<Reference Material>
December 1, 2022
TEPCO Holdings
Fukushima Daiichi D&D Engineering Company

Fukushima Daiichi Nuclear Power Station
Examining the tritium analysis values of fish during sea area monitoring in preparation for ALPS treated water discharge into the sea

- TEPCO published TEPCO holding's action in response to the Japanese government's policy on the handling of ALPS treated water on April 16, 2021. The response includes strengthening and expansion of its sea area monitoring efforts to minimize adverse impact on reputation that may be sustained a result of discharging ALPS treated water into the sea.
- TEPCO, as the organization responsible for discharging ALPS treated water, evaluated how the treated water will disperse based using dispersion simulations at sea, and developed a sea area monitoring plan for measuring tritium levels to check on dispersion *2in seas off the coast of Fukushima near the station where tritium concentrations will likely increase after discharge.*1. This was made public on August 25, 2021.
- Based on the government's more robust comprehensive monitoring plan, TEPCO has created sea area monitoring plan that stipulates the lowest detection values for tritium based upon the results of the August 25, 2021 deliberation (increase the number of measurement points, specimens to be measured, and measurement frequency), and put this plan into use in April of this year in order to continually examine the dispersion of tritium and how it is impacting marine organisms.

< Announced as of May 26, 2022 >

Background

- According to the government's comprehensive monitoring plan, though tritium concentrations in fish was to be measured once a month at one location, TEPCO has added 10 more measurement locations for a total of 11, and analysis of measurements taken from these added 10 locations began in May of this year using official methods (Radiation Measurement Methods Series of the Ministry of Education, Culture, Sports, Science and Technology) by TEPCO and Kaken Co., Ltd. (hereinafter referred to as, "Kaken"), to which this task has been outsourced by TEPCO. (The Kyushu Environmental Evaluation Association (hereinafter referred to as, "KEEA") will continue to analyze the measurement results from the existing one location)

- As this was the first time that TEPCO and Kaken analyzed tritium concentrations in fish, we compared them with the analysis results from the KEEA. As a result, in August, we confirmed that the concentrations of free water tritium (hereinafter referred to as, "FWT") analyzed by TEPCO, and the concentrations of organically bound tritium (hereinafter referred to as, "OBT") analyzed by both TEPCO and Kaken were higher than tritium concentrations in the surrounding seawater. We also found that the analysis results from the KEEA were approximately the same as they have always been (OBT concentrations were below detectable levels).

- Therefore, we enlisted the help of experts apart from the three parties mentioned above to investigate the cause of the discrepancy in analysis results between TEPCO, Kaken and the KEEA (discrepancies in the analysis methods of all three parties).

Investigation results

- The analysis methods of the KEEA and Kaken were observed directly, and the analysis methods of all three parties, including TEPCO, were compared. Upon identifying factors that impact analysis results, the methods were examined while asking experts apart from the three parties opinions. The following conclusions were drawn.
  - Discrepancies in methods for removing impurities
    - When comparing impurity (organic material) removal methods, it was found that compared to TEPCO and Kaken, the KEEA took more time to remove impurities and added more reagent. When analyzing tritium, correct measurements cannot be obtained if impurities have not been adequately removed.
  - Discrepancies in the amount of time that specimens are left to stand after addition of luminescent liquid (impact of chemical reactions)
    - When the amount of time that specimens are left to stand after addition of the luminescent liquid (liquid scintillator) were compared, it was found that TEPCO did not leave the specimens stand as long as other analysis parties. When analyzing tritium, after removing impurities a luminescent liquid is added, and if the specimens are not left to stand along enough for the chemical reaction to conclude, correct measurements cannot be obtained.
- Based on these results, TEPCO and Kaken used the same method employed by the KEEA to remove impurities and measured/compared tritium concentrations once again.
  - Kaken: Impurities were removed just like the KEEA and OBT concentrations were below detectable levels.
  - TEPCO: Impurities could not be completely removed, and OBT levels were much higher than actual concentrations.
- After confirming that the KEEA's analysis methods are scientifically-based and logical, Kaken formulated new analysis procedures based on the KEEA's analysis methods and recommenced analysis on October 24. It was confirmed there after that OBT concentrations are below detectable levels. In light of the fact that the analysis results from the KEEA and the analysis results from Kaken after changes were made to their analysis method both show OBT concentrations to be below detectable levels, it has been deemed that actual OBT concentrations are below detectable levels and that the analysis method currently employed by TEPCO yields OBT concentration results that are much higher than actual levels.

Steps going forward

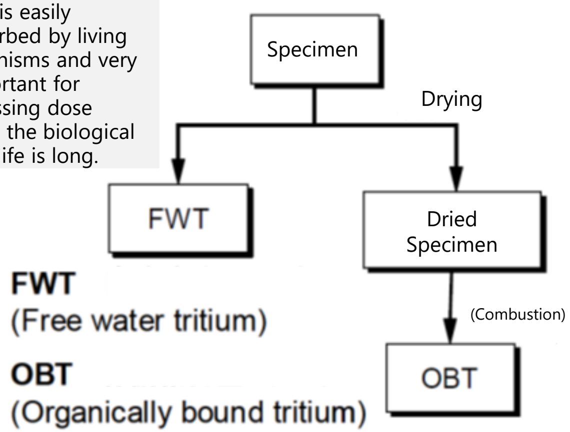
- Kaken will continue to measure tritium concentrations in fish.
- Since extremely small concentrations of tritium are being measured during fish tritium analysis, we are being very careful not to contaminate other specimens. Since TEPCO’s analysis is being conducted within controlled area, we will continue to closely examine our methods for removing impurities as well as investigate the possibility that specimens are being contaminated by tritium from the surrounding environment.

1． Background

- As a result of adding additional measurement locations and targets to the sea area monitoring plan regarding the discharge of ALPS treated water, which was announced on March 24 of this year, the number of locations from which specimens for analyzing the concentration of tritium in fish will be sampled has increased from one to 11. (The KEEA will continue to analyze tritium concentrations from the one location stipulated in the original sea area monitoring plan)
- According to the monitoring plan, the results from each agency engaged in monitoring shall be compared to confirm validity, and accordingly, since TEPCO and Kaken Co., Ltd. (hereinafter referred to as, "Kaken") have analyzed tritium concentrations in fish for the first time, the analysis was conducted based on official methods (Radioactivity Measurement Methods Series) and the results were compared with the results from the KEEA, which has already produced a substantial amount of analysis results.
- After commencing the analysis of tritium concentrations in fish from 11 locations in May of this year, we confirmed that the concentrations of free water tritium (hereinafter referred to as, "FWT") analyzed by TEPCO, and the concentrations of organically bound tritium (hereinafter referred to as, "OBT" ※Refer to the diagram below) analyzed by both TEPCO and Kaken were higher than tritium concentrations in the surrounding sea water. The analysis results from TEPCO and Kaken differed from those from the KEEA, which were approximately the same as they have always been.
- Therefore, TEPCO and Kaken have temporarily suspended analysis since August when the aforementioned analysis results from TEPCO and Kaken were compared with those from the KEEA, and the cooperation of experts apart from these three parties was elicited to perform an investigation into the cause of the discrepancies in analysis methods.

Measuring tritium in organic substances

OBT is easily absorbed by living organisms and very important for assessing dose since the biological half-life is long.



Pre-treatment equipment

- When measuring tritium concentrations in fish, two types are measured, FWT and OBT.
- FWT refers to the concentration of tritium in the water inside the fish, so the water in the fish is evaporated, recovered and then measured.
- OBT is tritium that exists as part of hydrogen atoms in fish tissue (protein tissue, such as muscles, etc.), so the water generated during the burning of this tissue is recovered and measured. Since impurities that could impact measurements migrate into the specimens during combustion, they must be removed from the recovered water.
- Fish tritium measurement is very difficult and there are only a few analysis agencies that are capable of performing this measurement.

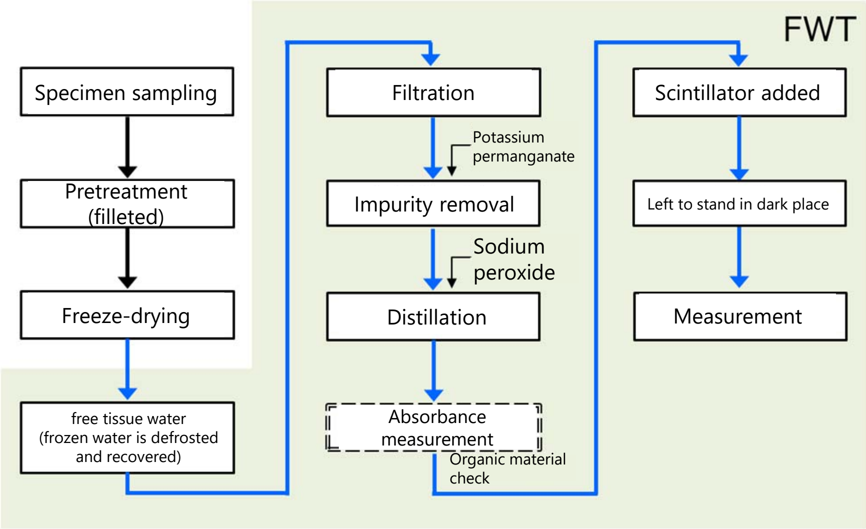
※ Excerpt from reference document 3: "The Environmental Dynamics of Tritium (^{3}H) (Institute for Environmental Sciences; Hideki Kakiuchi) in the Tritiated Water Task Force Report (June 2016 Tritiated Water Task Force)

2． Fish tritium analysis method

■ When measuring tritium concentrations in fish, two types are measured, FWT and OBT.

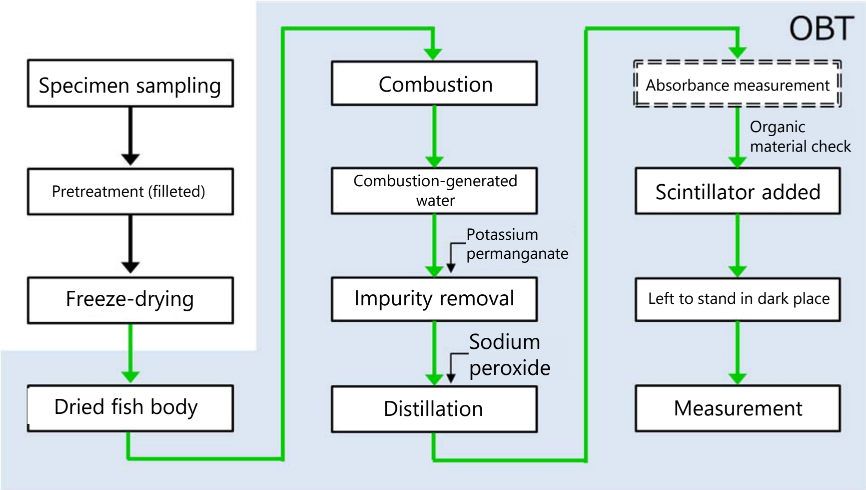
<FWT analysis method>

- After the parts of the specimen that are subject to analysis are harvested, they are frozen in a freezer and then the frozen specimens are freeze-dried after which water (free tissue water) is extracted and an analysis specimen is prepared.
- After this, an oxidant is added to the remaining organic material in the free tissue water (analysis specimen) to dissolve it, the water is distilled, and the specimen is measured using a liquid scintillation counter.



<OBT analysis method>

- Dry tissue that remains after freeze-drying is burned, and the vapor (combustion-generated water) generated is recovered and prepared as an analysis specimen.
- After this, an oxidant is added to the organic material that remains in the combustion-generated water (analysis specimen) to dissolve it (remove impurities), the water is distilled, and the specimen is measured using a liquid scintillation counter.



3． Factors that affect fish tritium analysis

- There are many more factors that affect fish tritium analysis compared to sea water tritium analysis.
- In addition to factor analysis, analysis methods employed by the KEEA and Kaken were directly observed, and the analysis methods of all three parties, including TEPCO, were compared. Experts apart from the three parties were also asked for their opinion during the process of factor identification in the course of the investigation.
- As a result, we identified "the impact of measurement equipment," "the impact of impurities (organic material)," and "the impact of chemical reactions," as factors that affect analysis results.

① The impact of measurement equipment

- Since the performance and installation environments of measurement devices differ, the differences in equipment may have an impact on analysis results.

② The impact of impurities (organic material)

- The concentration of tritium in fish is the same as the concentration in the seawater, and if the fish analysis contains even a few impurities, the concentration of tritium cannot be measured accurately. As a result of differences in pretreatment processes, it is possible that impurities are not being adequately removed. (Deliberation of pretreatment processes)

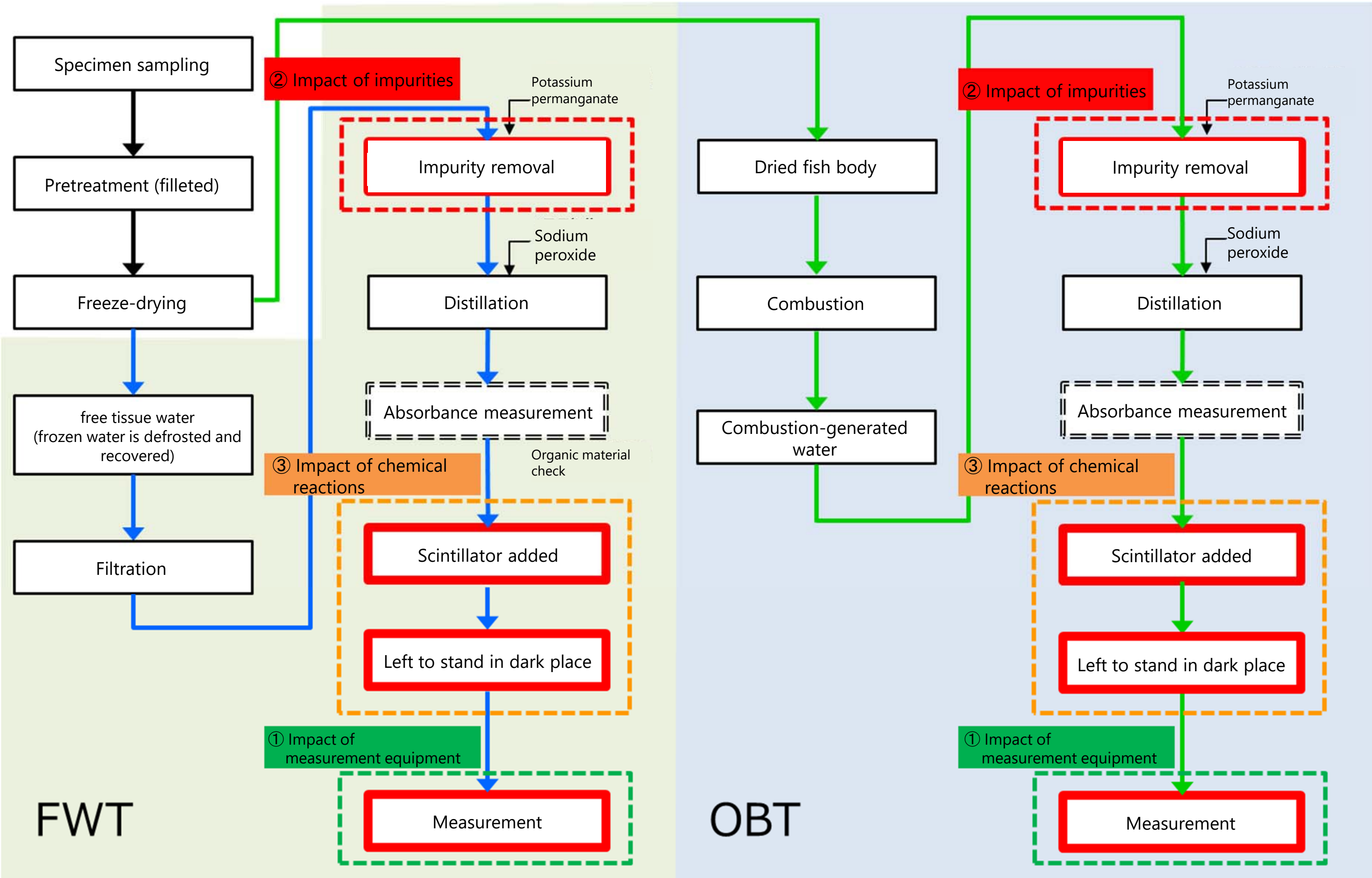
③ The impact of chemical reactions

- Since tritium cannot be directly measured, the radiation must be converted into light that can be measured. However, due to the differences in the way the specimen is left to stand, it is possible that the specimen may remain luminescent due to chemical reactions that have yet to conclude. (Deliberation of measurement stages)

3. Factors that affect fish tritium analysis

TEPCO

- Factors identified this time
  - ① The impact of measurement equipment ② The impact of impurities (organic material)
  - ③ The impact of chemical reactions



4． Verification method

TEPCO

- The following examination of the impact of measurement equipment, impurities (organic material) and chemical reactions was conducted while listening to the opinions of experts apart from the three parties.
- The same examination was conducted at TEPCO and Kaken.

① The impact of measurement equipment

- The same piece of equipment (LSC-LB-8) located at TEPCO (1F on-site analysis facility), off-site facilities, and Kaken was used to measure an empty measurement vessel (no specimen) at the same time on the same day and under the same conditions in order to check for any impact from differences in measurement equipment.

② The impact of impurities (organic material)

- TEPCO (1F on-site analysis facility) and Kaken performed analysis using the pretreatment process of the KEEA (impurity removal). Results after the pretreatment process conducted at each analysis facility were compared to check for any impact from impurities (organic material).

③ The impact of chemical reactions

- TEPCO (1F on-site analysis facility) and Kaken took chronological measurements of measured specimens in which OBT had already been detected to check changes over time and checked for any impact from chemical reactions.

5. Results from verification of measurement equipment

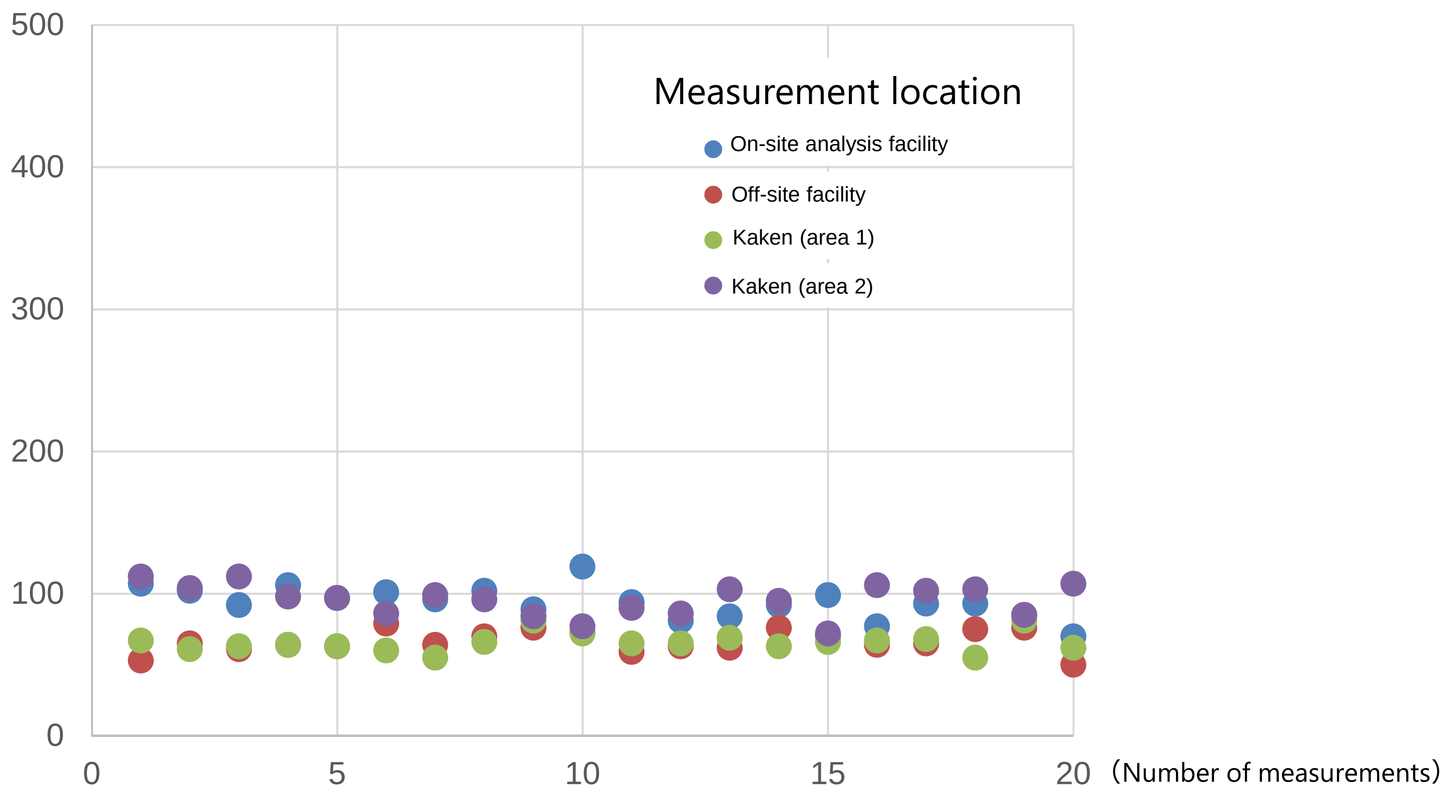
TEPCO

- The same piece of equipment (LSC-LB-8) located at TEPCO (1F on-site analysis facility), off-site facilities, and Kaken was used to measure an empty measurement vessel (no specimen) at the same time on the same day and under the same conditions.
- There was no significant difference in the data from each of the four facilities that repeatedly conducted the same test.
- Therefore, it was determined that there is no impact from any difference in measurement equipment.

(Numerical value※)

※ Number of Radiation Measured

The graph gives numerical values for the range of energy measured from tritium



6. Results from investigation into impurities (organic material)

TEPCO

- Impurities (organic material) are removed based on official methods (Radioactivity Measurement Methods Series). The official methods stipulate the type and amount of reagent that must be added in order to remove [impurities], however it is also noted to, "adjust the amount of reagent added appropriately in accordance with the amount of organic material in the specimen," so the party taking the measurements must ascertain the appropriate amount of additive to be used in accordance with the attributes of the fish to be measured, etc.

- When a check was performed to see if the impurities had been removed as noted on the next page, it was discovered that impurities were better removed using the method employed by the KEEA compared to the methods employed by TEPCO and Kaken. When the methods were compared, it was found that the KEEA took more time to remove impurities and added more reagent. Through this discovery we learned that the method employed by TEPCO and Kaken did not allow enough time for the chemical reaction to completely remove impurities.

Comparison of official analysis methods employed by KEEA/Kaken/TEPCO (1F on-site analysis facility)

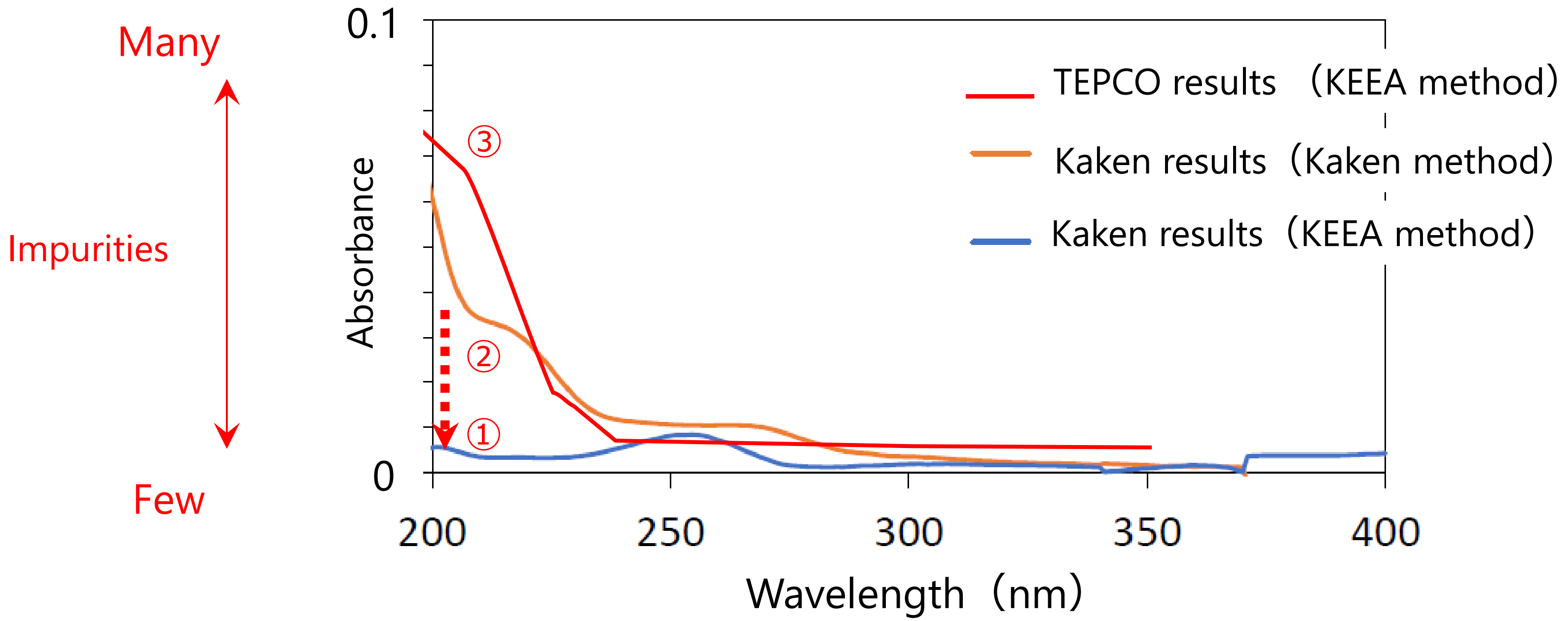
Analysis agency	KEEA	Kaken	TEPCO (1F on-site analysis facility	Official method※
Impurity removal method				
Reagent	Potassium permanganate (Approx. 1g added)	Potassium permanganate (Approx. 1g added)	Potassium permanganate (Approx. 0.3g added)	Potassium permanganate (Approx. 0.5g added per approx. 70ml of analysis specimen)
Temp.	100℃	100℃	60℃	100℃
Time	7 hours/day for 4 days	8 hours	More than 6 hours	More than 4 hours

※Radioactivity Measurement Method Series 9 Tritium Analysis Methods (2002 Revision) MEXT

6. Results from investigation into impurities (organic material)

TEPCO

- In order to examine any impact from the differences in impurity removal methods, TEPCO and Kaken both removed impurities using the same method employed by the KEEA and then measured/compared absorbance (the lower the value, the fewer the impurities), which is the standard for determining if there are residual impurities.
- Kaken found that by using the KEEA method absorbance near the 200nm range decreased greatly compared with the conventional method thereby confirming that the removal of impurities (decrease in residuals) is being promoted. (②→① in the graph below)
- When this method was tested during the analysis of fish specimens, Kaken found that it was able to eliminate the impact of chemical luminescence caused by impurities and that OBT fell below detectable levels. It also confirmed that the impurity dissolution method employed by KEEA is effective for preventing chemical luminescence originating from residual impurities.
- However, when TEPCO employed the method used by the KEEA, it was unable to satisfactorily decrease absorbance and was not able to obtain the anticipated improvement in results. (③ in the graph below)



7. Results from investigation into chemical reactions

TEPCO

- After dissolving impurities and distilling the water, a luminescent liquid (liquid scintillator) was added and measurements were taken.
- According to official methodology (Radioactivity Measurement Methods Series) it is recommended that the specimen be left to stand for approximately a whole day to one week after addition of the liquid scintillator.
- A comparison of the time that each party left the specimens stand after addition of the additive showed that the KEEA and Kaken left the specimens data for one week, while TEPCO left it stand for more than three days, which is based on its experience with analyzing seawater, etc.
- Since TEPCO was not leaving the specimens stand as long as other analysis agencies, it is possible that it was not allowing enough time to eliminate chemical reactions.

Comparison of official analysis methods employed by KEEA/Kaken/TEPCO (1F on-site analysis facility)

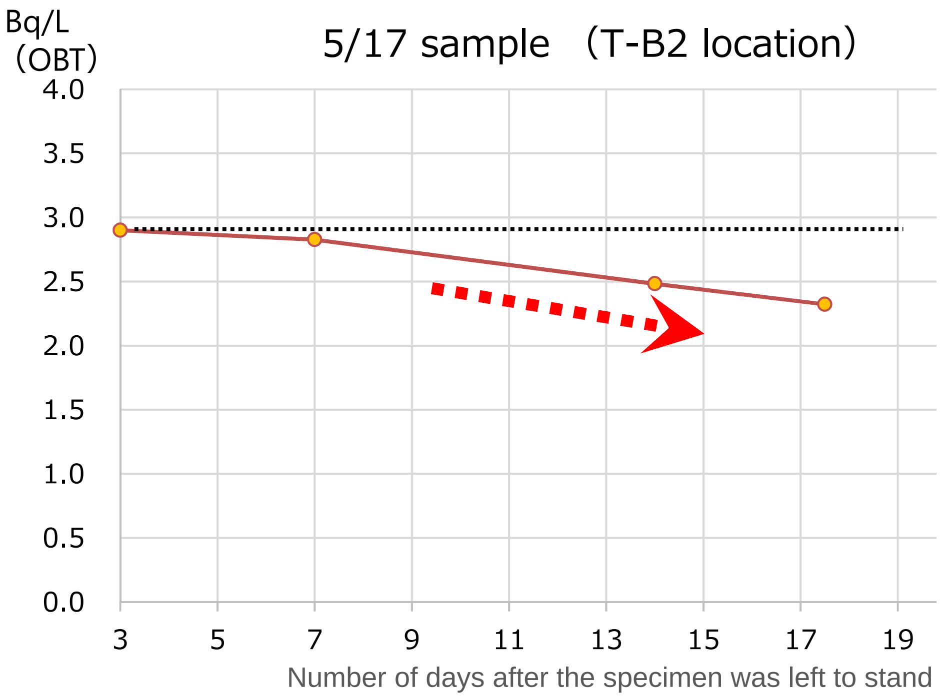
Analysis agency	KEEA	Kaken	TEPCO (1F on-site analysis facility)	Official method※
Measurement equipment	LSC-LB-5, LB-7 (Made by Nippon RayTech Co., Ltd.)	LSC-LB-7, LB-8 (Made by Nippon RayTech Co., Ltd.)	LSC-LB-7, LB-8 (Made by Nippon RayTech Co., Ltd.)	Liquid scintillation counter
Liquid scintillator	Ultima Gold uLLT	Ultima Gold LLT	Ultima Gold LLT	Emulsification scintillator (Commercial products such as Scintisol and Ultima Gold LLT, etc.)
Amount of time left to stand after liquid scintillator is added to the specimen	One week	One week	More than three days	A whole day and approximately one week and after specimen adjustment
Environment in which the specimen is left to stand	20℃ (Air-conditioning temperature) Dark location	15℃ Inside measurement equipment (dark location)	15℃ Inside measurement equipment (dark location)	Inside of the measurement equipment or in an incubator set to approximately 16° C

※Radioactivity Measurement Method Series 9 Tritium Analysis Methods (2002 Revision) MEXT

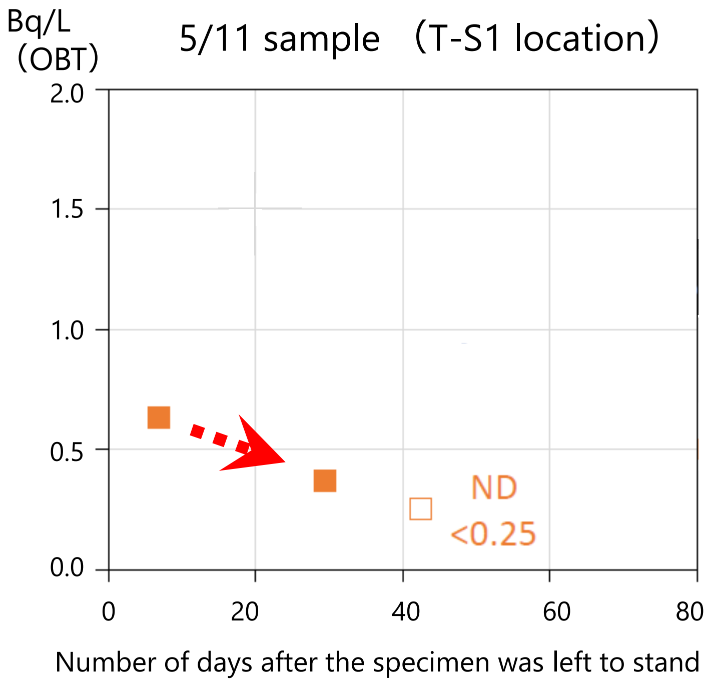
7． Results from investigation into chemical reactions

TEPCO

- When Kaken and TEPCO used a specimen in which OBT had been detected to perform chronological measurements, it was confirmed that analysis values decreased because chemical reactions were allowed to conclude.
- Therefore it was determined that chemical reactions that occur in conjunction with the addition of the liquid scintillator had not been allowed to run their course, and that measurements had been taken without allowing the specimen to stand for a sufficient amount of time.
- When using the aforementioned specimens, it is true that chemical luminescence from residual impurities had a great impact, but this was compounded by the impact of chemical luminescence from ongoing chemical reactions.



TEPCO investigation results



Kaken investigation results

8． Kaken initiatives

TEPCO

- Based on the investigation results, Kaken determined that the method employed by the KEEA is scientifically-based and logical, and will now employ the method for removing impurities used by the KEEA during OBT analysis (pretreatment). When this method was tested during the analysis of fish specimens, Kaken found that it was able to eliminate the impact of chemical luminescence caused by impurities, and that OBT fell below detectable levels.
- Kaken created new analysis procedures based on the method employed by the KEEA and when analysis of fish tritium recommenced on October 24, OBT results showed that tritium concentrations were below detectable levels.

Kaken analysis method (After revision)

Analysis agency	Kaken（After revision）	KEEA
Impurity removal method		
Reagent	Potassium permanganate (Approx. 1g added)	Potassium permanganate (Approx. 1g added)
Temperature	100℃	100℃
Time	More than 28 hours	7 hours/day for four days
Method for confirming residual impurities		
Absorbance	<0.1(near 200nm)	<0.1(near 200nm)
Conductivity rate	<10μS/cm	<10μS/cm
pH	Neutral	Neutral

9． TEPCO initiatives

- TEPCO's analysis results differed from those by the KEEA and Kaken, and determined that OBT measurements showed higher values than the actual amount of OBT.
- Since extremely small concentrations of tritium are being measured during fish tritium analysis this time, we are being very careful not to contaminate specimens. However, since analysis is being conducted within TEPCO's controlled area, we will continue to closely examine our methods for removing impurities as well as investigate the possibilities that specimens are being contaminated by tritium from the surrounding environment.
  - The KEEA impurity removal method will be used in an analysis area within the on-site analysis facility that is assumed to result in minimal tritium contamination, and the specimen will be left to stand for the same amount of time that the KEEA employs after which test analysis of fish specimens will be conducted. (Action 1)
  - After these tests have been concluded, the measurement results will be compared with fish specimens on trial by TEPCO and Kaken, and the skills of analysis personnel shall also be reviewed. (Action 2)
- Kaken will analyze specimens from the TEPCO’s five locations while the investigation is ongoing.

Using the impurity (organic material) removal device

[Schedule for recommencing analysis by TEPCO]

・Action 1: To be implemented in December

・Action 2: To commence as soon as preparations have been completed after the completion of action 1

【Reference】Data from all three parties

- Originally, analysis results from TEPCO (1F on-site analysis facility) and Kaken showed higher concentrations of tritium then in the surrounding seawater. (Chart 1 in red)
- Analysis results from the KEEA were approximately the same as they have always been (OBT concentrations were below detectable levels). (Chart 2)
- After employing the same methodology as the KEEA, OBT levels measured by Kaken also dropped to below detectable levels. (chart 3)

Chart 1 Data confirmed after the start of the analysis by TEPCO and Kaken

Sampling location	Sampling date	Fish type	Concentrations in surrounding sea water (Bq/L)	Free water tritium (Bq/L)	Organically bound tritium (Bq/L)	Analysis location
T-B1	May 17	Flounder	ND(0.33)	0.48	1.2	TEPCO
T-B1	June 21	Flounder	ND(0.33)	ND(0.23)	0.40	TEPCO
T-B2	May 17	Slime flounder	ND(0.32)	0.58	2.9	TEPCO
T-B2	May 24	Flounder	ND(0.34)	0.45	0.74	TEPCO
T-B2	June 21	Marbled sole	ND(0.33)	ND(0.23)	–※1	TEPCO
T-B3	May 31	Flounder	ND(0.33)	0.45	0.58	TEPCO
T-B4	May 31	Flounder	ND(0.33)	0.42	0.87	TEPCO
T-S1	May 11	Flounder	0.077	0.11	0.63	Kaken
T-S2	May 11	Flounder	ND(0.065)	ND	0.96	Kaken
T-S3	May 12	Flounder	0.067	0.11	0.94	Kaken
T-S4	May 12	Flounder	ND(0.067)	0.053	0.52	Kaken
T-S7	May 31	Flounder	ND(0.33)	0.36	0.68	TEPCO

Chart 2 KEEA data

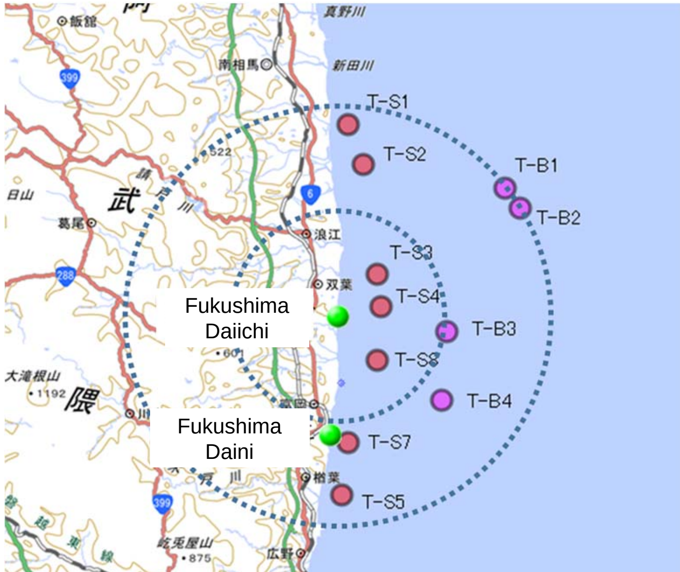
Sampling location	Sampling date	Fish type	Concentrations in surrounding sea water (Bq/L)	Free water tritium (Bq/L)	Organically bound tritium (Bq/L)	Analysis location
T-S8	May 10	Flounder	0.070	0.057	ND(0.27)	KEEA
T-S8	June 28	Flounder	0.075	0.075	ND(0.27)	KEEA

Chart 3 Kaken data (After recommencing analysis)

Sampling location	Sampling date	Fish type	Concentrations in surrounding sea water (Bq/L)	Free water tritium (Bq/L)	Organically bound tritium (Bq/L)	Analysis location
T-S1	June 3	Flounder	ND(0.068)	0.15	Analysis underway	Kaken
T-S2	June 3	Flounder	ND(0.074)	0.12	ND(0.39)	Kaken
T-S3	June 23	Flounder	0.14	0.12	Analysis underway	Kaken
T-S4	June 23	Flounder	0.14	0.13	ND(0.27)	Kaken
T-S5	June 30	Flounder	0.11	0.15	ND(0.28)	Kaken

※ KEEA analysis results were the same as always showing that the concentration of tritium in fish was the same, or lower, as the surrounding seawater

※1 No measurements could be performed due to a lack of specimens



Map created by Tokyo Electric Power Company Holdings, Inc. based on the Geographical Survey Institute website

Fish sampling point map