Meeting Material 4-3 of 107th Committee on Oversight
and Evaluation of Specified Nuclear Facilities
Tentative Translation by NRA

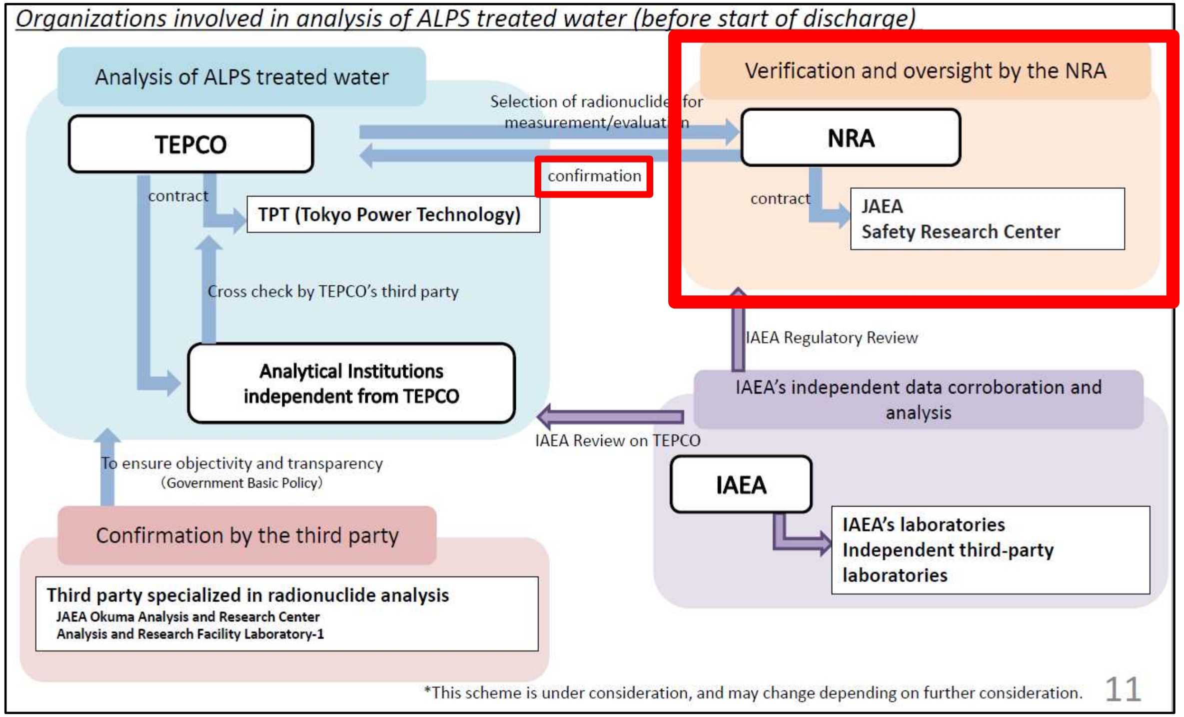
Analysis result of ALPS treated water

14th April 2023

JAEA
Nuclear Safety Research Center (NSRC)

Purpose

- The NRA is inspecting TEPCO's organizational framework for analyzing "nuclides to be measured and evaluated" and their quality assurance activities, and checking whether they are following the approved Implementation Plan.
- Also, considering the statement in the Governmental Policy "monitoring with objectivity and transparency", the NRA confirms the validity of TEPCO's analysis by conducting independent monitoring.
- JAEA NSRC analyzed radionuclides in ALPS treated water under the contract from the NRA.



"Update Material" explained in 2nd IAEA review mission for NRA

Compare the analytical results (radionuclide concentration) by JAEA NSRC and TEPCO with consideration of uncertainty ranges, for supporting the overall oversight by the NRA.

Sampling

Observation by IAEA review team of the sampling of ALPS treated water at the TEPCO Fukushima Daiichi Nuclear Power Station (March 24)

<Reference material>
March 25, 2022
Tokyo Electric Power Company Holdings, Inc.
Fukushima Daiichi D&D Engineering Company

○ On March 24, the IAEA review team in Japan visited the Fukushima Daiichi Nuclear Power Station and observed the sampling of ALPS treated water from the measurement/confirmation facility.

○ This is the second time that the IAEA has observed sampling this year (1st time: February).

○ A portion of the sampled water shall be analyzed at an IAEA research facility in order to verify TEPCO's analysis method and results.

Observing sampling

Sampling ALPS treated water

Photographed by TEPCO Holdings, Inc.
on March 24, 2022

Received at the hot laboratory of JAEA on 12th October 2022

TEPCO's news release material:
https://www.tepco.co.jp/en/hd/decommission/information/newsrelease/reference/pdf/2022/reference_20220325_01-e.pdf

Target nuclides

➢ Analyzed 13 radionuclides contracted from NRA

- Nuclides mainly detected in ALPS treated water (to compare with TEPCO's result) :

  Co-60, Sr-90, Ru-106, Sb125, I-129, Cs-134, Cs-137, H-3, C-14, Tc-99

- Nuclides rarely detected in ALPS treated water (confirmation of existence) :

  Cl-36, Fe-55, Se-79

1. Overview TEPCO

- For the 35 nuclides of the nuclides to be measured/assessed and monitored, the analytical results at the ALPS inlet (FY 2021) and ALPS outlet (K4, J1-C, J1-G) are reported based on the results of checking the sum of the ratios to regulatory concentrations limits in the classification in the table below. Note that in the calculation of the regulatory concentration limit ratio of α-nuclides, the total-α value is divided by 4 Bq/L, which is the lowest regulatory concentration limit among the α-nuclides selected.

Classification					Specific nuclides	ALPS inlet	ALPS outlet K-4	ALPS outlet J1-C	ALPS outlet J1-G
Nuclides mainly detected in ALPS treated water					7 major nuclides including radioactive equilibrium Y-90, Te-125 m) C-14, Tc-99	1.7E+03	2.7E-01	1.6E-01	5.8E-02
Nuclides rarely detected in ALPS treated water	α				U-234, U-238, Np-237, Pu-238, Pu-239, Pu-240, Am-241, Cm-244	5.4E+00 →1.0E+00	8.2E-04 →1.6E-04	4.2E-02 →8.1E-03	3.7E-02 →7.0E-3
	Other than α nuclides	Subject to removal by ALPS (other than the above)			Mn-54, Ni-63, Cd-113m, Ce-144, Pm-147, Sm-151, Eu-154, Eu-155, Pu-241	2.2E+00	1.4E-03	1.3E-02	1.2E-02
		Other than those subject to removal	A large number of measurements		Cl-36, Se-79, Nb-94	5.0E-02	1.2E-02	1.2E-02	1.2E-02
			Small number of measurements	[1] Countable for gross β or Ge	Ba-133	8.7E-03	1.5E-03 →1.8E-05	1.4E-03 →1.4E-04	1.4E-03 →1.3E-04
				[2] Not countable for gross β and Ge	Fe-55, Nb-93m, Mo-93	2.1E-02	9.3E-03	6.8E-03	6.8E-03

*For J1-C and J1-G, the analysis and evaluation results for Cl-36, Se-79, ~~Ba-133,~~ Fe-55, Nb-93 m, and Mo-93 are not available, and the results from the additional ALPS outlet are used.

The Japanese version shall prevail.

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Meeting Material 1-1-2 of 3rd Technical Meeting on Specified Nuclear Facility
https://www.tepco.co.jp/en/hd/decommission/information/committee/pdf/2022/technical_22122102-e.pdf

Analytical methods（1/2）

Nuclides	Principal radiation emitted	Analytical equipment	Analytical method （pretreatment）	Basis for Analytical Method
Co-60	βγ	Ge	without pretreatment	The Series of Environmental Radioactivity Measuring Methods (SERMM) No.7
Sr-90	β	LSC	Sr was purified by using Sr resin. β -Ray was measured after reaching radioactive equilibrium between ^{90}Sr and ^{90}Y.	JAEA-Technology 2009-051
Ru-106	β	Ge (Measure Rh-106)	without pretreatment	SERMM No.7
Sb-125	βγ	Ge	without pretreatment	SERMM No.7
I-129	βγ	ICP-MS	I was purified with Anion-SR	SERMM No.32
Cs-134	βγ	Ge	without pretreatment	SERMM No.7
Cs-137	βγ	Ge	without pretreatment	SERMM No.7

Ge：Ge Semiconductor Detector
LSC：Liquid Scintillation Counter
ICP-MS：Inductively Coupled Plasma Mass Spectrometry

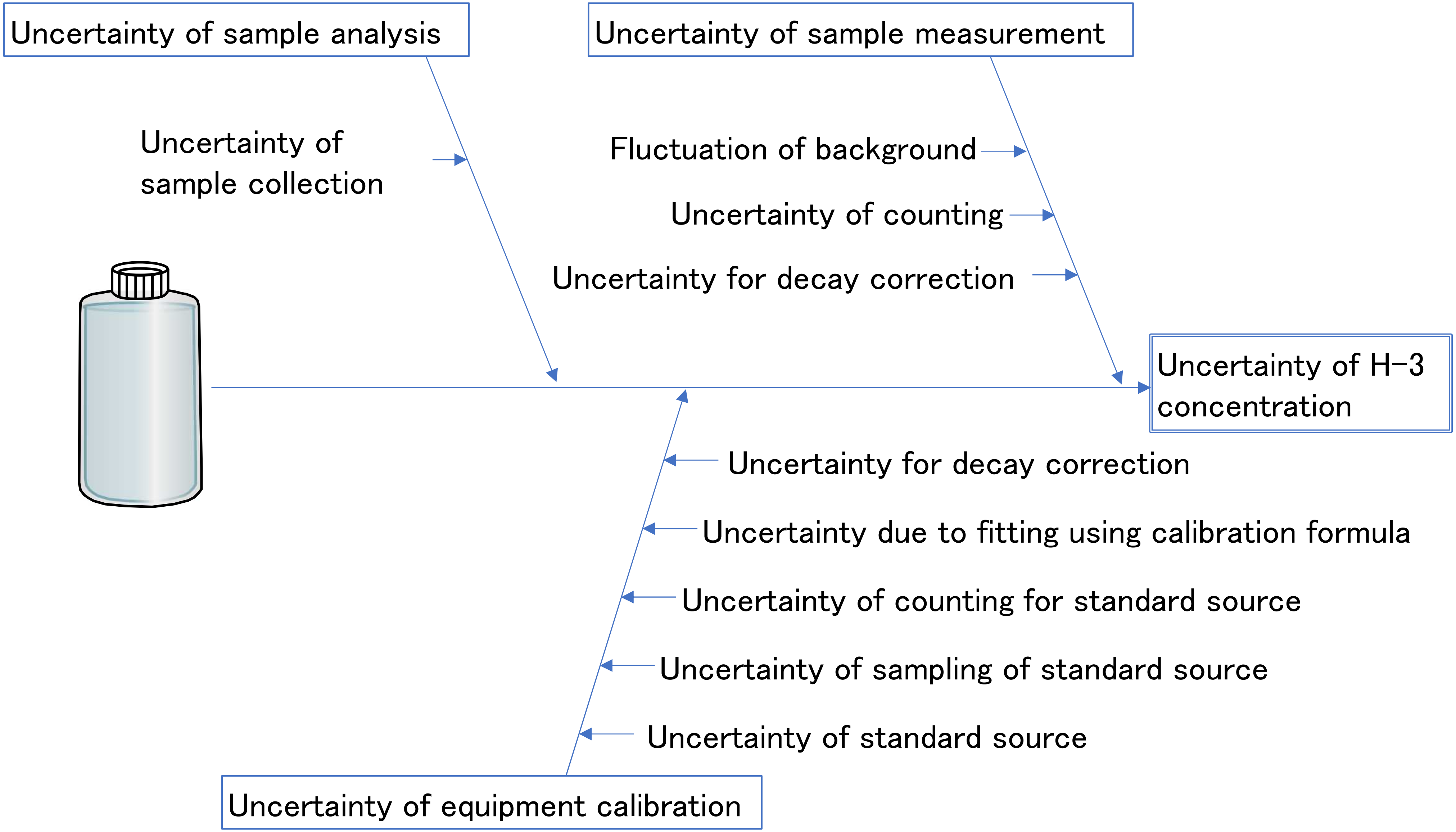
Analytical methods（2/2）

Nuclides	Principal radiation emitted	Analytical equipment	Analytical method （pretreatment）	Basis for Analytical Method
H-3	β	LSC	Sample solution was purified by distillation, and then, mixed with scintillator.	SERMM No.9
C-14	β	LSC	1.5 M HNO_3 was added into sample solution and N_2 gas was insufflated to the solution to evaporate CO_2. CO_2 was tapped by absorbent and absorbent was mixed with scintillator.	JAEA-Technology 2009-051
Tc-99	β	ICP-MS	Tc was separated by TEVA resin	S. Uchida et al.,[*1] and K. Tagami[*2]
Cl-36	β	GFC	Cl was separated by AgCl precipitation. After dissolution of AgCl, I was eliminated by SDB-XD Disk. Finally, Cl was recovered by AgCl precipitation.	JAEA-Technology 2009-051
Fe-55	EC	Ge	Fe was recovered by coprecipitation of iron hydroxide and calcium oxalate.	H. Ichige et al.,[*3]
Se-79	β	LSC	Cs was eliminated by AMP. Co, Ni, Ca, Sr and Ra were eliminated by carbonate precipitation. Al, Y, Th, U were eliminated by iron hydroxide precipitation, and Tc was eliminated TEVA resin. Se in the treated sample solution was precipitated by reduction. Precipitated Se was dissolved by HNO_3 and the solution was neutralize with NaOH and finally mixed with scintillator	JAEA-Technology 2009-051

Ge：Ge Semiconductor Detector
LSC：Liquid Scintillation Counter
GFC：Gas Flow Counter
ICP-MS：Inductively Coupled Plasma Mass Spectrometry

*1：S. Uchida et al., (1998) Hoken-butsuri, 1998, vol. 33, No. 1, p. 35-39
*2：K. Tagami (2003) Housha-Kagaku-news, 2003, No.8, p. 3-8
*3：H. Ichige et al., (2010) *RADIOISOTOPES*, 2010, 59, p.367-378

Example of Uncertainty（H-3）



Notes：Cause of uncertainty depends on the analysis method

Example of evaluation of uncertainty（H-3）

Cause of uncertainty	Relative standard uncertainty*1	Value (%)
Uncertainty of sample analysis		
・Uncertainty of sample collection	μ_1	0.618
Uncertainty of equipment calibration		
・Uncertainty of standard source	μ_2	2.550
・Uncertainty of sampling of standard source	μ_3	1.020
・Uncertainty of counting for standard source	μ_4	0.854
・Uncertainty due to fitting using calibration formula	μ_5	0.654
・Uncertainty for decay correction	μ_6	0.004
Uncertainty of sample measurement		
・Fluctuation of background	μ_7	2.438
・Uncertainty of counting	μ_8	0.432
・Uncertainty for decay correction	μ_9	0.007

・Combined standard uncertainty $= \sqrt{\mu_1^2 + \cdots\cdots + \mu_9^2} = 3.9$ (%)

・Relative expanded uncertainty*2＝

（Combined standard uncertainty ）×2＝7.8 (%)

Value of uncertainty is different by each analysis. Also, if the concentration is very small, uncertainty becomes large

＊1：[Relative standard uncertainty (%)]＝[standard uncertainty]÷[analysis result]×100

＊2：It shows about 95% confidence interval, based on "Guide to the expression of uncertainty in Measurement(1995)".

Comparison of analytical result (*E*n number)

Evaluated analytical results by using *E*n number shown in B.3 of ISO/IEC17043: 2010(JIS Q 17043:2011), with consideration of uncertainty in analytical results

→If the absolute value of *E*n number exceed 1 (|*E*n|>1), the cause of discrepancy will be investigated.

$$En = \frac{X_{TEPCO} - X_{\mathrm{JAEA}}}{\sqrt{U_{TEPCO}^2 + U_{\mathrm{JAEA}}^2}}$$

X_{TEPCO} : Measured value (radionuclide concentration) by TEPCO

X_{JAEA} : Measured value (radionuclide concentration) by JAEA NSRC

U_{TEPCO} : Uncertainty of TEPCO's value

U_{JAEA} : Uncertainty of JAEA NSRC's value

Analysis result(1/2)

➢ **Nuclides which were not detected in the analysis of JAEA**

Nuclides		JAEA (Bq/L)	TEPCO* (Bq/L)	Concentration limit (Bq/L)
To be Compared	Ru-106	<0.66	<0.415	100
	Sb-125	<0.18	0.150±0.0749	800
	Cs-134	<0.066	<0.0573	60
Confirmation of existence	Cl-36	<1.5	—	900
	Fe-55	<0.36		2,000
	Se-79	<0.77		200

・Any detection limit is lower than 1/100 of regulatory concentration limit
・Abundance of Cl-36, Fe-55, Se-79 were lower than detection limit

＊ : materials of hearings from the operators on 22nd February 2023
https://www2.nra.go.jp/data/000421817.pdf (only in Japanese)

Analysis result(2/2)

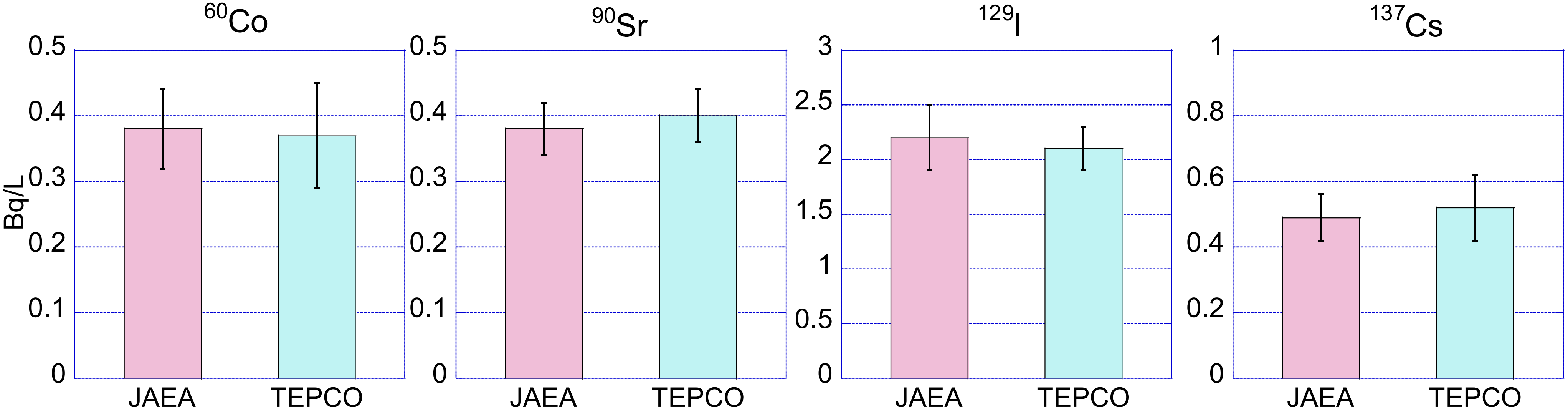
- Nuclides which were detected in the analysis of JAEA

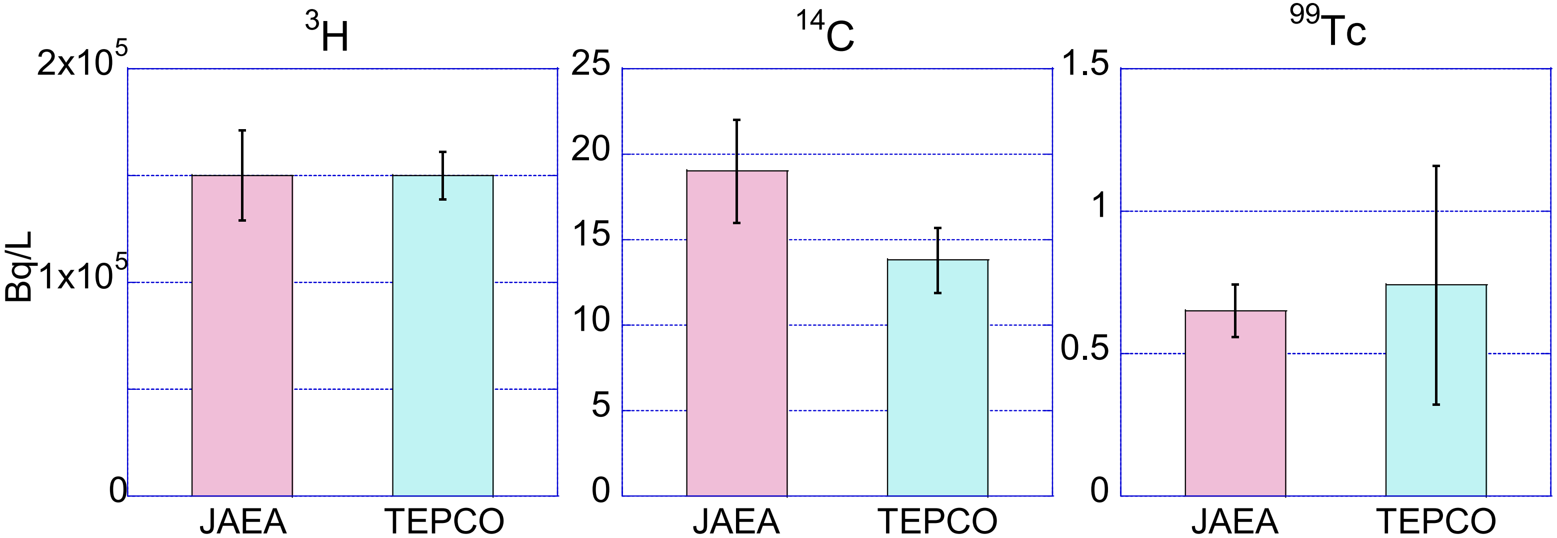
Nuclides		JAEA (Bq/L)	TEPCO* (Bq/L)	Concentration limit (Bq/L)	|*En*|
To be Compared	Co-60	0.38±0.06	0.373±0.0745	200	0.11
	Sr-90	0.38±0.04	0.399±0.0383	30	0.39
	I-129	2.2±0.3	2.13±0.162	9	0.21
	Cs-137	0.49±0.07	0.517±0.100	90	0.21
	H-3	(1.5±0.2)E+05	(1.46±0.102)E+05	60,000	0.0072
	C-14	19±3	13.8±1.90	2,000	1.5
	Tc-99	0.65±0.09	0.735±0.412	1,000	0.2

・The values of |*En*| were below 1, except for C-14.

＊ : materials of hearings from the operators on 22nd February 2023
https://www2.nra.go.jp/data/000421817.pdf (only in Japanese)

Analysis result(2/2)





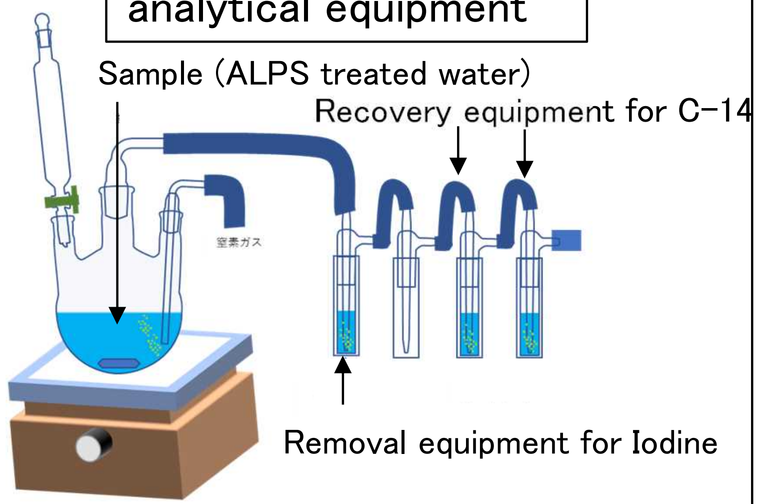
Error bar shows relative expanded uncertainty

Discussion: Analysis of C-14

- The discrepancy in the measured concentrations of C−14 between JAEA NSRC and TEPCO would be induced by volatile I−129 in ALPS treated water which exists at concentration of 2.2Bq/L.

→Reanalysis was performed by adding removal equipment for iodine (silver nitrate solution).

Schematic drawing of analytical equipment



Result of reanalysis

・Result：14±2 Bq/L
（TEPCO：13.8±1.9 Bq/L）

・|*En*| ＝0.07 （＜1）