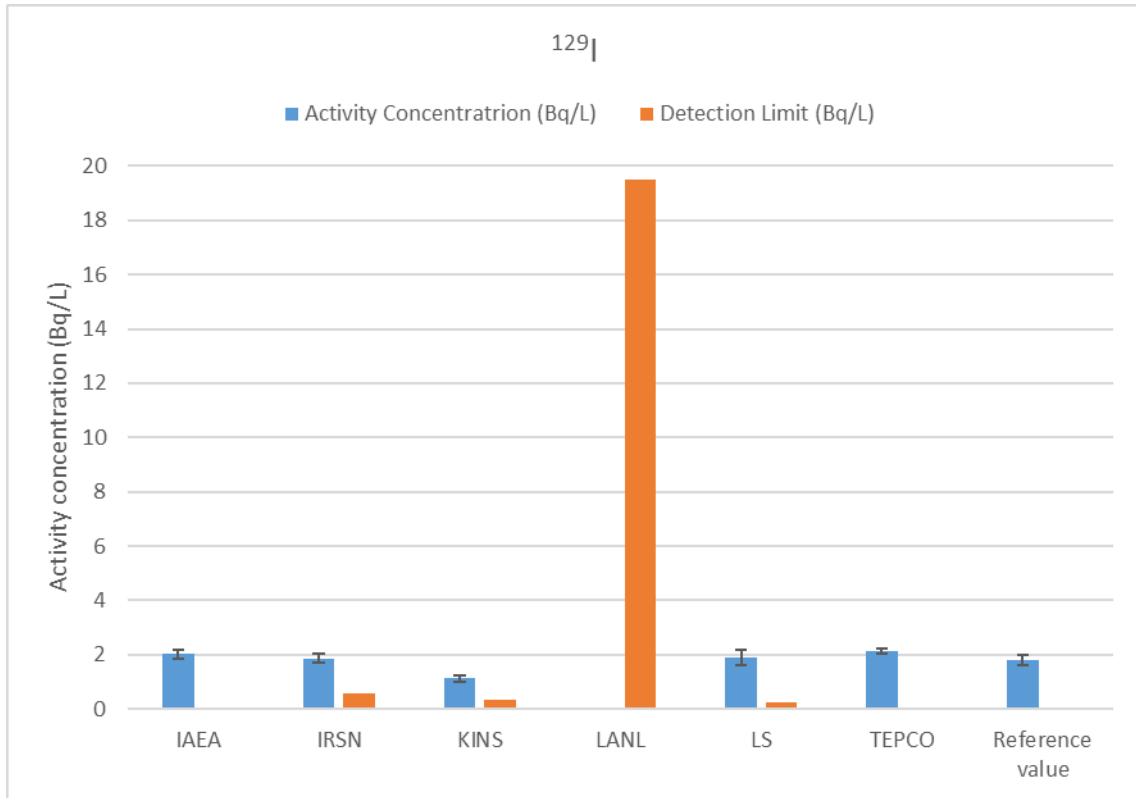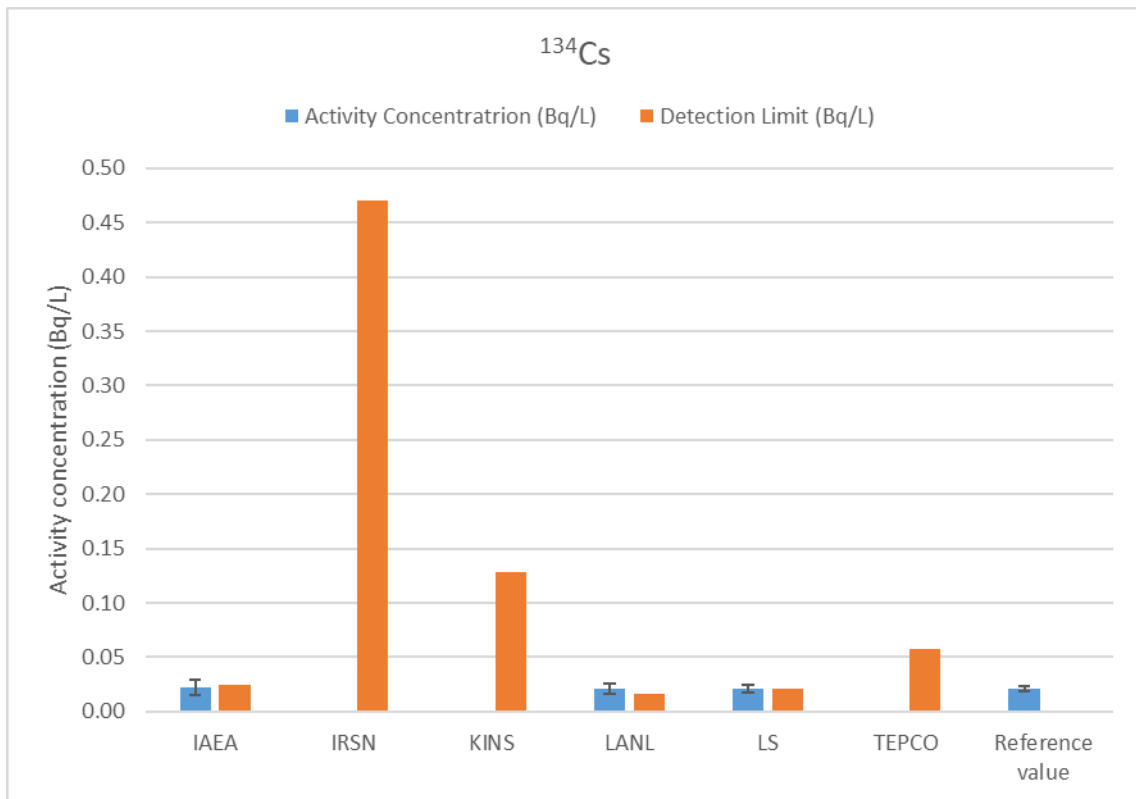
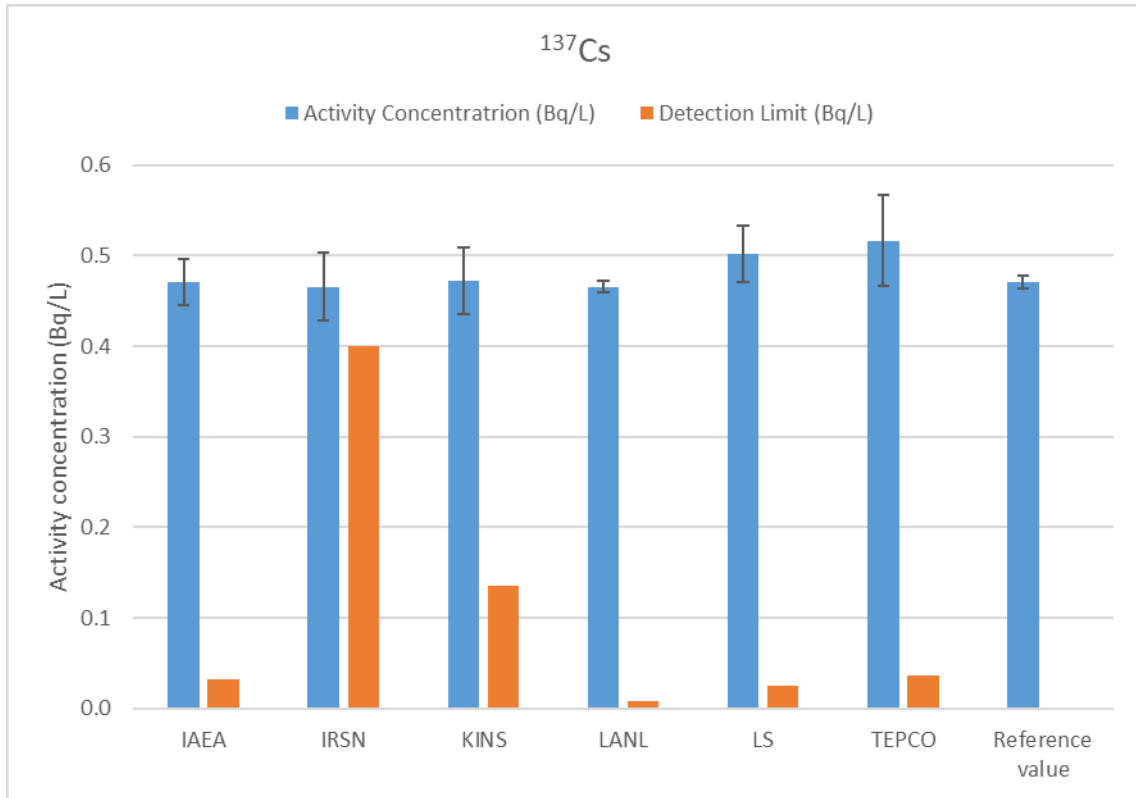*FIG. 19. Detection limits for ⁵⁴Mn.*
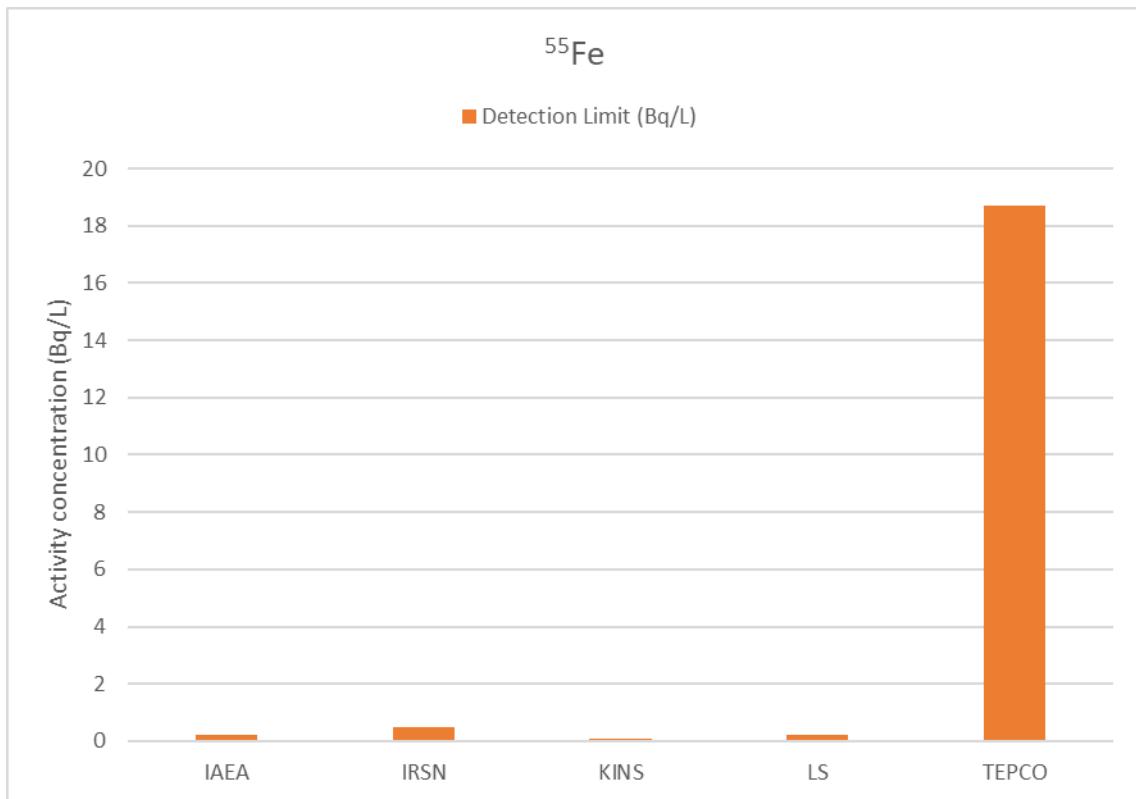
FIG. 20. Detection limits for ⁵⁵Fe.
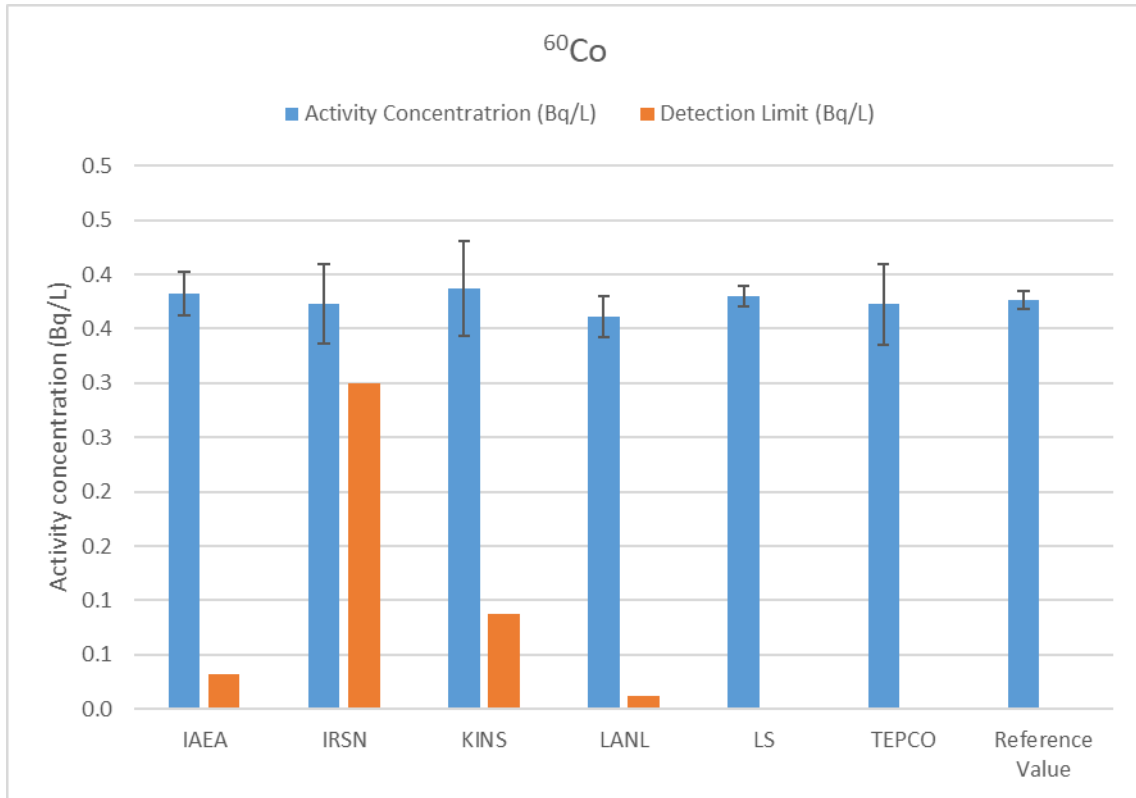
FIG. 21. Activity concentrations and detection limits for ^{60}Co.
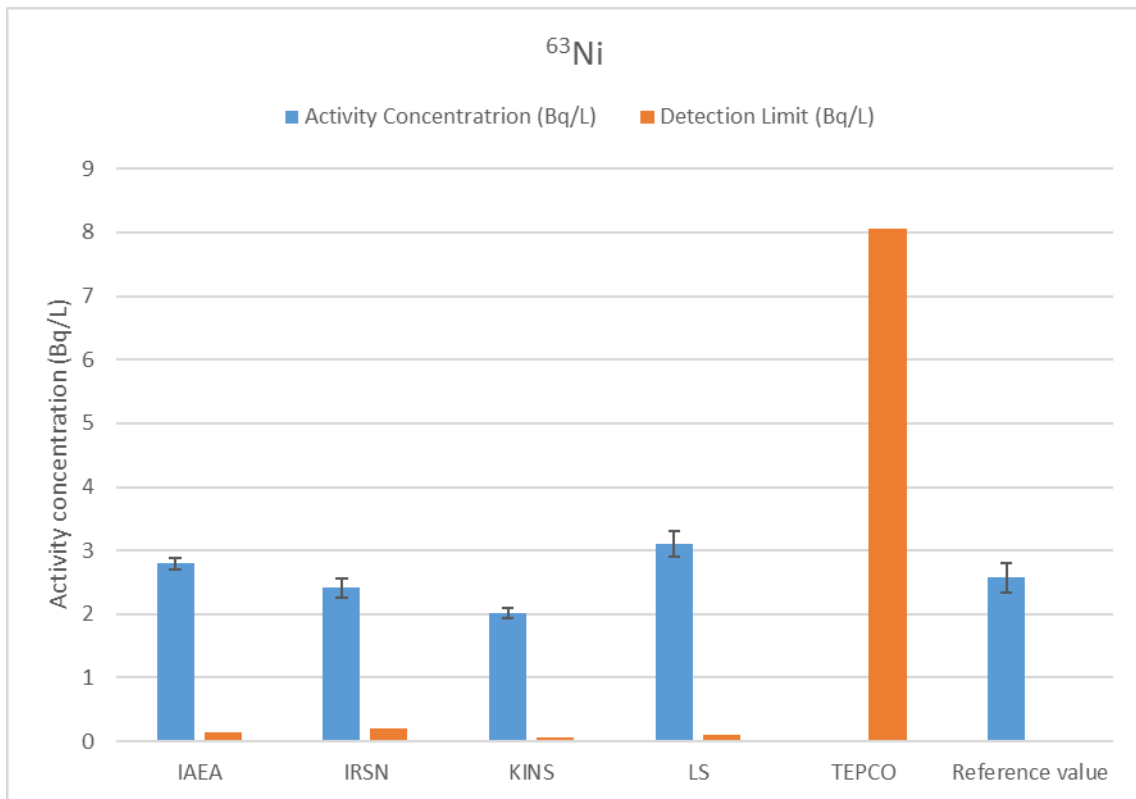
FIG. 22. Activity concentrations and detection limits for ^{63}Ni.
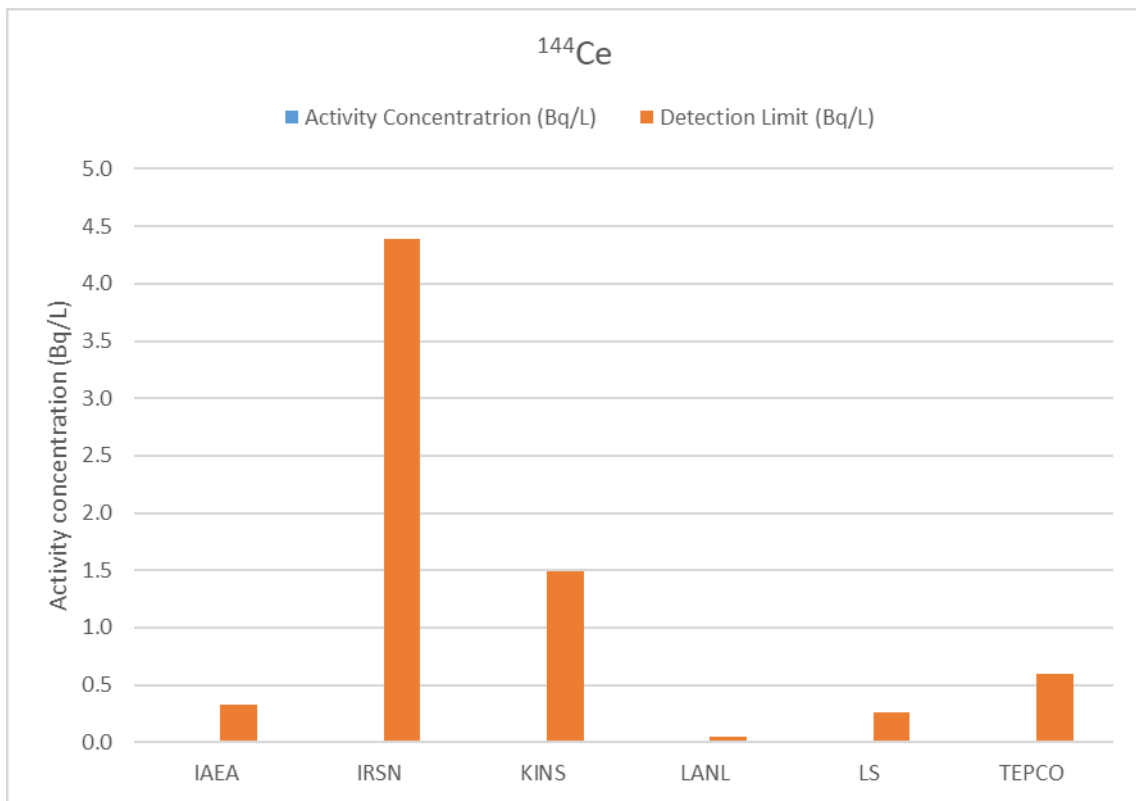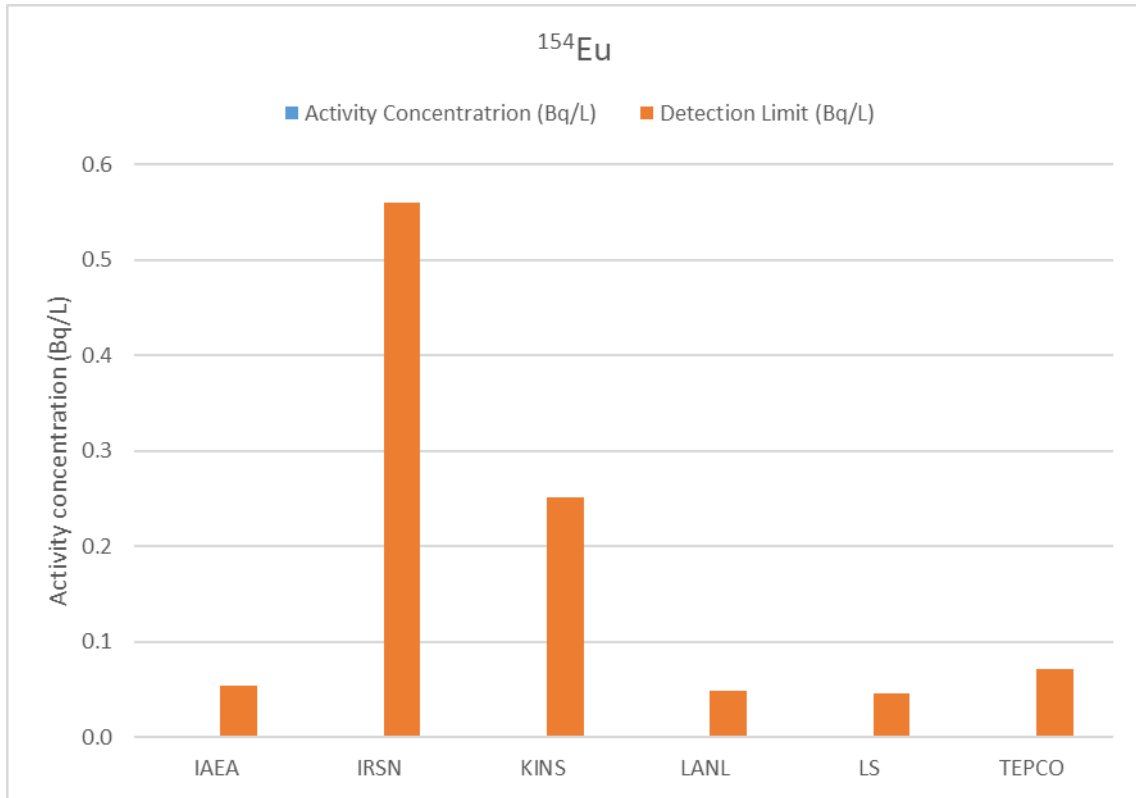
44
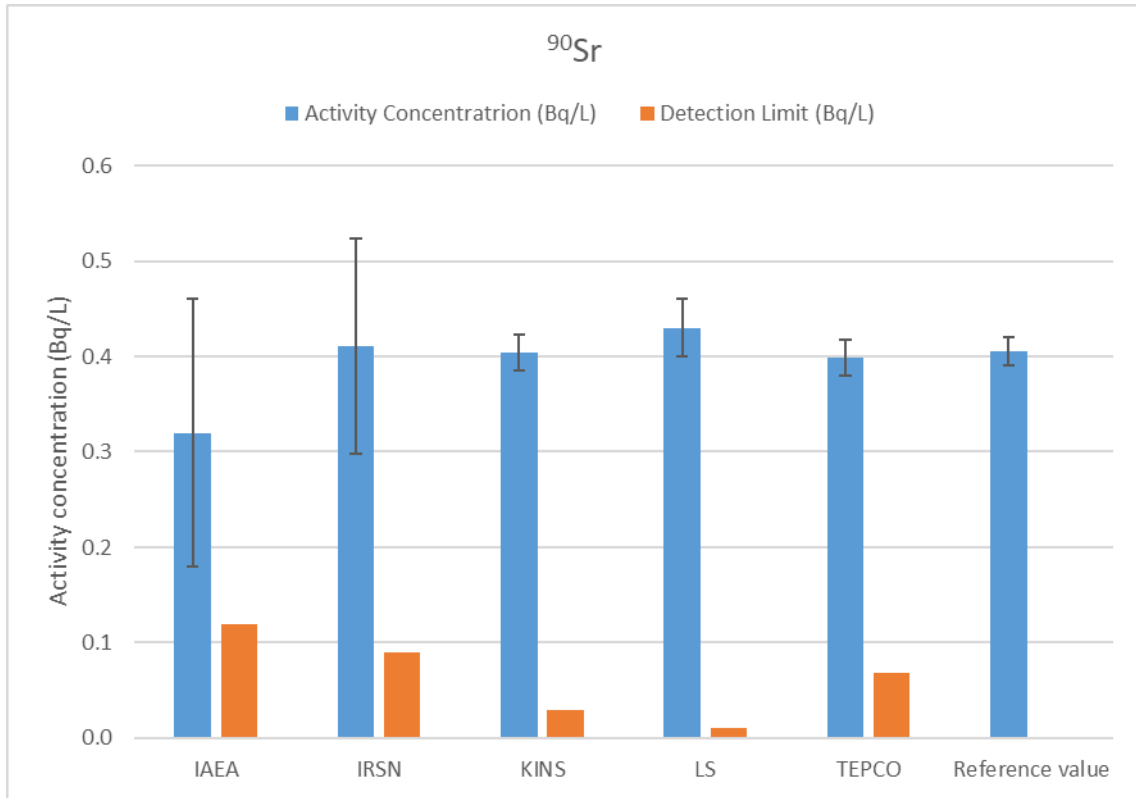
FIG. 23. Activity concentrations and detection limits for ^{90}Sr.
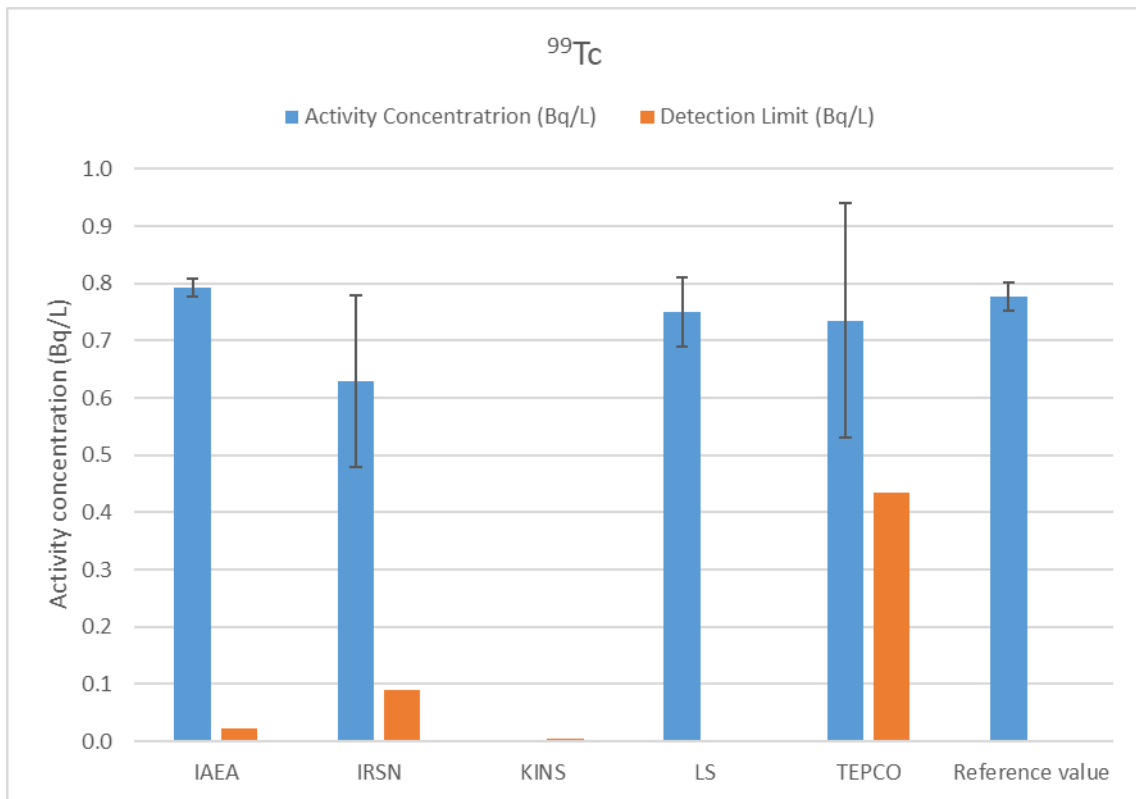
FIG. 24. Activity concentrations and detection limits for ^{99}Tc.

FIG. 25. Detection limits for ^{106}Ru.

FIG. 26. Activity concentrations and detection limits for ^{125}Sb.

FIG. 27. Activity concentrations and detection limits for ^{129}I.

FIG. 28. Activity concentrations and detection limits for ^{134}Cs.

FIG. 29. Activity concentrations and detection limits for ^{137}Cs.

FIG. 30. Detection limits for ^{144}Ce.

FIG. 31. Detection limits for ^{154}Eu.
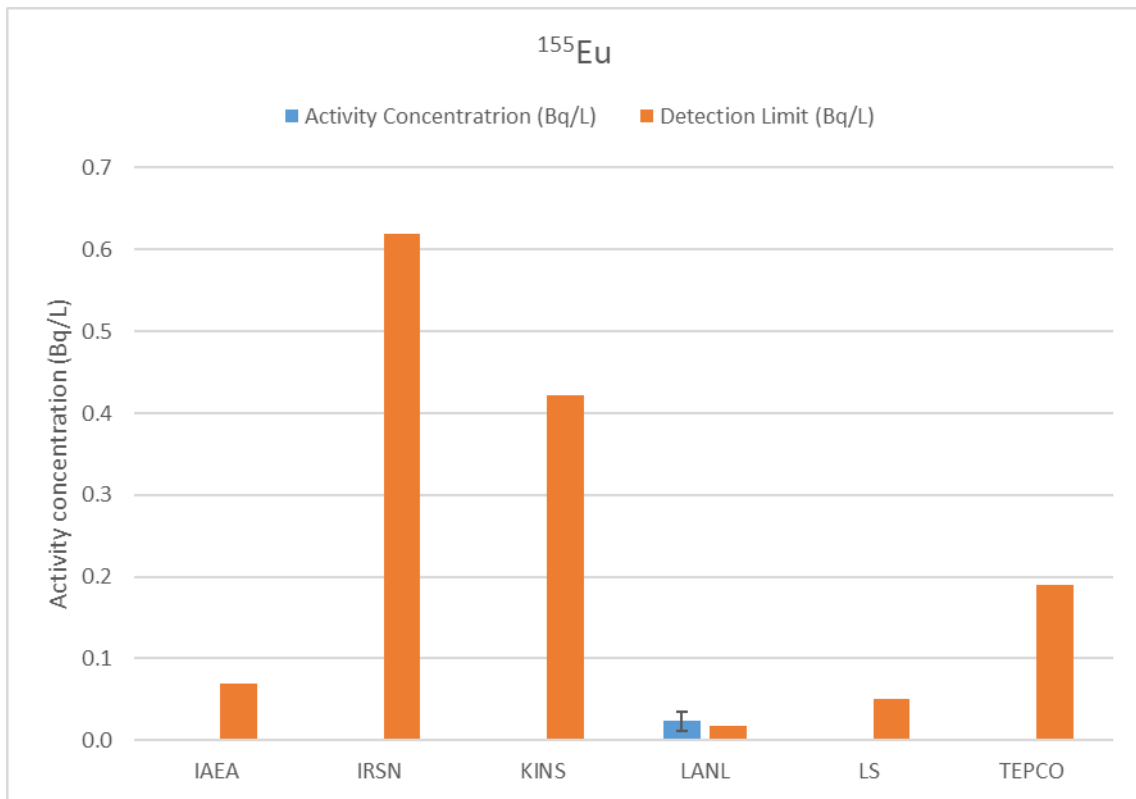
FIG. 32. Activity concentrations and detection limits for ^{155}Eu.
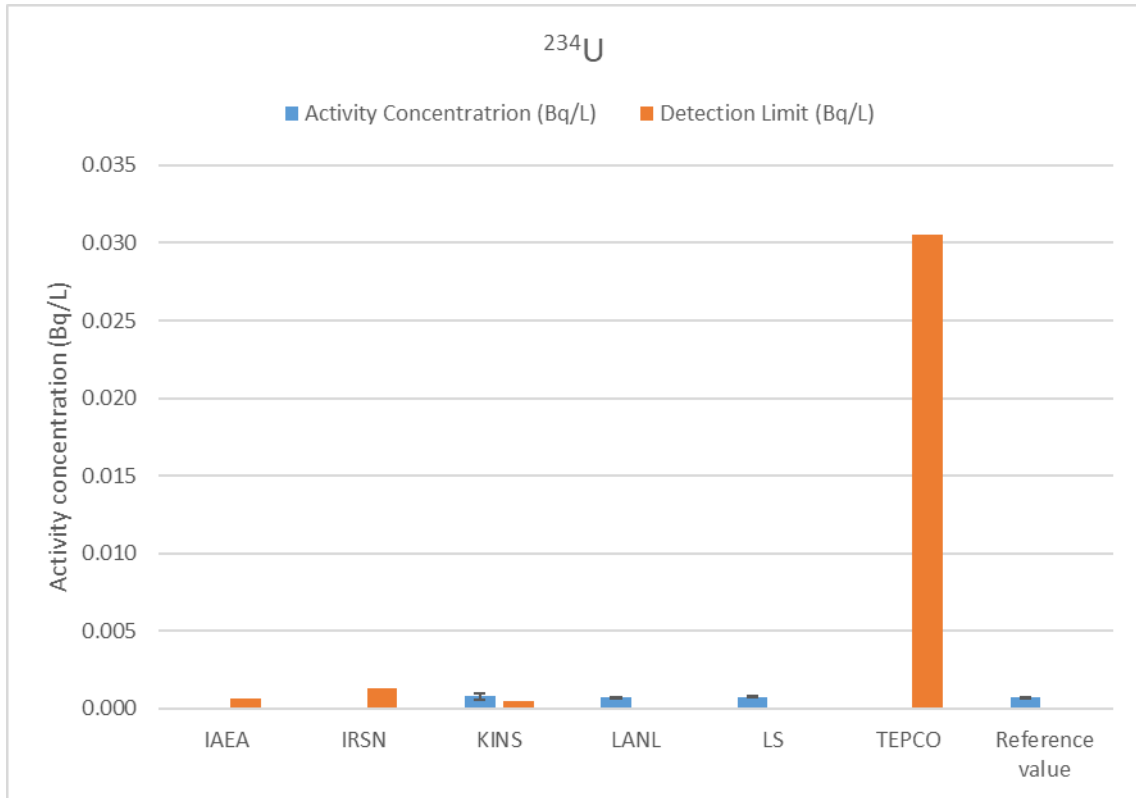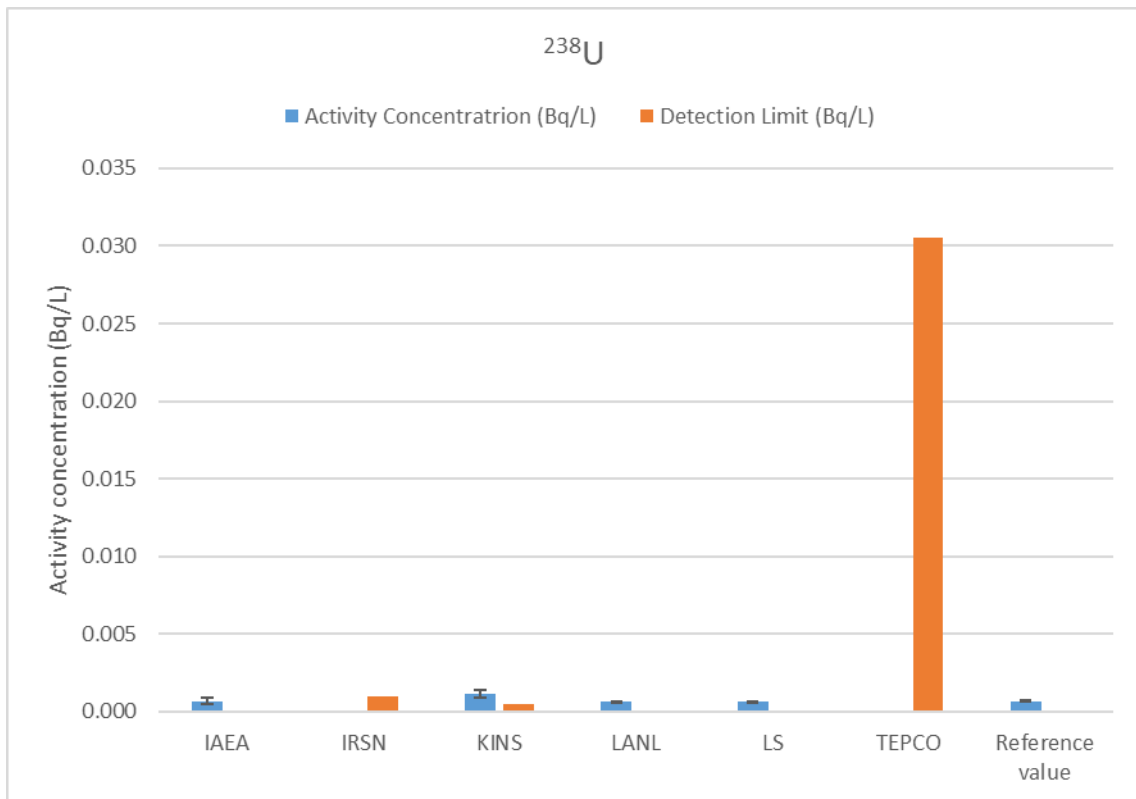
FIG. 33. Activity concentrations and detection limits for ^{234}U.

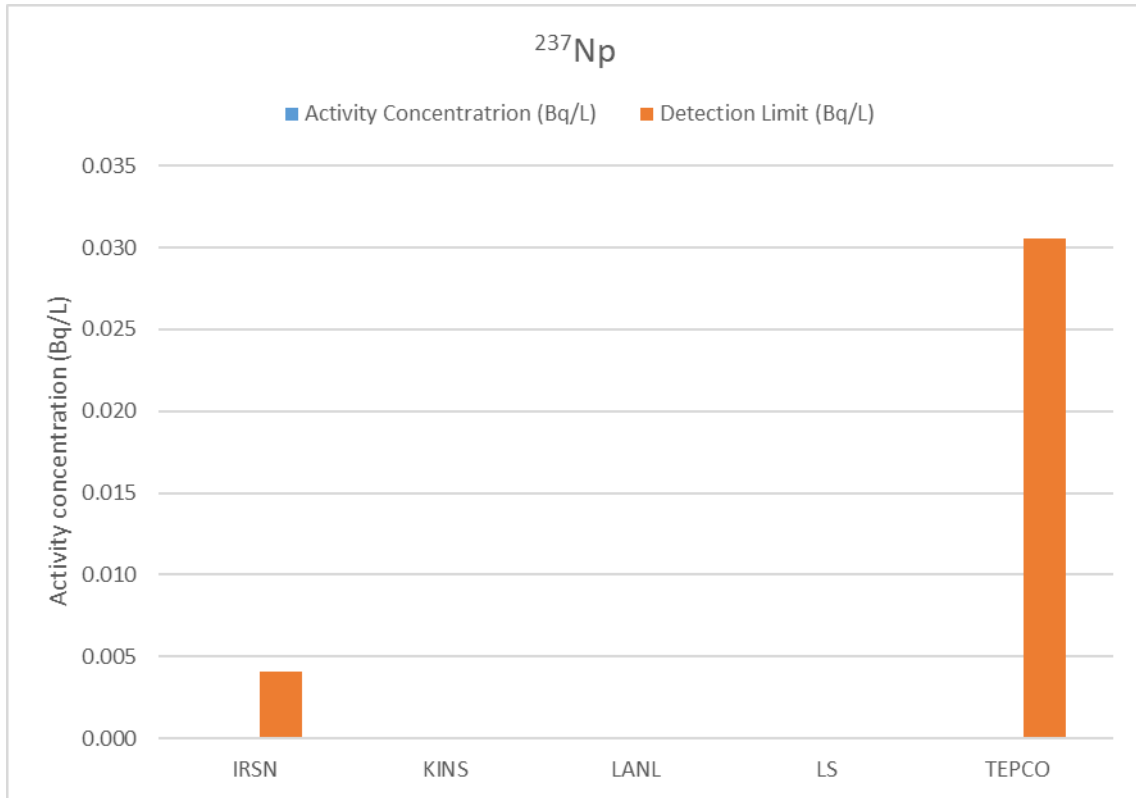*FIG. 34. Activity concentrations and detection limits for ^{238}U.*

50

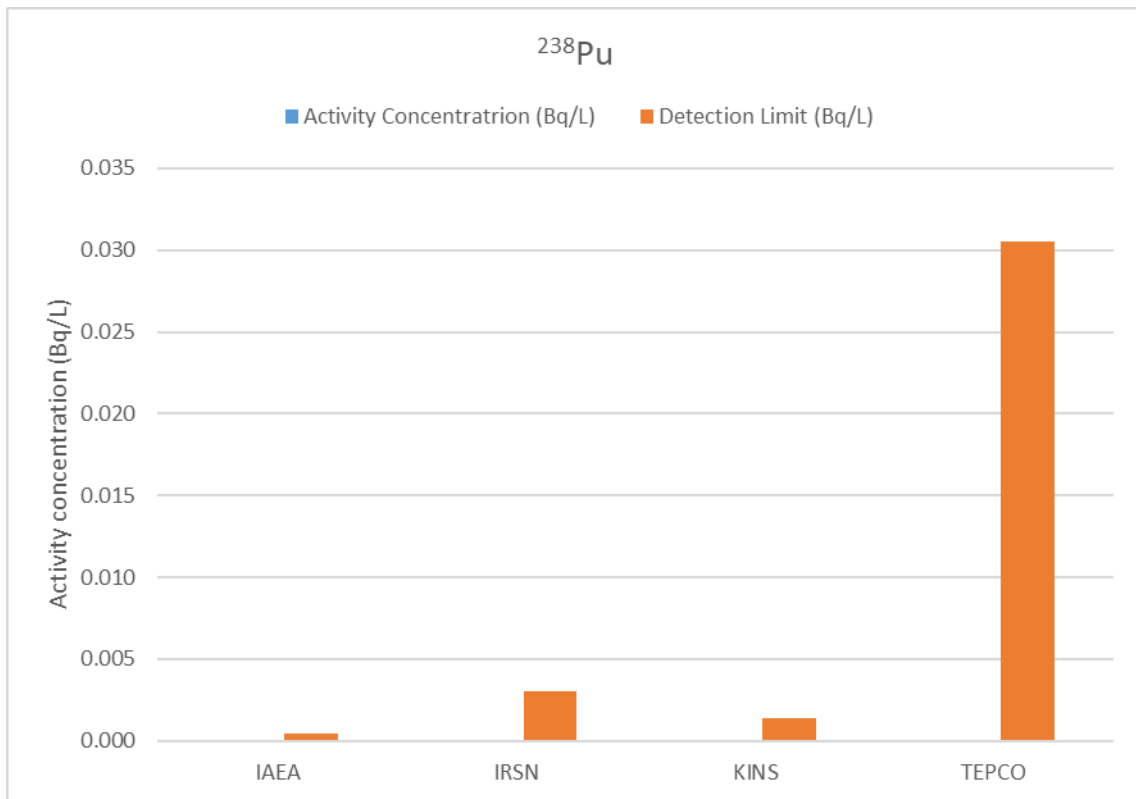*FIG. 35. Activity concentrations and detection limits for ^{237}Np.*
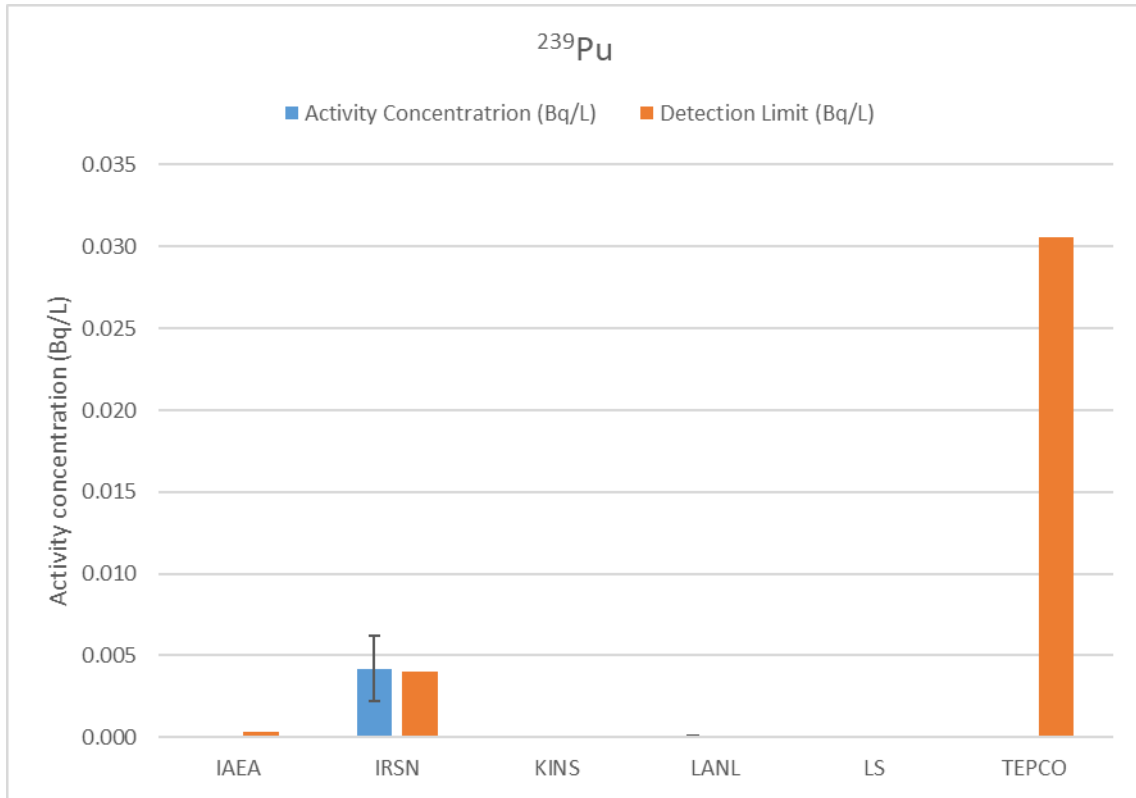

FIG. 36. Detection limits for ^{238}Pu.

FIG. 37. Activity concentrations and detection limits for ^{239}Pu. (A combined value for ^{239,240}Pu was reported by IAEA and IRSN.)
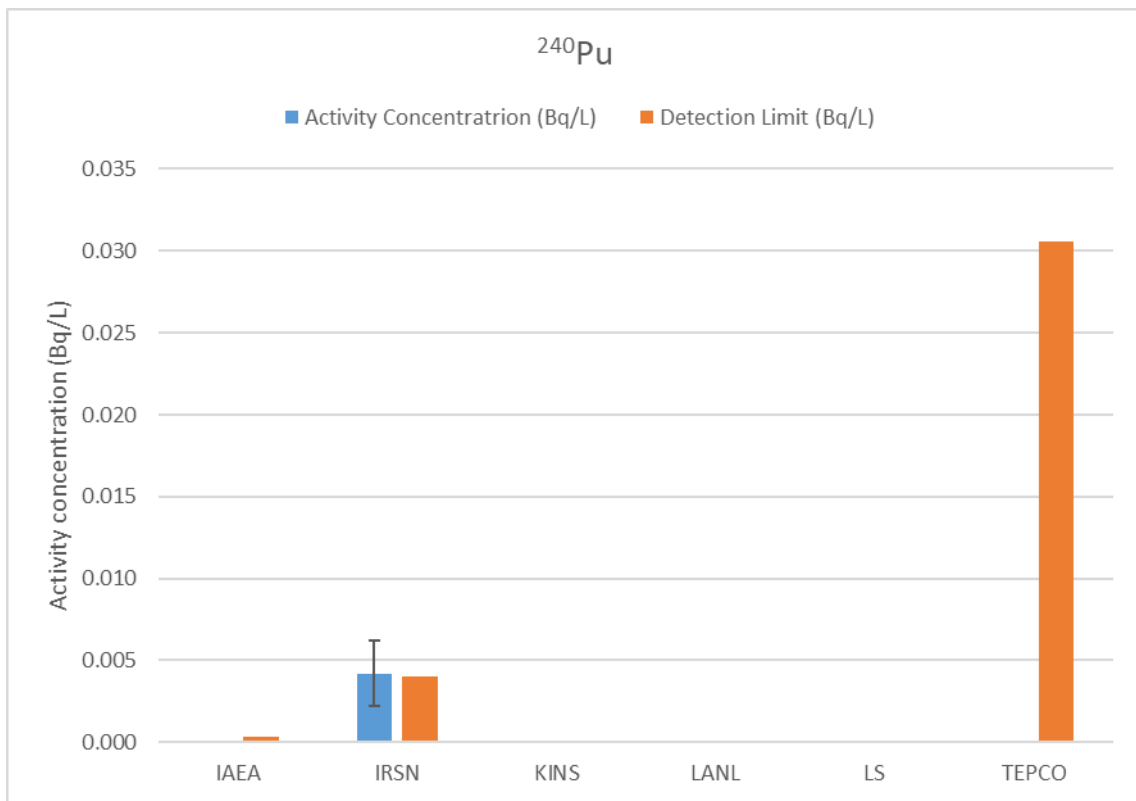

FIG. 38. Activity concentrations and detection limits for ^{240}Pu. (A combined value for ^{239,240}Pu was reported by IAEA and IRSN.)

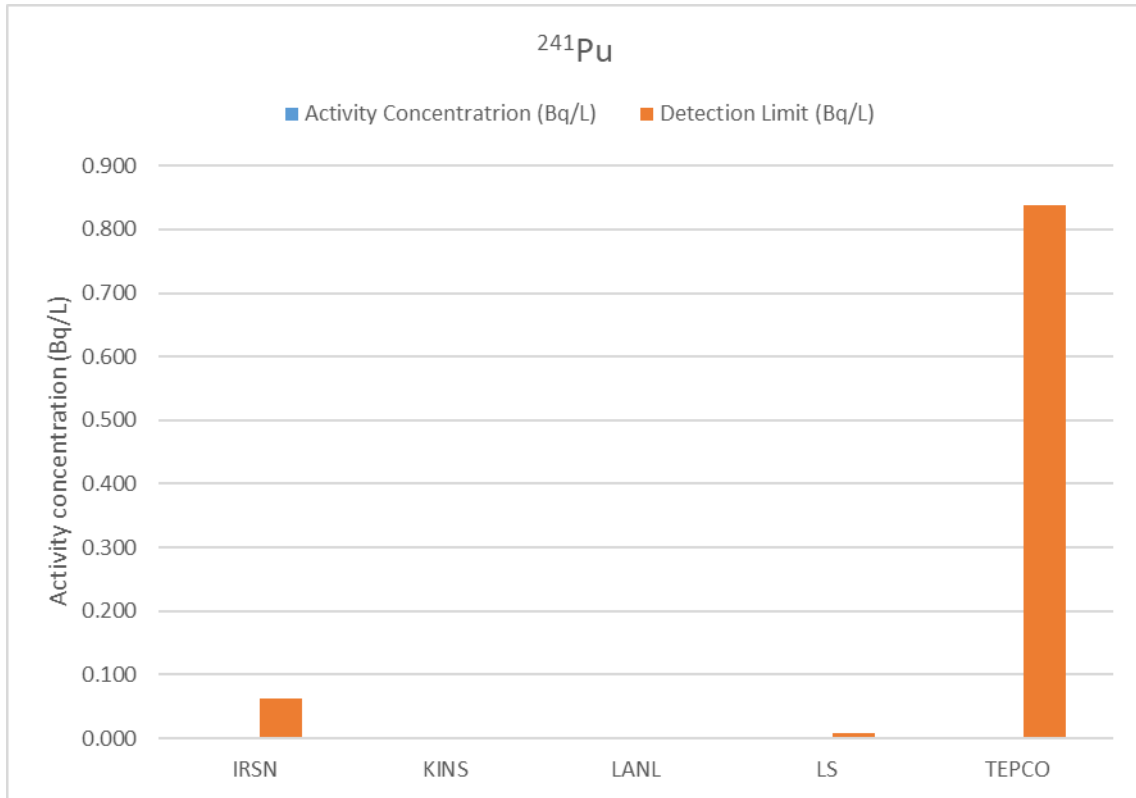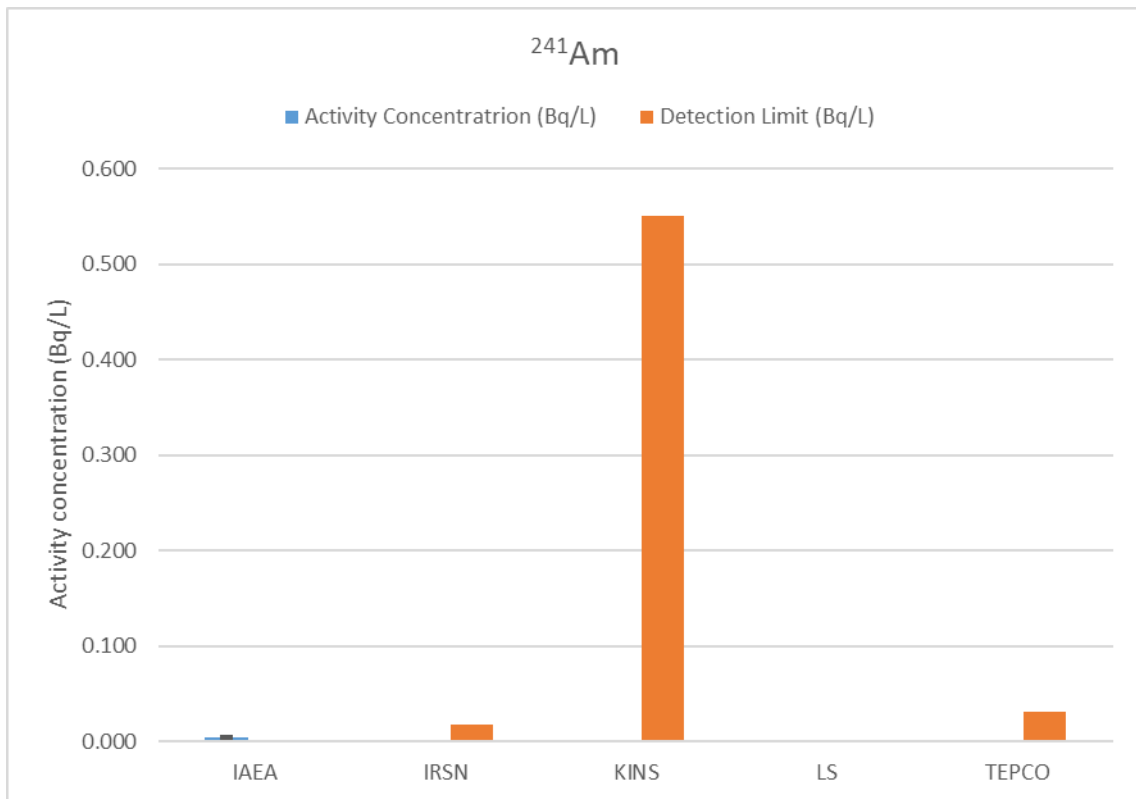*FIG. 39. Activity concentrations and detection limits for ^{241}Pu.*

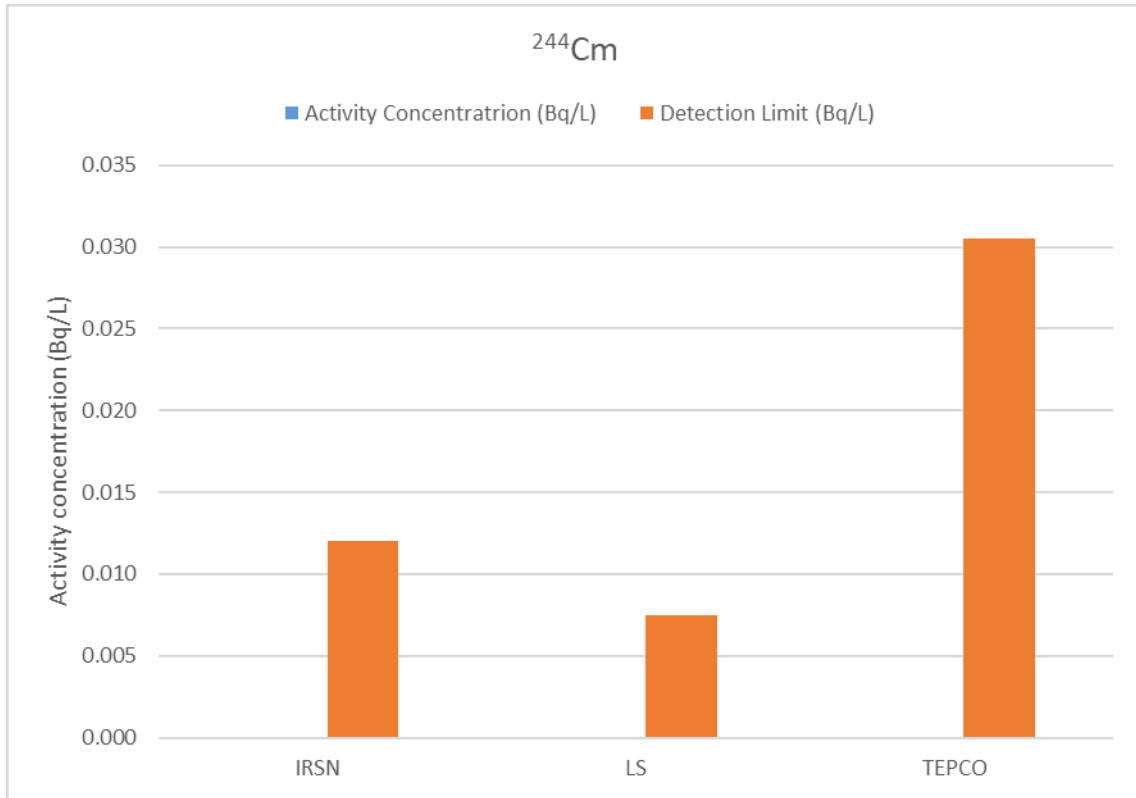*FIG. 40. Activity concentrations and detection limits for ^{241}Am.*

FIG. 41. Activity concentrations and detection limits for ^{244}Cm.

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CONTRIBUTORS TO DRAFTING AND REVIEW

Abraham-Ponti, C.	International Atomic Energy Agency
Bartocci, J.	International Atomic Energy Agency
Blinova, O.	International Atomic Energy Agency
Cook, M.	International Atomic Energy Agency
Copia, L.	International Atomic Energy Agency
Dioszeghy, A.	International Atomic Energy Agency
Fujak, M.	International Atomic Energy Agency
Groening, M.	International Atomic Energy Agency
Horsky, M.	International Atomic Energy Agency
Le Normand, J.	International Atomic Energy Agency
Levy, I.	International Atomic Energy Agency
Massinger, B.	International Atomic Energy Agency
McGinnity, P.	International Atomic Energy Agency
Murphy, N.	International Atomic Energy Agency
Nadalut, B.	International Atomic Energy Agency
Osvath, I.	International Atomic Energy Agency
Patterson, S.	International Atomic Energy Agency
Pham, M. K.	International Atomic Energy Agency
Pommé, S.	European Commission Joint Research Centre, Geel, Belgium
Rovan, L.	International Atomic Energy Agency
Seel, P.J.	International Atomic Energy Agency
Seslak, B.	International Atomic Energy Agency
Sobiech-Matura, K.	International Atomic Energy Agency
Titiano, M.J.	International Atomic Energy Agency
Tucakovic, I.	International Atomic Energy Agency
Ulanowski, A.	International Atomic Energy Agency

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA