Results of the Investigation into the Deviation between Gross Beta Data and Primary 7 Nuclide Totals for Water Treated with Multi-nuclide Removal Equipment, etc.

June 17, 2019

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1．Reasons for the investigation into the deviation

- Results of analyses for tank groups performed during FY2018 showed a deviation between the measurement values taken by gross beta radiation measurement methods (hereinafter referred to as, “gross beta※”) and the total values for the primary 7 nuclides (Cs-134/137, Co-60, Sb-125, Ru-106, Sr-90, I-129), so an investigation into the cause of this deviation was performed under the suspicion that an unidentified nuclide was present.

- When the H4N-A6 tank, which showed a large deviation, was analyzed, significant amounts of C-14 and TC-99 were detected.
  Furthermore, the concentration of C-14 detected at this time was approximately 45Bq/L (legally acceptable concentration: 2,000Bq/L) and the concentration of TC-99 was approximately 29Bq/L (legally acceptable concentration: 1,000Bq/L), which fall below the upper limit of legally accepted concentration levels.

- During the 67th meeting of the Monitoring and Assessment Review Committee held on January 21, 2019, we were instructed to perform further analysis of multiple tanks other than the H4N-A6 tank, so additional tank investigations were performed.

※Gross beta data is used for the following two objectives:
①Gross beta data is used as a substitute value for Sr-90 for wastewater samples that have low self-absorption, such as water from the groundwater bypass and subdrains
②Gross beta data is used to check for the presence of nuclides other than nuclides being measured in samples with large self-absorption upon looking at the radioactive substance concentration level trends of the samples.

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

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◆ A total of five tanks were selected for further investigation.
A breakdown of the five tanks is shown below:

①Three tanks from tank groups for which the deviation between gross beta data and the primary 7 nuclides was large
②One tank from a tank group for which the deviation between gross beta data and the primary 7 nuclides was small
③One tank from the K4 tank group for which detailed analysis of 62 nuclides has already been implemented

◆ The degree of deviation was determined by the ratio of gross beta data to primary 7 nuclides and a selection was made [from tanks] which fulfill the following criteria.

①Large deviation: 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)
②Small deviation: Gross beta data/Primary 7 nuclides (conversion※)＝1±0.05(as close to 1 as possible)

※Since the contribution to gross beta from each nuclide differs, the contribution to gross beta of 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.

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

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◆ Tanks to be further investigated were selected as shown in the following graph based on the approach explained on the previous page

No.	Condition	Selected tank	Notes (FY2018 analysis results)
1	Large deviation	**G1S-B7**	Gross beta/primary 7 nuclides＝4.63　Δ23.45Bq/L
2		**H4-D1**	Gross beta/primary 7 nuclides＝4.16　Δ22.33Bq/L
3		**H4-A11**	Gross beta/primary 7 nuclides＝3.31　Δ17.05Bq/L
4	Small deviation	**J3-B1**	Gross beta/primary 7 nuclides＝0.97
5	K4 tank group	**K4-D1**	K4 tank group tank with smallest deviation Gross beta/primary 7 nuclides＝1.13
6	Analysis completed	H4N-A6	Gross beta/primary 7 nuclides＝4.45　Δ31.55Bq/L

＜Reference＞

Number of tank groups for which analysis has been completed as of the end of FY2018	Number of tank groups for each gross beta/primary 7 nuclides ratio (gross beta/primary 7 nuclides)			
	Less than 2 times	2 times or more, but less than 3 times	3 times or more, but less than 4 times	4 times or greater※
153	**100**	**36**	**13**	**4**

※The largest deviation found during this investigation was for tank G1S-B7(4.63)

3. Investigation details

◆ The following analyses were performed for all five tanks

No.	Analysis	Notes
1	Gross beta	
2	**γ 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	
7	Nuclide other than those mentioned above (according to beta ray energy spectrum)	See Note

Note: The spectrum is used to check for the presence of beta ray-emitting nuclides other than C-14 and Tc-99, and if such presence is detected further analysis is performed to identify the nuclide.

4. Investigation results

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◆ Take a group analysis results

Primary 7 nuclides

Unit: Bq/L

No.	Selected tank	Cs-134	Cs-137	Co-60	Sb-125	Ru-106	Sr-90	I-129	C-14	Tc-99
1	G1S-B7	<0.061	0.19	0.60	0.45	1.2	1.1	3.0	**78**	3.8
2	H4-D1	<0.071	0.14	0.51	0.32	1.9	0.35	3.4	**71**	6.5
3	H4-A11	<0.063	0.067	0.95	0.42	<0.46	0.49	7.3	**140**	<0.70
4	J3-B1	0.16	0.96	0.92	0.75	<0.47	<0.27	9.0	**14**	<0.70
5	K4-D1	0.16	0.12	0.64	0.17	<0.48	<0.19	3.0	**10**	<0.70

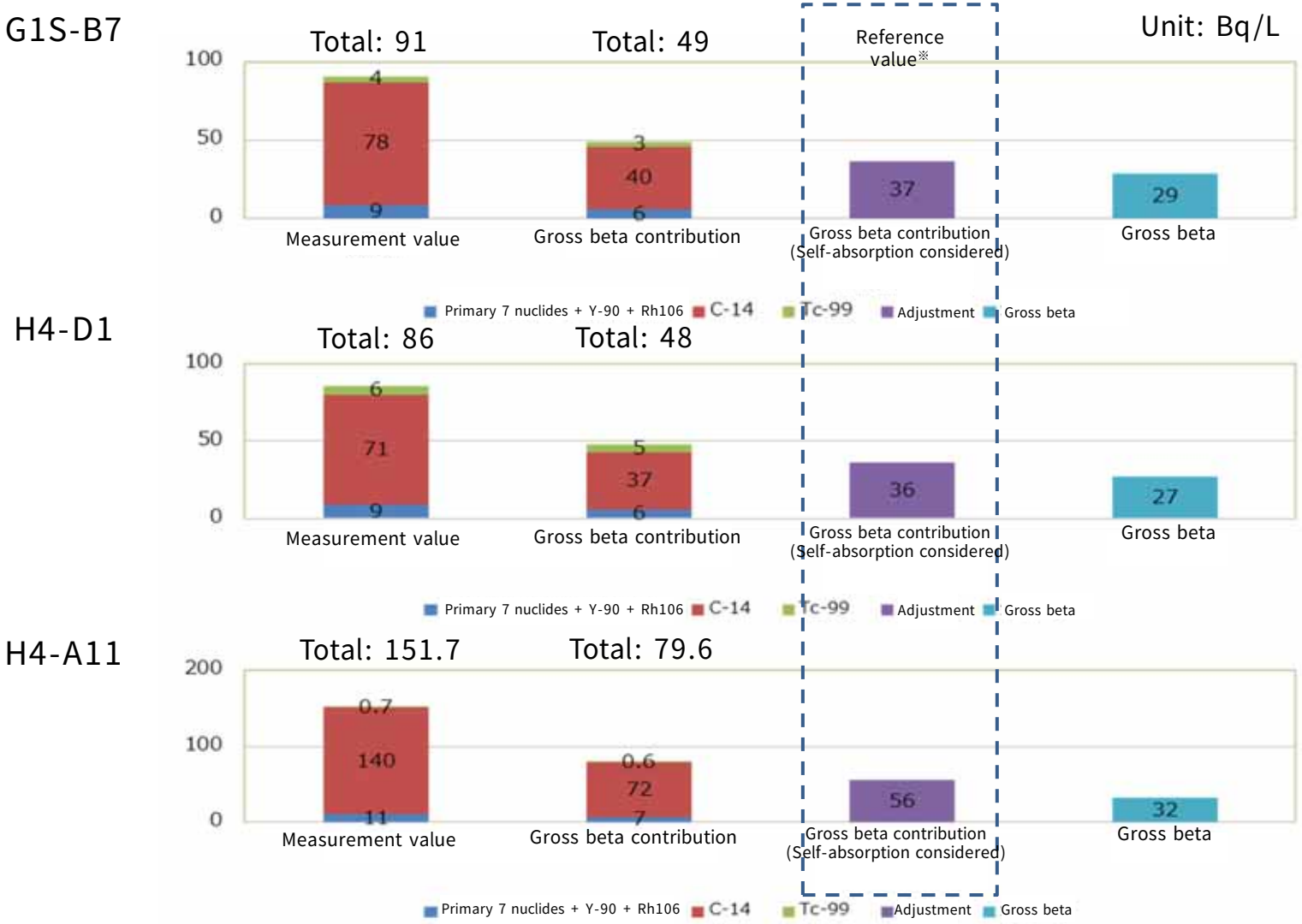
No.	Selected tank	Gross beta*	Residual amount* (mg)	Deviation
1	G1S-B7	29	59.62	**Large**
2	H4-D1	27	54.38	
3	H4-A11	32	63.39	
4	J3-B1	12	68.47	**Small**
5	K4-D1	5	42.55	

*Data for gross beta and residual amount are average of 3 measurements.

- A significant amount of C-14 was found in tanks with large deviations. Meanwhile, low concentrations of C-14 were found in tanks with small deviations.
- This investigation only found the low concentrations of Tc-99, which was significantly detected in tank H4N-A6.

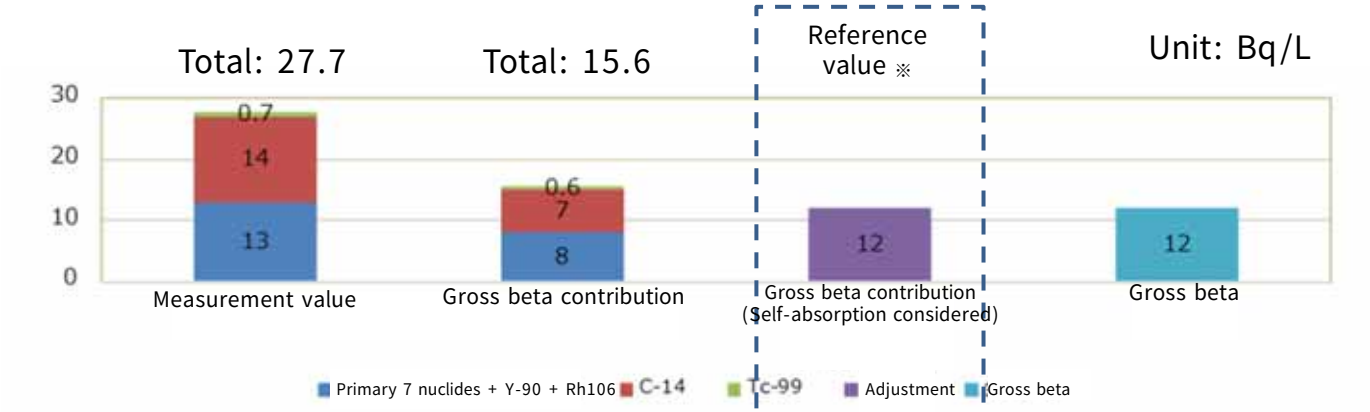
4. Investigation results: Tank groups with large deviations

- Gross beta contribution (considering self-absorption) from C-14 did not fall below gross beta.
- The same results were found for all three tanks with large deviations.

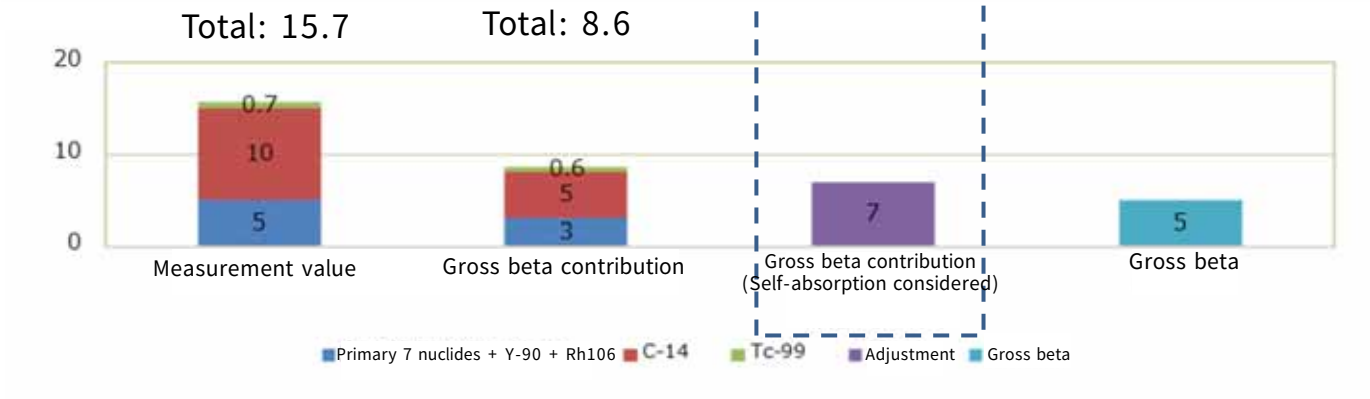


4. Investigation results: Tank groups with small deviations

J3-B1



K4-D1



※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.

5. Observations/Conclusions

◆ Causes of the deviation between the primary 7 nuclide total values and gross beta values

・The investigations conducted to date have shown that the primary causes of the deviation are C-14 and Tc-99.

・The investigation 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.

・Furthermore, the causes of the fluctuations in the radiation concentrations of ALPS-treated water are fluctuations in the radiation concentrations of water before it has been treated, and a decrease in the performance of adsorbents. These fluctuations can be reduced by increasing the frequency at which adsorbents are replaced.

・During this investigation we were able to establish an analysis method for identifying unidentified nuclides when there is a deviation between gross beta values and the totals for measured nuclides, and we believe this analysis method to be accurate.

◆ Future handling

・If disposing of ALPS-treated water by discharging it into the environment, the water shall be subjected to secondary treatment to ensure that the sum of the ratios is less than 1 as legally required.

・Going forward, the contribution of that C-14 makes to the legally required sum of the ratios shall be considered. (Tc-99 has already been considered because it is a nuclide that is subject to removal)

・Measuring]gross beta values is an effective method for detecting the presence of nuclides that are not subject to measurement and we will continue to use this method not just to monitor tanks, but for various types of trend monitoring.

End of document

＜Reference＞List of analysis methods

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◆ Analysis methods used during this investigation

■ Primary 7 nuclide analysis methods

Nuclide	Analysis method	Chemical pretreatment	Measurement instrument	Specimen size	Measurement time
Cs-137	γ ray spectrometry	None	Ge detector	2L	100,000 seconds
Cs-134					
Sb-125					
Co-60					
Ru-106					
I-129	ICP-MS		ICP-MS	2 or 5mL	100 seconds
Sr-90	Sr resin	Sr extraction using Sr resin	Pico beta	1L	1,000 seconds

■ Gross beta analysis methods

Nuclide	Analysis method	Chemical pretreatment	Measurement instrument	Specimen size	Measurement time
Gross beta	Gross beta radiation measurement	Evaporation to dryness	Low background gas flow counter	10mL	6,000 seconds

■ C-14, Tc-99 analysis methods

Nuclide	Analysis method	Chemical pretreatment	Measurement instrument	Specimen size	Measurement time
C-14	Wet digestion	Oxidization using oxidant	Liquid scintillation counter	100mL	15,000 seconds
Tc-99	ICP-MS	None	ICP-MS	2mL	100 seconds

＜Reference＞Treatment period of each tank group

Tank Name	G1S-B7	H4-D1	H4-A11	J3-B1	K4-D1	H4N-A6
Treatment Period	FY2018	FY2018	FY2017～FY2018	FY2014	FY2016	FY2017