The Commission on Supervision and Evaluation of the Specified Nuclear Facilities (79<sup>th</sup> meeting) Reference Document 4

Results of Investigation into the Deviation of Total Values for the Primary Seven Nuclides from Gross- Values for Water Treated with Multi-Nuclide Removal Equipment, etc.

March 16, 2020



Tokyo Electric Power Company Holdings, Inc.

# 1 . Background Information

Due to the deviation between measurement values taken by gross-(hereinafter referred to as, "gross beta") radiation measurement methods and the total values for the primary 7 nuclides (Cs-134/137, Co-60, Sb-125, Ru-106, Sr-90, I-129), tank H4N-A6, which showed the largest deviation, was investigated. The cause of this deviation was found to be a significant amount of C-14 and Tc-99, which were detected during the investigation. This was reported during the 67<sup>th</sup> meeting of the Commission on Supervision and Evaluation of the Specified Nuclear Facilities on January 21, 2019.

- In order to substantiate the investigation results three tanks with large deviations and two tanks with small deviations were investigated and it was confirmed that the primary cause of the deviation is the presence of C-14 and Tc-99 (June 17, 2019, 72<sup>nd</sup> meeting of the Commission on Supervision and Evaluation of the Specified Nuclear Facilities).
- More analysts were trained in order to enhance our mechanism for analyzing C-14, which is a nuclide that is difficult to measure.
- Analysis was completed in FY2018 and tanks that showed large deviations were subjected to additional investigation. At the same time, similar investigations were implemented for tanks for which analysis was newly completed in FY2019.

## 2. Selecting tanks to be analyzed (1/3)



As with the previous investigation, the tanks that were selected for analysis met the following conditions.

#### Selection criteria

Gross beta data/Primary 7 nuclides (conversion ) > 3 (deviation of 300% or more) and, Gross beta Primary 7 nuclides (conversion ) > 10Bq/L(absolute value deviation is 10 or higher)

Since the contribution to gross beta from each nuclide di ers, the contribution to gross beta from the primary 7 nuclides was calculated using the gross beta conversion coefficient noted in the High Energy Accelerator Research Organization report "Using egs5 to Calculate the Gross Beta Conversion Coefficients for Each Measured Nuclide at the TEPCO Fukushima Daiichi Nuclear Power Station" (KEK Internal 2018-6 January 2019 R). Furthermore, the contribution to gross beta from the primary 7 nuclides was added under the assumption that Y-90 and Rh-106, which are daughter nuclides of Sr-90 and Ru-106, are in radioactive equilibrium with their parent nuclides.

◆ Tanks that met investigation selection criteria were as follows.

- Eight tanks out of the tanks that were analyzed during FY2018
- Nine tanks out of the tanks that are analyzed during FY2019

# 2. Selecting tanks to be analyzed (2/3)



## [Tanks for which analysis was completed during FY2018]

The tanks shown in the chart below were selected for further analysis based upon the analysis results from FY2018

No.	Selected tank	Notes (FY2018 analysis results)	
1	H2-A1	Gross beta/primary 7 nuclides=3.21	16.94Bq/L
2	J7-D1	Gross beta/primary 7 nuclides=3.90 (sublevel measurement results)	11.84Bq/L
3	H4-D7	Gross beta/primary 7 nuclides=4.13	14.31Bq/L
4	H4-D8	Gross beta/primary 7 nuclides=3.85	35.95Bq/L
5	H1E-C8	Gross beta/primary 7 nuclides=3.11	11.81Bq/L
6	K3-A3	Gross beta/primary 7 nuclides=3.25	11.18Bq/L
7	H4N-C1	Gross beta/primary 7 nuclides=3.12	12.68Bq/L
8	G1S-A5	Gross beta/primary 7 nuclides=3.54	24.24Bq/L

# 2. Selecting tanks to be analyzed (3/3)



#### [Tanks for which analysis was completed during FY2019]

The tanks shown in the chart below were selected for further analysis based upon the analysis results from FY2019

No.	Selected tank	Notes	
1	H4-B1	Gross beta/primary 7 nuclides=4.10	19.91Bq/L
2	H4-B6	Gross beta/primary 7 nuclides=5.13	18.07Bq/L
3	H4-B7	Gross beta/primary 7 nuclides=4.82	48.97Bq/L
4	H6(1)-B1	Gross beta/primary 7 nuclides=4.54	29.99Bq/L
5	G6-B6	Gross beta/primary 7 nuclides=5.48	25.46Bq/L
6	G6-B1	Gross beta/primary 7 nuclides=4.56	17.16Bq/L
7	H5-B11	Gross beta/primary 7 nuclides=4.13	14.58Bq/L
8	H6(2)-A1	Gross beta/primary 7 nuclides=3.87	22.58Bq/L
9	H3-B5	Gross beta/primary 7 nuclides=3.19	20.44Bq/L

## 3. Investigation details

The following analyses were conducted for the tanks subject to investigation

No.	Analysis	Notes
1	Gross beta	
2	ray-emitting nuclides (Cs-134, Cs-137, Co-60, Sb-125, Ru-106)	Primary 7 nuclides
3	Sr-90	Primary 7 nuclides
4	I-129	Primary 7 nuclides
5	C-14	
6	Tc-99	

< Note >

- New samples were taken from the eight tanks for which analysis concluded during FY2018, and those new samples were subjected to the analyses shown above .
- The analyses shown above were conducted for all 30 tanks for which analysis was completed during FY2019. This report covers nine tanks that showed large deviations.
- Approximately 20 days per tank was needed for analysis.
- The analysis process commenced with tanks for which sampling had been completed and all parts of the process were performed simultaneously thereby enabling all 38 tanks to be analyzed over approximately two months.



 Gross beta contribution (considering self-absorption) from the 7 primary nuclides, C-14 and Tc-99 did not fall below gross beta values.



H4-D7

0.80

Gross beta

contribution

60

50

40

30

20

10

0

Measurement

value

Primary 7

nuclides

+ Rh106

+ Y-90

Bq/L



The analysis results shown above are from specimens that were resampled during FY2019

11

Gross beta

Reference

Gross beta contribution

(Self-absorption considered

■ C-14 ■ Tc-99 ■ Adjustment ■ Gross beta

value

Gross beta contribution (self-absorption considered) values calculated using the self-absorption adjustment formula noted in the radioisotope pocket data book under the assumption that the self-absorption-causing substance is equally distributed throughout the sample. This is provided as a reference value since the degree of self-absorption may change depending on the existence form.

+ Y-90 + Rh106













The analysis results shown above are from specimens that were resampled during FY2019





H4-B7



 Gross beta contribution (considering self-absorption) from the 7 primary nuclides, C-14 and Tc-99 did not fall below gross beta values in tanks for which analysis had been completed since FY2019.



31

. .

Gross beta



# \_\_\_\_

Reference

value

G6-B1



H5-B11







- It is assumed that the self-absorption attribute of the specimens played a large part in the difference between gross beta contribution (self-absorption considered) and gross beta values.
- In order to substantiate this claim, multiple specimens for gross beta measurements were created from the water from tank H4-D7 and it was found that the minimum value was approximately 33% lower (approximately 10Bq/L) than the maximum value (approximately 15Bq/L)
- The investigations conducted to date have shown that the primary causes of the deviation are C-14 and Tc-99 since the gross beta contribution (self-absorption considered) is larger than gross beta values.
- Beta-ray energy spectral analysis performed to date did not find the presence of any nuclide other than C-14 and Tc-99 that would cause a large impact on gross beta values, so it is assumed that the cause of the deviation was the presence of C-14 and Tc-99.

# 5 . Conclusion

- Causes of the deviation between the primary 7 nuclide total values and gross beta values
- Analysis did not find the presence of any nuclide other than C-14 and Tc-99 that would cause a large impact on gross beta values, so it is assumed that the primary cause of the deviation was the presence of C-14 and Tc-99.

## Future handling

 C-14, Tc-99 and gross beta analyses will continue for the primary seven nuclides in tanks for which analysis has been completed, and if the gross beta contribution falls below gross beta values even when taking into consideration the gross beta contribution from the primary seven nuclides, C-14 and Tc-99, further investigation will be performed to look for the presence of unidentified nuclides.

End of document



#### Radiation concentration unit: Bq/L

Selected Tank	Cs-134	Cs-137	Co-60	Ru-106	Sb-125	Sr-90	I-129	Gross-	C-14	Tc-99	Precipitate weight (mg)
H2-A1	<0.43	<0.25	0.23	<1.7	<0.66	6.2	5.2	42	108	<1.0	56.78
J7-D1	<0.24	<0.25	0.95	<1.4	<0.45	0.75	2.8	11	17	4.4	57.16
H4-D7	<0.18	0.31	0.49	<1.4	<0.48	0.69	2.2	11	40	<1.0	52.51
H4-D8	<0.21	<0.20	1.3	<1.4	0.81	<0.40	13	35	139	<1.0	60.07
H1E-C8	<0.22	0.47	0.86	<2.0	<0.64	<0.51	2.8	22	15	15	47.58
K3-A3	<0.39	1.3	0.51	<2.8	<1.1	<0.47	3.8	19	17	9.1	48.40
H4N-C1	<0.15	<0.24	1.6	<1.4	<0.46	<0.42	1.0	26	67	<1.2	64.37
G1S-A5	<0.19	0.34	1.3	<1.4	<0.48	<0.50	7.6	26	80	<1.2	59.44

## TEPCO

#### Radiation concentration unit: Bq/L

Selected Tank	Cs-134	Cs-137	Co-60	Ru-106	Sb-125	Sr-90	I-129	Gross-	C-14	Tc-99	Precipitate weight (mg)
H4-B1	<0.20	0.40	2.1	<1.5	<0.47	0.81	0.80	26	102	<1.2	60.17
H4-B6	<0.15	0.44	0.70	<1.2	<0.43	<0.40	1.3	22	76	<1.2	55.20
H4-B7	<0.17	<0.24	0.70	<1.2	0.56	<0.39	17	62	215	<1.2	51.78
H6(1)-B1	<0.13	0.70	2.9	<1.3	<0.42	1.1	2.3	38	122	5.7	52.45
G6-B6	<0.28	0.22	1.7	<1.2	<0.44	0.55	1.9	31	119	<1.3	62.07
G6-B1	<0.16	<0.23	0.94	<1.3	<0.47	<0.45	1.8	22	51	<1.3	63.45
H5-B11	<0.12	<0.20	0.68	<1.2	<0.39	0.41	2.3	19	59	<0.52	56.11
H6(2)-A1	<0.24	<0.23	1.3	<1.3	<0.46	1.2	3.7	30	107	<0.52	60.40
H3-B5	<0.27	0.44	1.7	<1.2	<0.39	2.3	1.4	30	61	<0.52	71.94

#### < Reference > Analysis results for tanks that did not show large deviations from amongst the tanks that were analyzed during FY2019 (1/2)



								Radiati	on concer	itration u	nit: Bq/L		
Tank	Cs-134	Cs-137	Co-60	Ru-106	Sb-125	Sr-90	I-129	Primary seven nuclide conversion values	Gross- contribution (self-absorption considered)	Gross-	C-14	Tc-99	Precipitate weight (mg)
H4-E1	<1.1	6.7	<0.95	<7.5	<2.3	3.1	2.2	27	40	35	34	<1.2	48.54
H6(1)-A1	<1.6	2.4	<3.0	<14	<4.5	0.84	1.1	28	76	39	119	<1.3	42.70
H6(1)-A5	2.6	43	<1.1	<9.5	<3.9	21	1.0	109	144	98	95	<1.3	45.26
H6(1)-B5	<1.3	28	<0.95	<8.6	<3.5	8.9	2.0	65	129	103	116	32	51.43
B-C1	<0.33	1.6	0.52	<1.5	1.9	1740*	45	3790	3640	3850*	10	4.6	75.54
B-D1	<0.16	0.30	<0.18	<1.3	<0.50	1.2	0.66	5.3	7.5	8.0	3.8	<1.3	48.77
B-E1	<0.21	0.39	0.48	<1.4	2.2	457*	46	1020	973	1040*	9.9	4.5	76.34
B-E6	<0.23	1.0	0.46	<2.3	2.4	7360*	41	15900	15200	15600*	13	4.8	90.92
BS-A1	<0.21	<0.24	<0.19	1.8	<0.72	3.8	0.91	12	14	8.7	5.4	<1.3	51.19
BS-A5	<0.18	0.39	0.77	<1.3	<0.40	3.5	2.6	12	16	7.3	13	<1.3	76.77
H5-A1	<0.41	<0.24	1.2	1.8	1.4	<0.34	2.0	6.4	36	18	83	<1.3	64.83

\*Welded tanks currently holding water that was transferred from G4, G5 tanks (flange tanks), in which water treated during FY2013-2014 was being stored, that became full during FY2019.

#### < Reference > Analysis results for tanks that did not show large deviations from amongst the tanks that were analyzed during FY2019 (2/2)

## TEPCO

Tank	Cs-134	Cs-137	Co-60	Ru-106	Sb-125	Sr-90	I-129	Primary seven nuclide conversion values	Gross- contribution (self-absorption considered)	Gross-	C-14	Tc-99	Precipitate weight (mg)
H5-B1	<0.24	<0.23	1.3	<1.4	3.4	<0.39	2.2	7.3	18	22	30	<1.3	62.23
H5-C7	<0.18	<0.23	1.6	<1.9	<0.72	<0.44	5.1	8.0	35	24	78	<0.52	62.96
H5-C1	<0.29	<0.20	1.5	1.1	0.70	<0.41	2.2	5.5	23	14	47	<0.52	59.50
H5-A12	<0.14	<0.23	0.66	<1.3	<0.46	<0.41	2.8	5.1	24	15	53	<0.52	60.87
H6(2)-B1	<0.18	<0.21	0.65	<1.2	0.51	<0.38	2.3	4.7	8.4	<5.4	11	<0.52	60.29
H6(2)-B5	<0.22	<0.24	1.6	1.8	1.8	<0.40	5.0	8.6	18	16	32	<0.52	74.73
H6(2)-A5	<0.17	<0.24	1.2	<1.5	<0.47	9.3	1.2	24	46	40	67	<0.52	71.49
G1S-B5	<0.42	2.6	0.62	<3.0	3.8	18500*	43	40000	37300	37700*	23	6.6	132.5
H3-A1	<0.19	<0.25	0.61	<1.2	<0.46	5.3	<0.19	14	52	32	104	<0.52	58.93
G3-D1	<1.7	9.3	12	<11	17	2280**	1.9	4960	4830	5620**	9.5	<0.52	52.75

Radiation concentration unit: Bq/L

\*Welded tanks currently holding water that was transferred from G4, G5 tanks (flange tanks), in which water treated during FY2013-2014 was being stored, that became full during FY2019.

\*\* Tanks in which Sr-treated water was previously being stored that have been repurposed for ALPS-treated water storage.