Supervision and Evaluation Committee
for Specified Nuclear Power Facilities
(83rd Meeting)
Document 5-3

Deviations between Gross Beta Values and the Totals for the Primary Seven Nuclides for ALPS-treated water

September 14, 2020

TEPCO

Tokyo Electric Power Company Holdings, Inc.

1. Selection of the primary seven nuclides

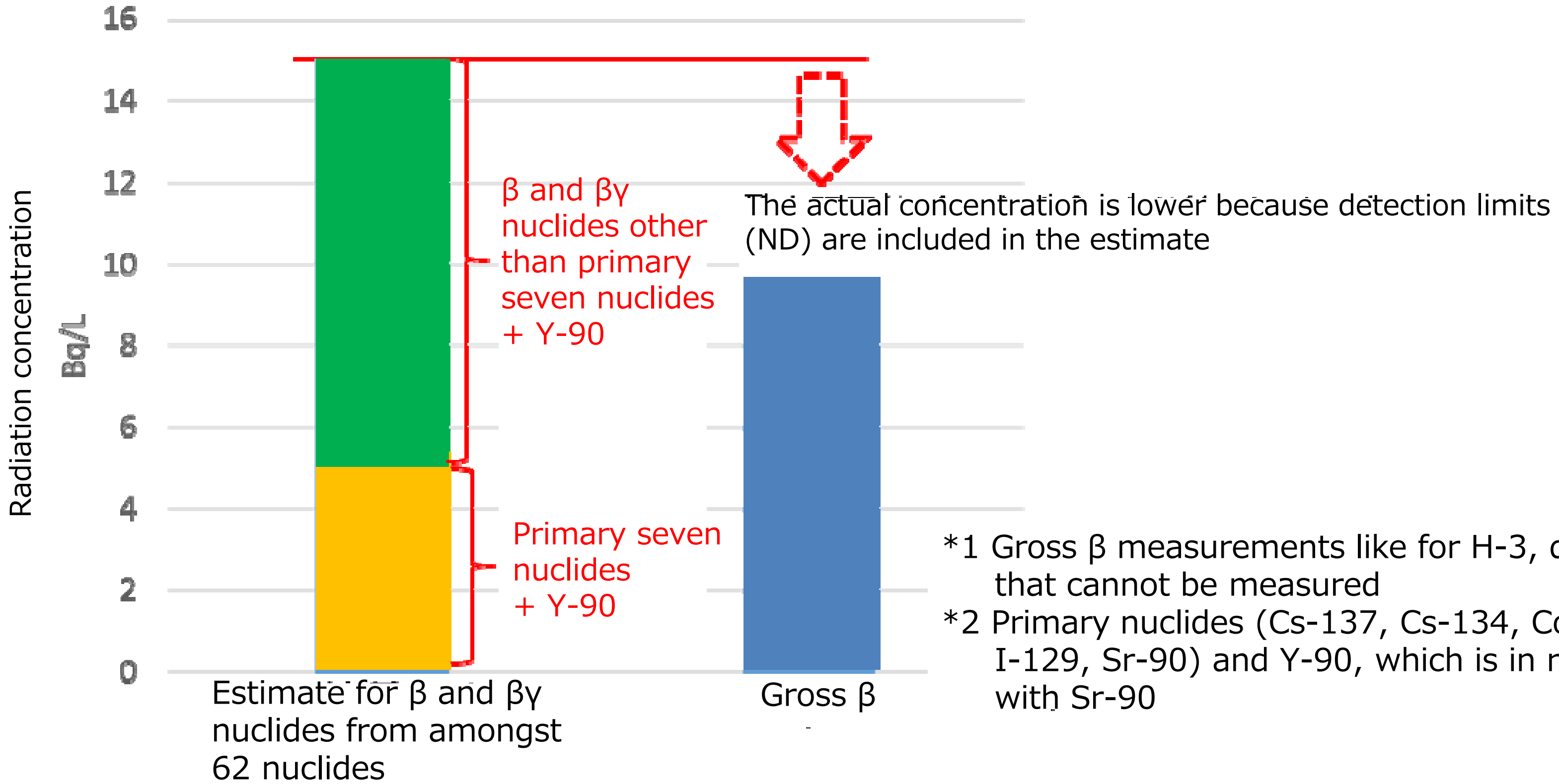
- Multi-nuclide removal equipment has the ability to remove 62 nuclides to the point where the sum of the ratios of concentrations required by law for these nuclides is less than 1.
- However, whereas much time is required to perform analyses for all 62 nuclides, it is necessary to select representative nuclides to measure and assess in order to ascertain the concentrations of nuclides contained in water stored in tank groups, and also verify ALPS performance, without causing delays to the decommissioning process.
- Therefore, an analysis of the 62 nuclides in treated water was performed, and the following "primary seven nuclides," for which significant concentrations required by law were detected, were selected.
- The sum of the ratios of concentrations required by law for the nuclides targeted for removal by ALPS during the water treatment process is calculated by setting the sum of the ratios of concentrations required by law for the 55 other nuclides as 0.3, and adding that to the actual concentration measurements for the primary seven nuclides.

The primary seven nuclides selected for ALPS-treated water analysis

Cs-134, Cs-137, Sr-90, I-129, Ru-106, Co-60, Sb-125

2. Awareness in regards to the deviation between the totals for the primary seven nuclides and gross beta values

- As of the first half of FY2018, we were aware that there were tanks in which the total values from the analysis results of the primary seven nuclides in ALPS-treated water deviated a certain extent from gross beta values.
- At the time, we believed that the reason why gross beta values were higher than the totals for the primary seven nuclides was because the nuclides that are targeted for removal other than the primary seven nuclides existed in concentrations that were below detectable limits, and that the beta rays emitted by these nuclides were affecting the assessment results.



*1 Gross β measurements like for H-3, do not include nuclides that cannot be measured
*2 Primary nuclides (Cs-137, Cs-134, Co-60, Sb-125, Ru-106, I-129, Sr-90) and Y-90, which is in radioactive equilibrium with Sr-90

Estimates for β and βγ nuclides (62 nuclides) in the K4 tank and comparison with gross β values

3. Investigations conducted (first)

- Since the assessment mentioned in the previous slide was just a guess, an investigation was conducted to identify the nuclides that were causing the deviation.

- The H4N-A6 tank was chosen to be investigated because it had the largest deviation between gross beta values and the radiation concentration totals for the seven primary nuclides.

- The beta ray spectral analysis of the ALPS outlet water and the water in the H4N-A6 tank suggested the presence of two spectrums that could not be identified (one with a maximum energy equal to that of I-129, and another with approximately two times that energy)

Gross beta values and concentrations of the primary seven nuclides※ in the H4N-A6 tank

Nuclide	Cs-137	Cs-134	Sr-90	Y-90	I-129	Ru-106	Rh-106	Co-60	Sb-125	Total	Gross β value
Concen-tration (Bq/L)	0.34	<0.17	0.19	0.19	1.77	5.77	5.77	0.46	0.65	15.31	40.74

※The assessment of the primary seven nuclides includes Y-90 and Rh-106, which are daughter nuclides that have a radiation equilibrium relationship to Sr-90 and Ru-106

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3. Investigations conducted (first)

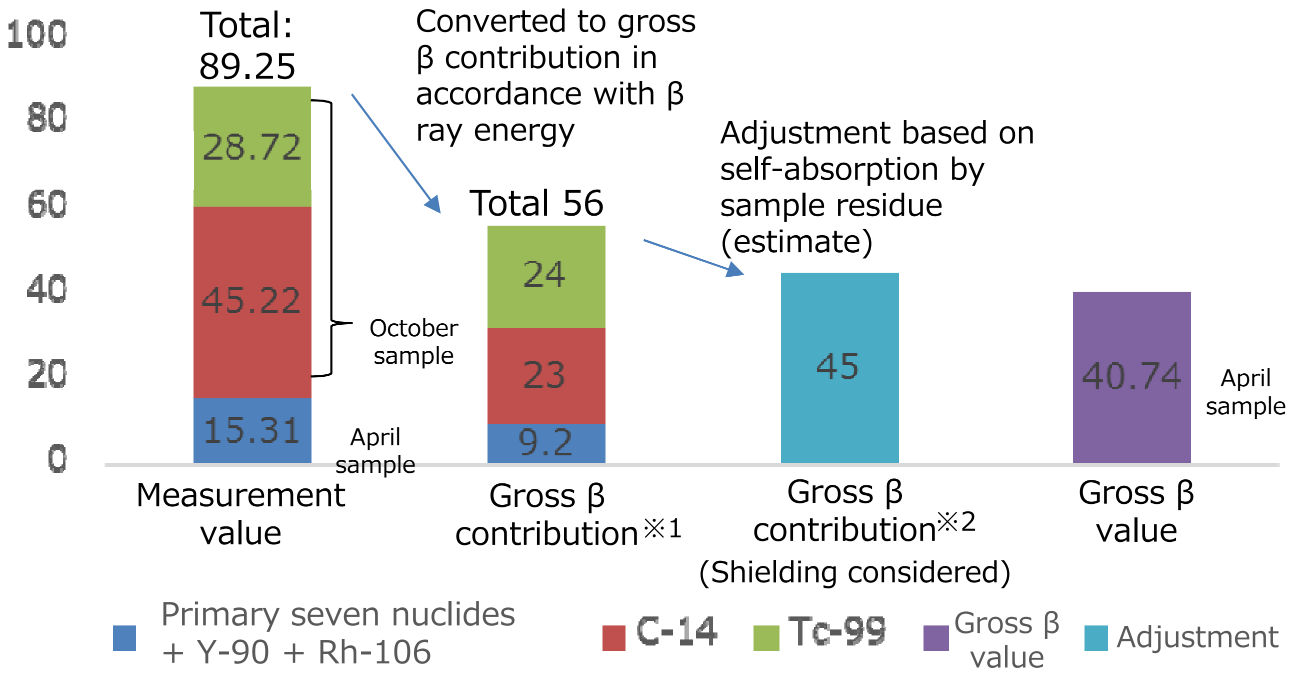
- This brought our attention to the existence of C-14 (which emits the same amount of maximum beta ray energy as I-129) and Tc-99 (which emits approximately two times the maximum energy of I-129), and when these nuclides were targeted for measurement, a significant concentration of both was detected.
- We also learned that the impact on gross beta values has a direct relationship with beta-ray emitting nuclides that have large maximum energies. Therefore, we performed an assessment using documented values that adds this impact to the gross beta values for each nuclide.

C-14 and Tc-99 measurement results

Nuclide	Measurement device	Concentration (Bq/L)
C-14	LSC	45.22
Tc-99	ICP-MS	28.72

Primary nuclides +

※1: The gross β contribution was calculated based on “Calculation of gross β conversion coefficients for each measured nuclide at the Fukushima Daiichi Nuclear Power Station by egs5” (KEK Internal 2018-6 January 2019 R)
※ 2 : The adjustment formula for self-absorption in the Radioisotope Pocket Data Book was used.



Gross β value assessment results including C-14 and Tc-99 (Bq/L)

- The results of this investigation were reported on at the 67th meeting of the Supervision and Evaluation Committee for Specified Nuclear Power Facilities (held on January 21, 2019)

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4. Investigations conducted (second)

- Thereafter we conducted analysis for the primary seven nuclides, C-14, Tc-99 and gross beta for three tanks with large deviations, and two tanks with small deviations.

- Tanks for which the difference between the total values for the primary seven nuclides (including Y-90 and Rh-106) and gross beta values exceeded 10 Bq/L, and for which the ratio was three times more, or greater, were defined as " tanks with large deviations."

- Analysis results showed that significant amounts of C-14 were detected in tanks with large deviations.

- Although C-14 was detected in tanks with small deviations, the concentrations were lower.

◆ Tank group analysis results

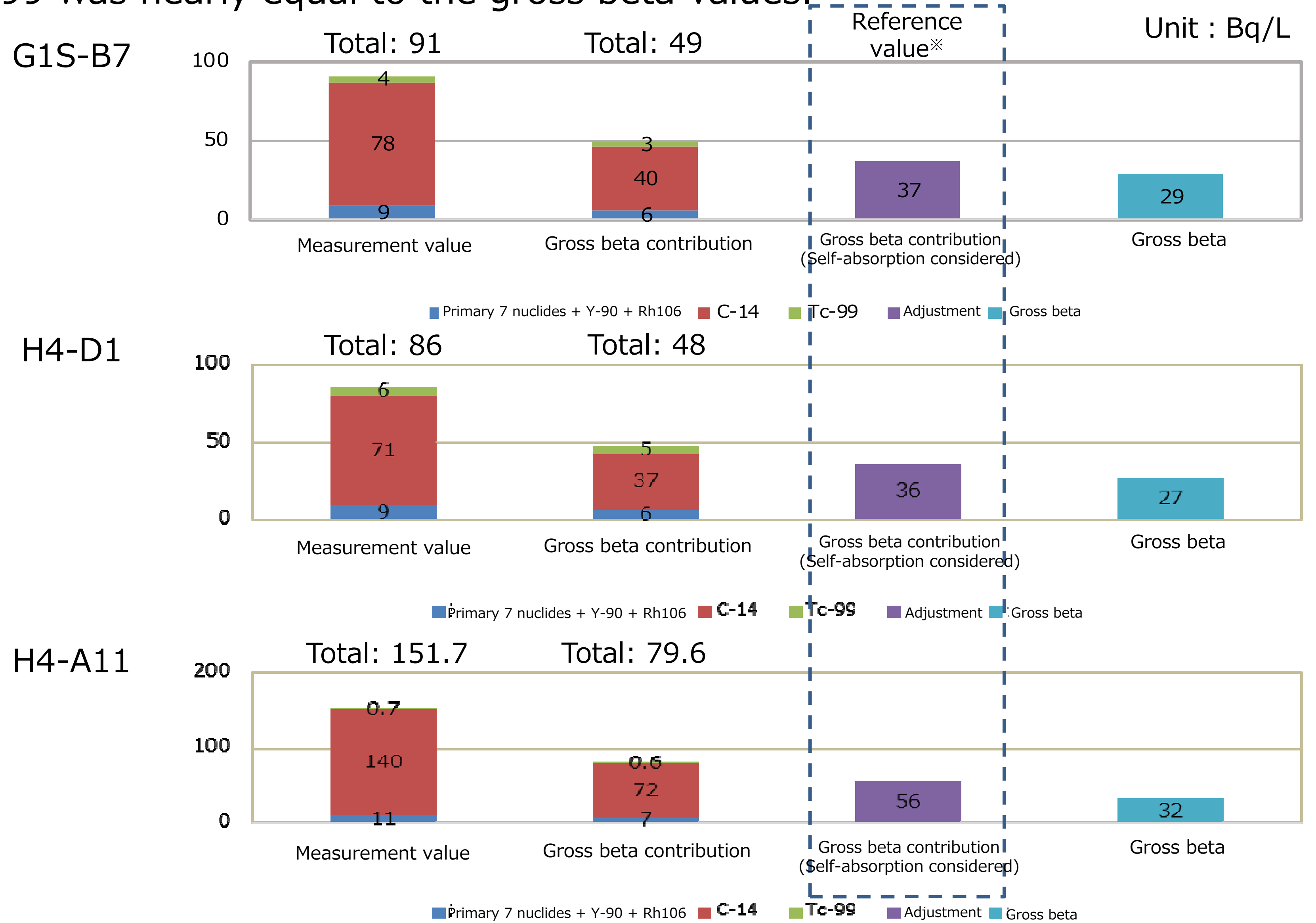
Primary seven nuclides (Cs-134 to I-129)

Units: Bq/L

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

4. Results of the second investigation

- The total value taking into consideration the gross beta contribution from C-14 and Tc-99 was nearly equal to the gross beta values.



※Gross beta contribution (self-absorption considered) values were 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.

- The results of this investigation were reported on at the 72nd meeting of the Supervision and Evaluation Committee for Specified Nuclear Power Facilities（held on June 17, 2019).

5. Investigations conducted (third)

- Even though the first and second investigations had already shown that the cause of the deviations was C-14 and Tc-99, we conducted an analysis of the primary seven nuclides, C-14, Tc-99 and gross beta for all the remaining tanks with large deviations in order to backup the results of previous investigations.
- The criteria for selecting tanks with large deviations were the same as the second investigation.
- Furthermore, we conducted an analysis of the primary seven nuclides, C-14, Tc-99 and gross beta for not just tanks with large deviations, but all tank groups that had become full since FY2019.
- Investigation results showed that the total values for the primary seven nuclides (including Y-90 and Rh-106), C-14 and Tc-99 was not less than gross beta values, thereby supporting our original conclusion that the cause of the deviations was the presence of C-14 and Tc-99.
  (refer to pages 14~18)

Selection criteria (same as second investigation)

Gross beta ÷ primary seven nuclides (converted) ＞ 3 (difference of three times or more), and;
Gross beta − primary seven nuclides (converted) ＞ 10Bq/L(difference in absolute value is 10 or higher)

- The results of this investigation were reported on at the 79th meeting of the Supervision and Evaluation Committee for Specified Nuclear Power Facilities (held on March 16, 2020)

6. Investigation conclusions

- Since investigation results for the tanks that have become full show that the total of the gross beta contributions from the primary seven nuclides, C-14 and Tc-99 approximately equal gross beta values, it is assumed that the cause of the deviation between gross beta values and the totals for the primary seven nuclides was the presence of C-14 and Tc-99.

- Therefore, tanks that become full will be subjected to analysis of not only the primary seven nuclides, but also of C-14 and Tc-99.

- In order to ensure that there is not an increase in unidentified nuclides in tanks in the future, we will continue to compare the results of nuclide analyses with gross beta values, and perform an investigation if we see any deviations that suggest the presence of other nuclides.

  (refer to p.9 “Future Tank Monitoring Plan”)

7. Future Tank Monitoring Plan

Full ALPS-treated water tanks

↓

Primary seven nuclides※1, C-14, Tc-99 and gross beta measured

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Comparison of the total gross beta contribution from the primary seven nuclides, C-14 and Tc-99※2 and gross beta values

↓

Gross beta contribution total ＜Gross beta values

- No → Normal monitoring continued
- Yes ↓

Tank groups for which gross beta values were larger than gross beta contribution totals for a certain period of time were selected as investigation targets, and gamma ray measurement results and beta rays spectral analysis were used to identify the presence of unknown nuclides. If it is determined that an unknown nuclide exists, the nuclide will be qualified and quantified.

⇒ If it is determined that the nuclides identified during these investigations have an impact on the sum of the ratios of concentrations required by law, we shall deliberate on how to proceed.

- ALPS-treated water tanks will be measured as they become full and monitored by comparing measurements with gross beta values in accordance with the flowchart on the left.

※1 : Cs-137, Cs-134, Co-60, Sb-125, Ru-106, I-129, Sr-90

※2: The gross β contribution (including the gross beta contribution from Y-90, which is in radioactive equilibrium with Sr-90, and Rh-106, which is in radioactive equilibrium with Ru-106) for each nuclide calculated based on "Calculation of gross β conversion coefficients for each measured nuclide at the Fukushima Daiichi Nuclear Power Station by egs5" (KEK Internal 2018-6 January 2019 R) while considering self-absorption.

8. Monitoring results since the third investigation

- Monitoring performed in accordance with the flowchart shown on the previous slide for tank groups that have become full since FY2019, and tank groups for which analysis was completed between the third investigation and June 2020, did not reveal any unknown nuclides.
- Among tank groups for which the gross beta contribution totals from the primary seven nuclides, C-14 and Tc-99 were lower than gross beta values tanks were selected as investigation targets based on the following criteria.
  1. Tanks that have the largest difference between the absolute values for gross beta values and gross beta contribution totals
  2. Tanks that have the largest gross beta ratio of the difference between gross beta values and gross beta contribution totals
  3. Tanks for which Sr-90 and Y-90 make up the smallest percentage of gross beta contribution totals
- The following investigations were performed for tanks targeted for investigation
  - ① Search for nuclides that have not been qualified through detailed gamma ray spectrometry report review
  - ② Examine beta rays spectrums to search for energy distributions from other beta-emitting nuclides not already detected
  - ③ Qualify and quantify the nuclides if it is determined through ① and ② that unknown nuclides are present.
- Refer to pages 11~13 for the analysis results for each tank group

8. Monitoring results since the third investigation

Radiation concentration unit: Bq/L

Tank	Cs-134	Cs-137	Sr-90	Y-90	Ru-106	Rh-106	I-129	Co-60	Sb-125	C-14	Tc-99	Gross-β contribution (conversion value)	Precipitate weight (mg)	Gross-β contribution (shielding considered)	Gross-β value
G6-C1	<0.20	<0.23	1.1	1.1	<1.8	<1.8	<0.35	0.36	<0.70	26	<1.4	20	38	17	11
G6-A1	<0.32	0.44	1.5	1.5	<1.9	<1.9	<0.35	0.92	<0.66	38	<1.4	28	54	22	8.5
G6-C10	<0.18	<0.26	1.9	1.9	<1.7	<1.7	2.6	0.36	<0.72	25	<1.4	23	54	19	13
G6-A9	<0.40	0.58	31	31	<1.8	<1.8	<0.35	0.45	<0.61	116	<1.4	132	94	104	56
H6(2)-C3	<0.22	0.42	5.1	5.1	<1.9	<1.9	<0.35	1.1	<0.69	57	<1.4	46	57	37	23
H6(2)-C1	<0.23	0.32	313	313	<2.2	<2.2	<0.35	1.1	<0.73	32	<1.4	699	60	672	720
K4-C5	<0.12	0.63	8.0	8.0	<1.3	<1.3	1.3	0.28	<0.40	17	<1.0	30	40	28	19
K4-E1	<0.15	0.59	7.3	7.3	<1.1	<1.1	1.9	0.43	<0.45	14	6.2	32	48	29	31 ※
K4-A1	<0.13	0.16	6.3	6.3	<1.2	<1.2	0.49	0.29	<0.37	14	<1.0	24	47	22	7.4
K4-B1	<0.19	0.47	8.6	8.6	<1.2	<1.2	1.3	0.56	<0.41	18	<1.0	32	46	29	13
J4-L1	<0.16	0.69	11	11	<1.3	<1.3	0.70	0.44	<0.60	21	<1.0	38	58	34	22
J4-C1	<0.20	1.2	12	12	<1.2	<1.2	2.2	<0.15	1.1	5.8	<1.0	35	34	34	25
J7-A1	<0.13	0.81	4.8	4.8	<1.2	<1.2	3.2	0.33	<0.41	14	<1.0	23	70	19	14

: Tanks for which the gross beta contribution was less than gross beta values

※ Tanks for which Sr-90 and Y-90 make up the smallest percentage of converted totals were investigated from amongst tanks for which the gross beta contribution was less than gross beta values.

8. Monitoring results since the third investigation

Radiation concentration unit: Bq/L

Tank	Cs-134	Cs-137	Sr-90	Y-90	Ru-106	Rh-106	I-129	Co-60	Sb-125	C-14	Tc-99	Gross-β contribution (conversion value)	Precipitate weight (mg)	Gross-β contribution (shielding considered)	Gross-β value
H1E-A1	<0.21	0.50	4.4	4.4	<1.2	<1.2	3.8	0.91	<0.45	14	<1.0	23	51	20	13
H2-C1	<0.22	0.87	2.5	2.5	<1.3	<1.3	6.3	0.27	<0.44	59	<1.2	44	43	36	20
H2-B1	<0.29	0.29	3.3	3.3	1.9	1.9	5.9	1.2	<0.46	22	13	36	58	30	26
H2-D1	<0.26	0.45	2.8	2.8	<1.2	<1.2	2.7	0.42	<0.42	14	<1.2	19	46	16	10
H2-J1	<0.18	0.51	2.1	2.1	<1.3	<1.3	2.5	0.56	0.52	39	12	39	94	28	25
J9-A1	<0.20	0.29	2.2	2.2	<1.2	<1.2	0.71	0.48	<0.45	17	<1.2	17	66	14	10
J1-N1	<0.13	1.3	2.0	2.0	<1.3	<1.3	2.2	0.43	<0.45	15	<1.2	18	65	15	12
K1-B1	<0.24	0.26	297	297	<1.3	<1.3	4.9	0.83	3.4	2.5	<1.2	650	22	642	678
K4-D1	<0.21	0.14	2.5	2.5	<1.3	<1.3	2.2	0.50	0.44	13	<1.7	17	46	15	12
G1S-B1	<0.17	0.44	2.4	2.4	<1.3	<1.3	3.0	0.63	0.51	96	5.6	64	65	47	35
G6-D1	<0.14	<0.13	2.2	2.2	<1.3	<1.3	<0.23	0.47	<0.43	24	<1.7	21	53	17	9.7
G6-D6	<0.22	<0.15	1.2	1.2	<1.3	<1.3	1.3	0.43	0.74	48	<1.7	32	57	25	22

[red box]: Tanks for which the gross beta contribution was less than gross beta values

8. Monitoring results since the third investigation

Radiation concentration unit: Bq/L

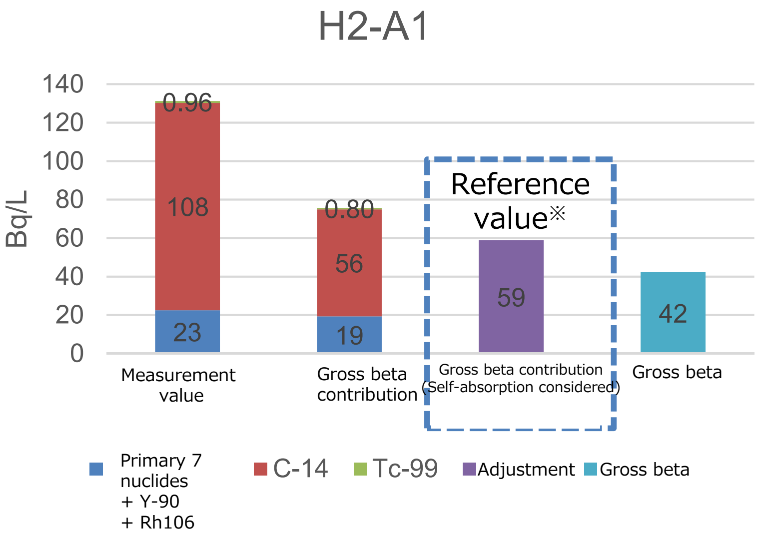
Tank	Cs-134	Cs-137	Sr-90	Y-90	Ru-106	Rh-106	I-129	Co-60	Sb-125	C-14	Tc-99	Gross-β contribution (conversion value)	Precipitate weight (mg)	Gross-β contribution (shielding considered)	Gross-β value	
B-B1	<0.14	<0.13	1.1	1.1	<1.2	<1.2	<0.23	0.43	<0.45	24	<1.7	18	55	15	11	
B-B5	<0.16	<0.12	4.1	4.1	<1.1	<1.1	<0.23	0.37	<0.31	32	<1.7	29	53	24	18	
B-A1	<0.43	1.3	9230	9230	<3.0	<3.0	52	0.69	2.7	16	5.8	20000	90	19000	20300	
B-A5	<0.30	0.48	2490	2490	<1.5	<1.5	54	0.66	2.0	15	5.9	5430	96	5150	5910	
B-D7	<0.70	3.0	22600	22600	<4.6	<4.6	45	0.43	4.8	14	5.9	48800	110	46000	52800	※1
B-D6	<0.50	2.2	17100	17100	<3.6	<3.6	47	0.43	2.8	14	5.5	36900	97	35000	40400	※2
B-D5	<2.0	2.8	11200	11200	<17	<17	47	<1.3	<6.2	16	5.2	24300	91	23100	24400	
B-D4	<1.5	1.5	9260	9260	<12	<12	48	<1.2	4.9	13	5.0	20100	82	19200	20200	
B-D3	<0.38	0.92	5920	5920	<2.7	<2.7	48	0.49	2.5	11	5.1	12800	77	12300	13700	
B-D2	<0.47	1.1	6100	6100	<3.1	<3.1	42	0.59	2.4	9.5	4.9	13200	81	12600	14200	
B-D9	<0.48	2.1	14200	14200	<3.4	<3.4	46	0.65	3.0	13	5.1	30800	86	29400	32700	
B-D8	<0.61	1.9	14200	14200	<4.2	<4.2	35	0.38	1.8	12	4.3	30800	85	29400	30200	
H2-E1	<0.19	0.34	2.0	2.0	<1.2	<1.2	4.2	0.21	0.50	17	18	33	56	27	19	

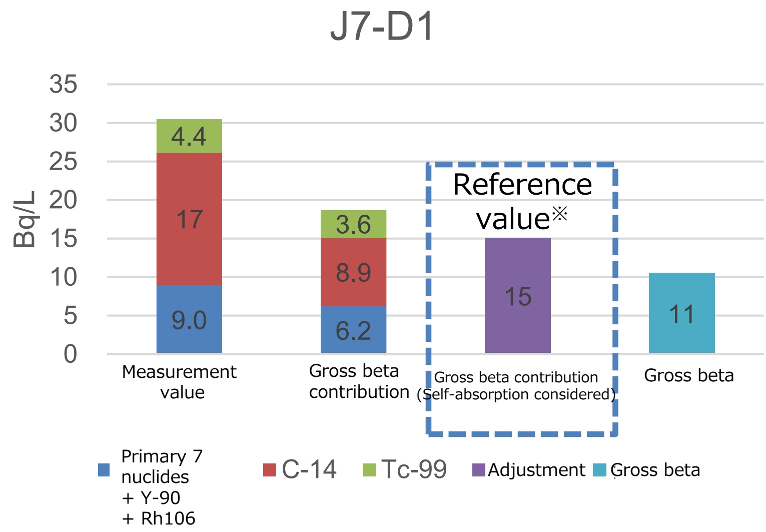
(red box) : Tanks for which the gross beta contribution was less than gross beta values

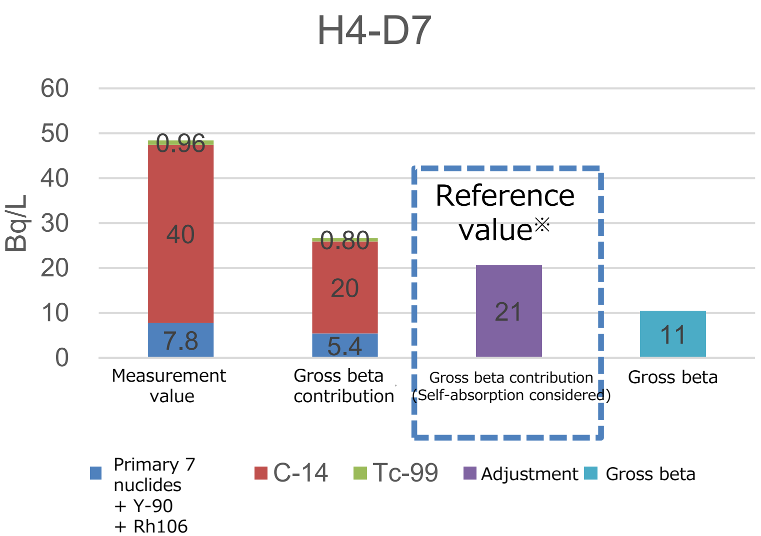
※1 Tanks that have the largest difference between the absolute values for gross beta values and converted totals were investigated from amongst tanks for which the gross beta contribution was less than gross beta values.

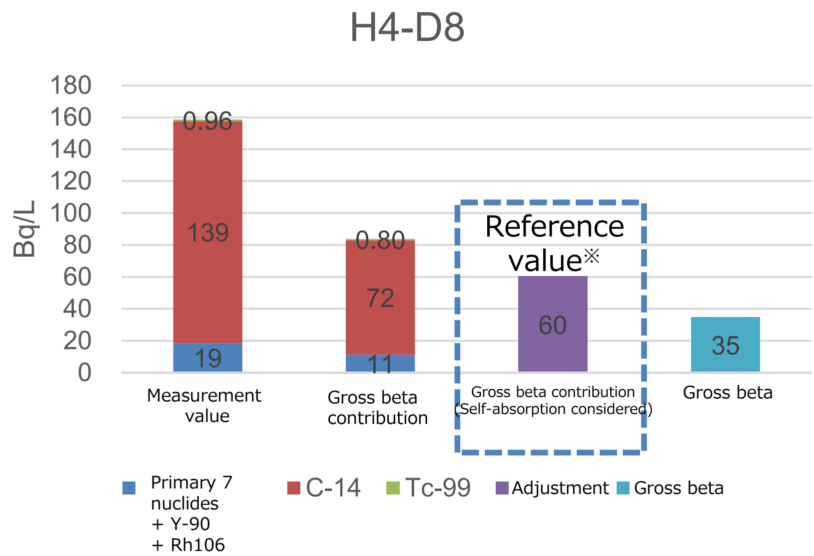
※2 Tanks that have the largest gross beta ratio of the difference between gross beta values and converted totals were investigated from amongst tanks for which the gross beta contribution was less than gross beta values.

<Reference> Results of the third investigation









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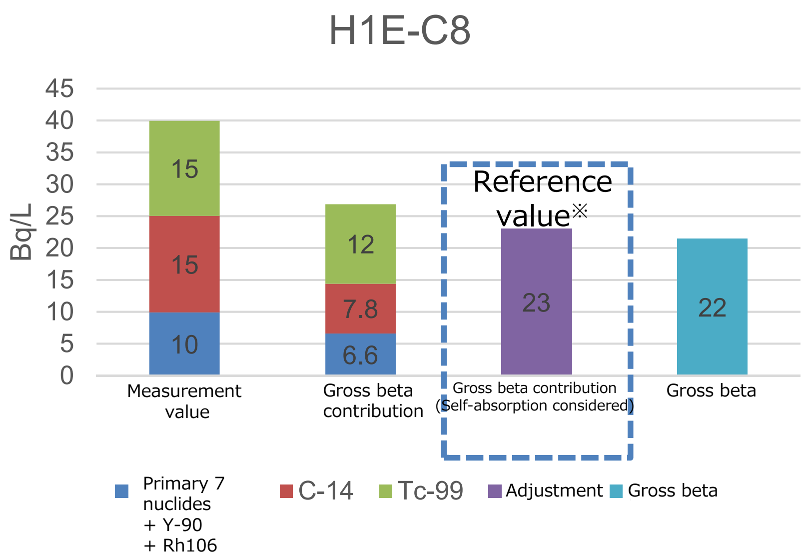

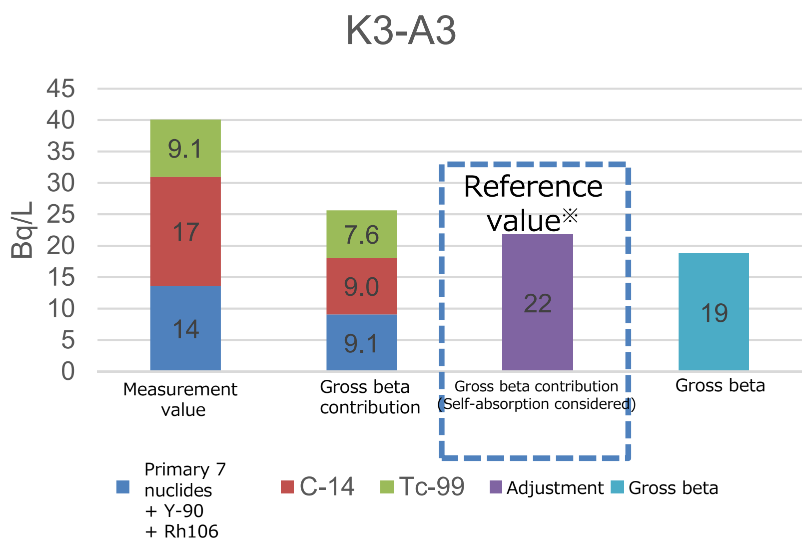

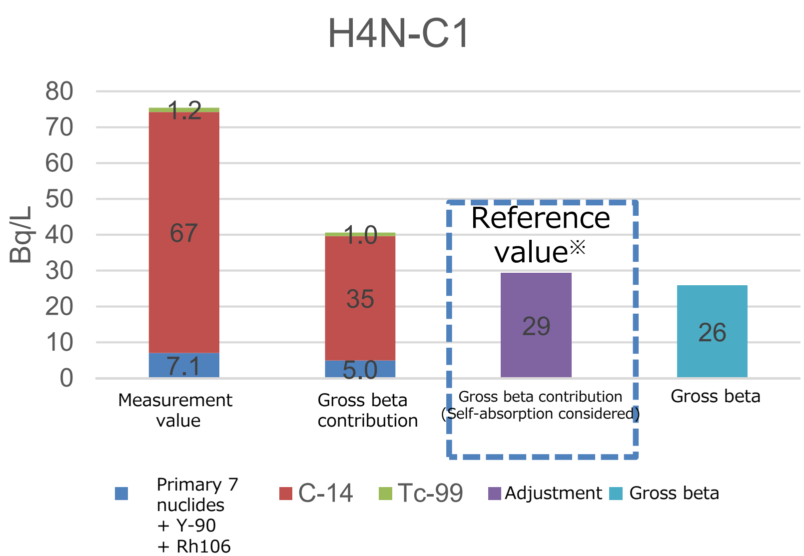

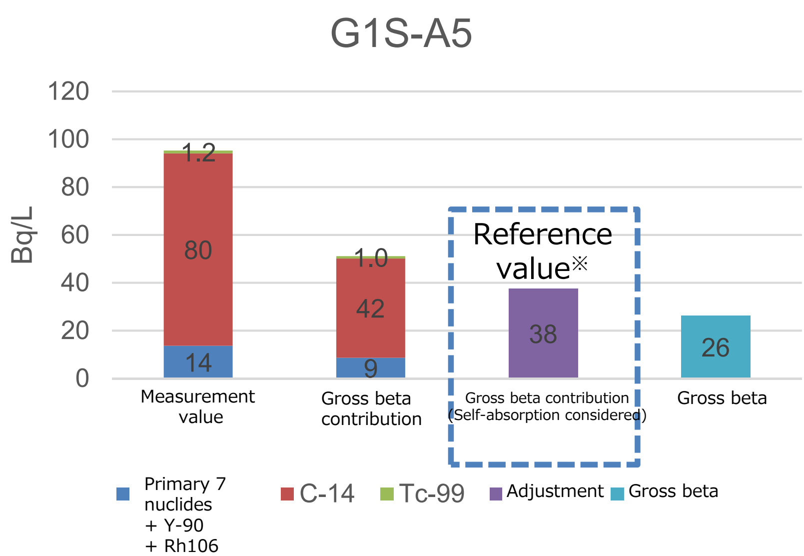

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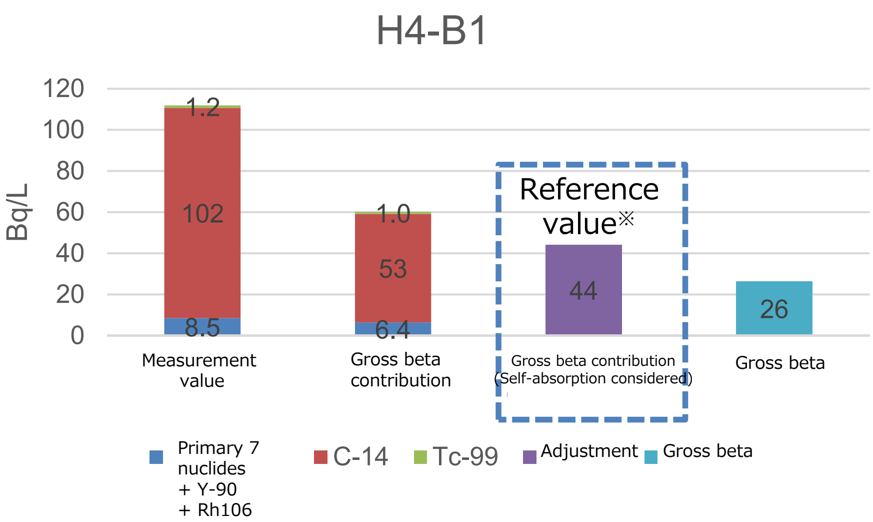

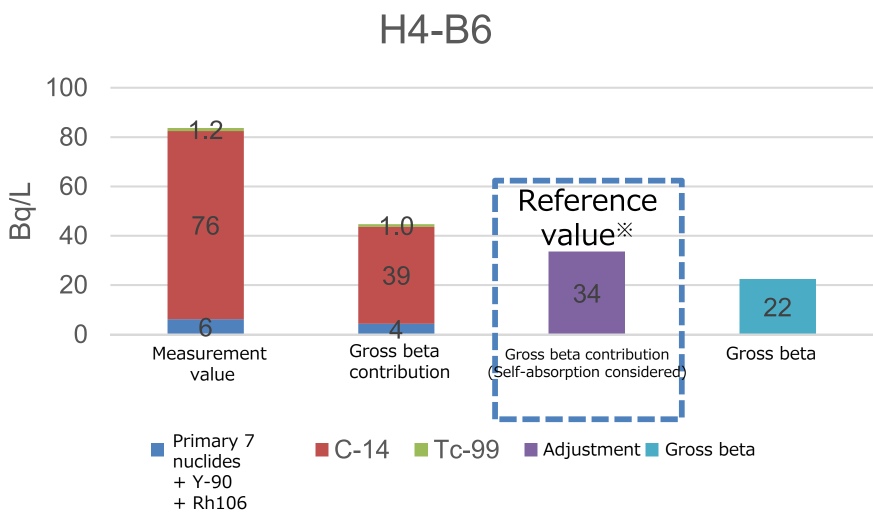

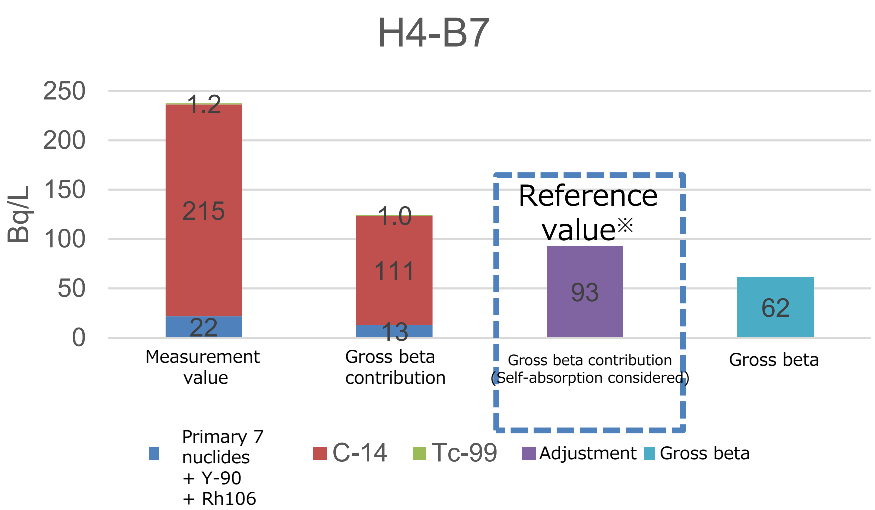

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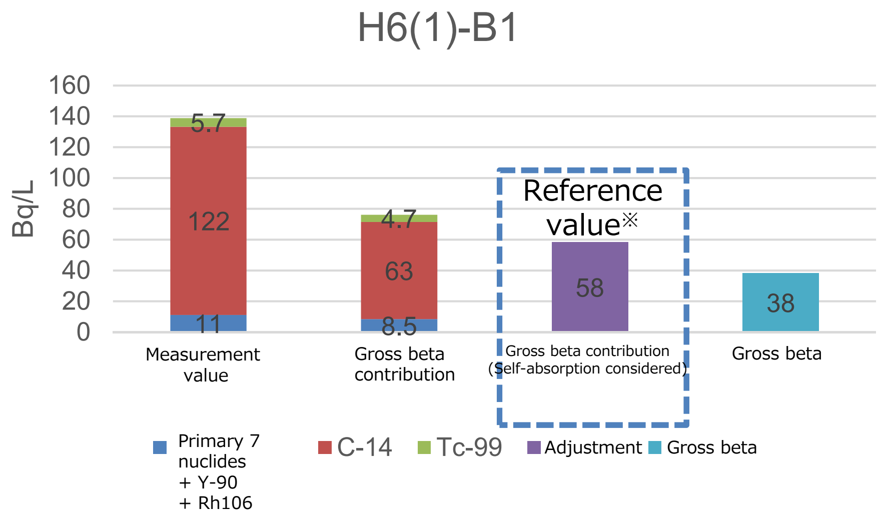

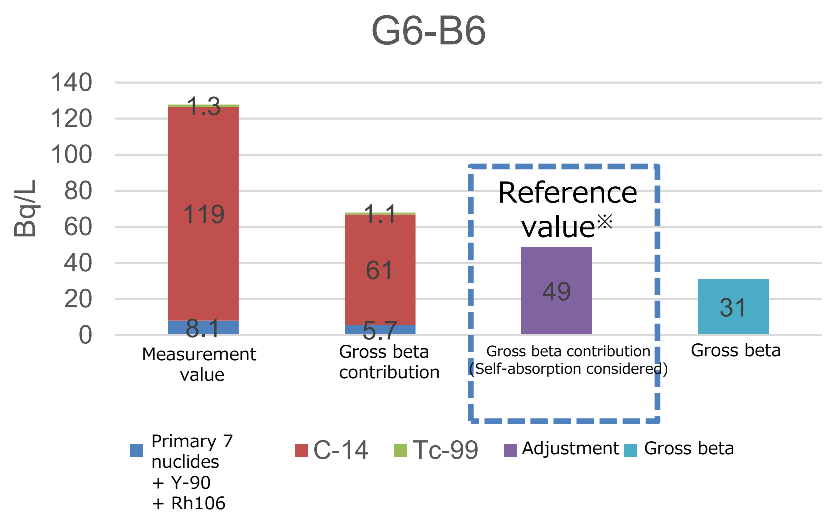

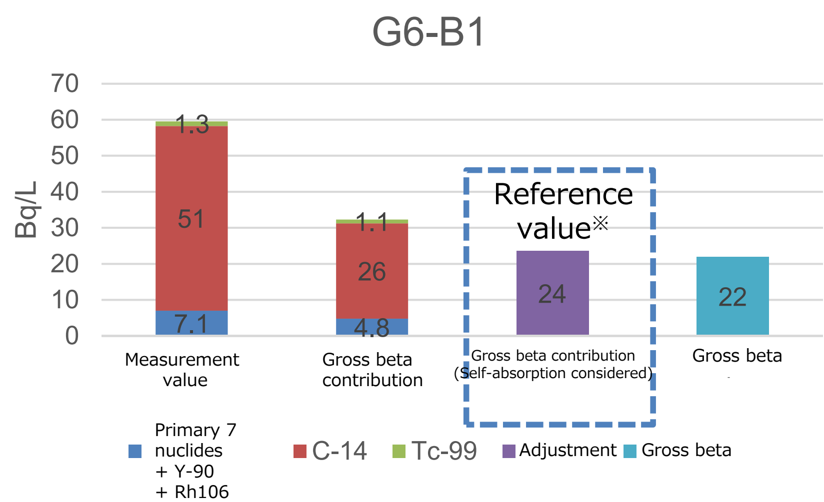

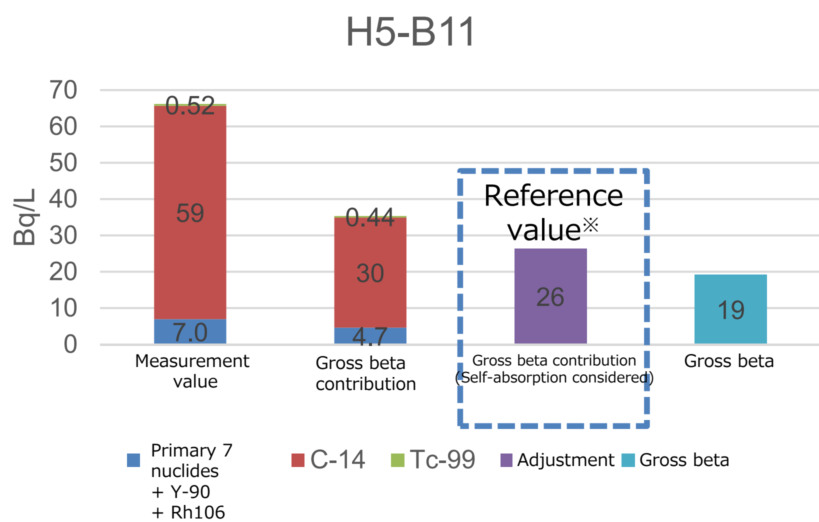

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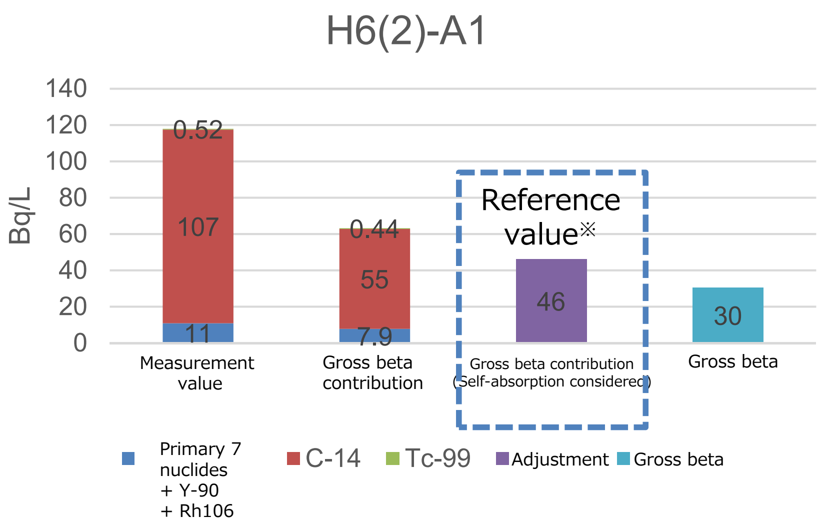
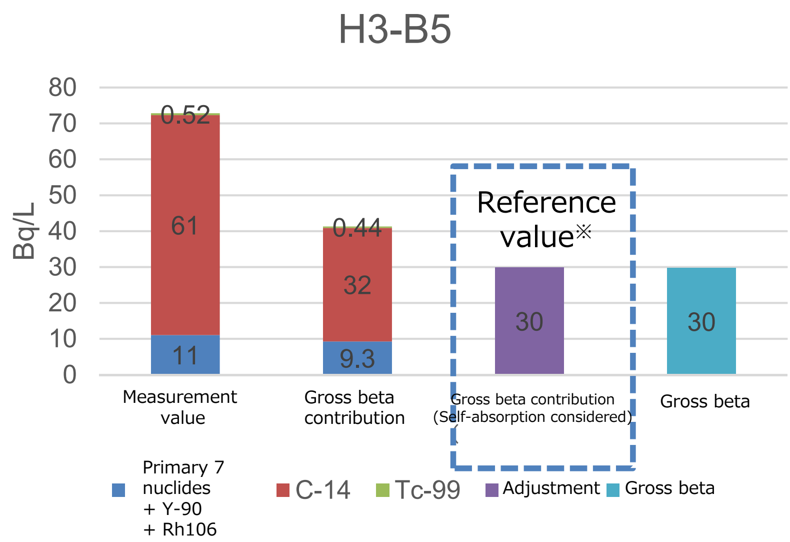
<Reference> Results of the third investigation





※Gross beta contribution (self-absorption considered) values were 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.

<Reference> Results of the third investigation

Radiation concentration unit: Bq/L

Selected Tank	Cs-134	Cs-137	Co-60	Ru-106	Rh-106	Sb-125	Sr-90	Y-90	I-129	C-14	Tc-99	Gross-β contribution (conversion value)	Precipitate weight (mg)	Gross-β contribution (shielding considered)	Gross-β
H2-A1	<0.43	<0.25	0.23	<1.7	<1.7	<0.66	6.2	6.2	5.2	108	<1.0	76	56.78	59	42
J7-D1	<0.24	<0.25	0.95	<1.4	<1.4	<0.45	0.75	0.75	2.8	17	4.4	19	57.16	15	11
H4-D7	<0.18	0.31	0.49	<1.4	<1.4	<0.48	0.69	0.69	2.2	40	<1.0	27	52.51	21	11
H4-D8	<0.21	<0.20	1.3	<1.4	<1.4	0.81	<0.40	<0.40	13	139	<1.0	84	60.07	60	35
H1E-C8	<0.22	0.47	0.86	<2.0	<2.0	<0.64	<0.51	<0.51	2.8	15	15	27	47.58	23	22
K3-A3	<0.39	1.3	0.51	<2.8	<2.8	<1.1	<0.47	<0.47	3.8	17	9.1	26	48.40	22	19
H4N-C1	<0.15	<0.24	1.6	<1.4	<1.4	<0.46	<0.42	<0.42	1.0	67	<1.2	41	64.37	29	26
G1S-A5	<0.19	0.34	1.3	<1.4	<1.4	<0.48	<0.50	<0.50	7.6	80	<1.2	51	59.44	38	26

<Reference> Results of the third investigation

Radiation concentration unit: Bq/L

Selected Tank	Cs-134	Cs-137	Co-60	Ru-106	Rh-106	Sb-125	Sr-90	Y-90	I-129	C-14	Tc-99	Gross-β contribution (conversion value)	Precipitate weight (mg)	Gross-β contribution (shielding considered)	Gross-β
H4-B1	<0.20	0.40	2.1	<1.5	<1.5	<0.47	0.81	0.81	0.80	102	<1.2	60	60.17	44	26
H4-B6	<0.15	0.44	0.70	<1.2	<1.2	<0.43	<0.40	<0.40	1.3	76	<1.2	45	55.20	34	22
H4-B7	<0.17	<0.24	0.70	<1.2	<1.2	0.56	<0.39	<0.39	17	215	<1.2	125	51.78	93	62
H6(1)-B1	<0.13	0.70	2.9	<1.3	<1.3	<0.42	1.1	1.1	2.3	122	5.7	76	52.45	58	38
G6-B6	<0.28	0.22	1.7	<1.2	<1.2	<0.44	0.55	0.55	1.9	119	<1.3	68	62.07	49	31
G6-B1	<0.16	<0.23	0.94	<1.3	<1.3	<0.47	<0.45	<0.45	1.8	51	<1.3	32	63.45	24	22
H5-B11	<0.12	<0.20	0.68	<1.2	<1.2	<0.39	0.41	0.41	2.3	59	<0.52	35	56.11	26	19
H6(2)-A1	<0.24	<0.23	1.3	<1.3	<1.3	<0.46	1.2	1.2	3.7	107	<0.52	63	60.40	46	30
H3-B5	<0.27	0.44	1.7	<1.2	<1.2	<0.39	2.3	2.3	1.4	61	<0.52	41	71.94	30	30