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A new approach to determine abnormality of radioactive discharges from pressurized water reactors and to derive abnormality indicators correlated with a specific causal event

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Abstract: A novel methodology to analyze the trend of radioactive effluent data from nuclear power plants (NPPs) has been proposed to estimate the possibility of abnormal releases of radioactive materials, and to identify sensitive radionuclides with high correlation to specific events and abnormal releases. Actual discharge data including abnormal releases for 21 years (1,610 reactor-years) from 62 US and 22 Korean pressurized water reactors were collected. A release correlation factor (RCF) was newly proposed as a quantitative indicator of the abnormality of radioactive discharges based on annual discharge data from each NPP. The probability of correct estimation of abnormality using the RCF for a test case ranges 27–36 %, and such a modest estimation power was ascribed to the relatively low contribution (i.e., about 12 %) of a specific causal event to the annual radioactivity release. Through this, the feasibility of the RCF for estimating the abnormality of radioactive discharges was demonstrated. The RCF was derived for three types of causal events, and subsequently the sensitive radionuclides to each specific event were derived as ^{88}Kr , ^{137}Xe , and ^{138}Xe for leakage from gas decay tank, ^3H for leakage from steam generator's power operated relief valve, and ^{85}Kr , ^{88}Kr , ^{133}Xe , $^{133\text{m}}\text{Xe}$, ^3H and ^{14}C for leakage from defected spent fuel during handling. The new approach proposed in this study is to be used to suppose the significant unreported abnormality in radioactive effluent data and the potential causal event, which may contribute to improving the safety and performance of radioactive effluent control as necessary.

1 Introduction

In the International Atomic Energy Agency (IAEA) General Safety Guide, the authorized discharge limit is described as typically limiting either the dose to the representative person, the activities of radionuclides discharged or the activity concentration of liquid and gaseous effluents (IAEA 2018). To ensure compliance with the authorized discharge limits for liquid and gaseous effluents, operators or regulatory bodies conduct monitoring (IAEA 2005). The operating organizations of nuclear facility must record and maintain records of monitoring and compliance with regulatory standards. Furthermore, the operating organizations should promptly report any discharges exceeding the authorized limits on discharges in accordance with criteria established by the regulatory body. Additionally, any significant increase in dose rate or concentrations of radionuclides in the environment that could be attributed to the facility or activity must also be reported (IAEA 2018).

In the United States Nuclear Regulatory Commission (USNRC)'s Code of Federal Regulations, it is stipulated that any event or situation related to the health and safety of the public, onsite personnel, or protection of the environment, for which a news release is planned or notification to other government agencies has been or will be made, including inadvertent release of radioactively contaminated materials, must be reported to the NRC (USNRC 2023a). According to the Regulatory Guide of the USNRC, licensees are required to submit an Annual Radioactive Effluent Release Report (ARERR) to the NRC annually (USNRC 2021). This report should document and report the concentrations or the activities of radioactive materials discharged to the environment during normal operations, as well as information on abnormal releases and discharges. The ARERR includes reporting of events that led to unplanned/abnormal releases and discharges annually for each nuclear power plant (NPP), and for some NPPs, it also includes reporting of

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the radionuclides (groups) that were released during unplanned/abnormal releases.

In Korean regulations, it is specified that in the event of unplanned or uncontrolled releases of radioactive materials from nuclear facilities to the environment, reporting to the Nuclear Safety and Security Commission (NSSC) is mandatory (NSSC 2023). The regulatory authority in Korea discloses information on accidents and malfunctions at NPPs through Operational Performance Information System for Nuclear Power Plant (OPIS), but the frequency of such reporting appears relatively lower compared to the United States (KHNP 2001; KINS 2024). In Japan, the Act requires operators to record and report abnormal releases to regulatory authorities; however, actual reports submitted by operators to regulatory agencies do not contain specific information on abnormal releases (NRA 1957, 2023).

When recording compliance with monitoring and regulatory standards, the operators should include main operational and discharge data for the period, along with conclusions on trends observed through comparison with previous results (IAEA 2018). This recommendation for discharge data and trend analysis can be interpreted to apply not only to normal discharges but also to unplanned/abnormal releases. In particular, if the association between the occurrence of unplanned/abnormal releases and events can be identified through unplanned/abnormal release data and trend analysis, it would be possible to estimate events leading to unplanned/abnormal releases by analyzing discharge characteristics from radioactive effluents data.

Through a comprehensive review of the literature, it has been confirmed that while there have been some studies on trend analysis of normal discharge of radioactive effluents, most preliminary studies on unplanned/abnormal releases simply report relevant information without conducting quantitative research on the correlation between unplanned/abnormal releases and the events leading to it (FENOC 2020; Harris and Miller 2008; Kong et al. 2017). The UK Environment Agency (EA) estimated that the peak points of discharge trends for 27 NPPs in 8 countries, including European Pressurized Reactor (EPR), Advanced Passive 1000 (AP1000), Economic Simplified Boiling Water Reactor (ESBWR), and Advanced Candu Reactor 1000 (ACR-1000), were associated with specific events (Copplestone et al. 2009; EA 2009). However, this study analyzed only a total of 13 cases, including 4 cases for pressurized water reactors (PWRs), indicating that the scope of the study was very limited and the analysis was predominantly qualitative. The USNRC reported 14 events of unplanned and unmonitored abnormal environmental releases of liquid tritium from US NPPs, including 10 PWRs and 4 boiling water reactors (BWRs), between 1996 and 2006 (USNRC 2006). While this

study mentioned a potential correlation between increased release the activities during abnormal releases and specific events, it also noted limitations in its scope.

Therefore, this study proposed a new approach that both estimates the possibility of abnormal releases through trend analysis of radioactive effluent data and identifies the correlation between the increase in the activities of radionuclides (groups) discharged and causal events. The new approach's validity was confirmed by collecting extensive discharge data, including detailed reports on abnormal releases and causal events, from the US ARERR, constructing an integrated radioactive effluent database. Subsequently, the derived methodology's applicability was verified using US and Korean PWRs.

2 Materials and Methods

2.1 Construction of an integrated radioactive effluent database for estimating abnormal release

Radioactive effluent data is partially available worldwide, with most reports publicly disclosing the activities of radioactive materials in normal, while data on abnormal releases is very limited (EC 2010, KHNP 2009, 2020, NRA 2023). Therefore, publicly available radioactive effluent information including abnormal releases was collected and processed from the US and Korean radioactive effluent data separately to construct and utilize an integrated database. To understand the correlation between discharge data and abnormal releases, a database was constructed based on comprehensive recording and reporting of normal and abnormal releases in the ARERR.

For 62 operating PWRs in the US, annual discharge data of radioactive effluents released to the environment for the period from 2000 to 2020 were collected and database-ized by liquid and gaseous radionuclides (groups) (FENOC 2020). A total of 1,300 reactor-years (RYs) out of 1,302 RYs of US PWRs were included in the integrated database, excluding 2 RYs (Beaver Valley Unit 1 in 2006 and Saint Lucie Unit 2 in 2018) due to typographical errors in the activities of radionuclides. The integrated database was constructed by reconstructing information such as NPP, year, liquid and gaseous radioactive effluents, radionuclides or radionuclide groups, the activities of annual discharge, status of abnormal releases for the respective year, radioactive effluents from specific events or specific event-related abnormal releases (if any available for some NPPs), and specific event information. The selection of radionuclides (groups) followed the criteria classified in

Regulatory Guide 1.21 of the USNRC (USNRC 2021). Gaseous radionuclides (groups) are represented in this paper as *G*, *I*, *P*, *T*, and *C*, corresponding to fission products, radioiodine, particulates, gaseous tritium, and gaseous ^{14}C , respectively. Liquid radionuclides (groups) are represented as *F*, *D*, and *H*, corresponding to fission and activation products, dissolved gases, and liquid tritium, respectively.

For Korean NPPs, radioactive effluent data by liquid and gaseous radionuclides (groups) from 22 operating PWRs were collected from the “Survey of Radiation Environment and Assessment of Radiological Impact on Environment in Vicinity of Nuclear Power Facilities” and “Radiation Management Yearbook” (KHNP 2009, 2020). A total of 310 RYs are included in the integrated database. The selected Korean NPPs include Kori Units 1 to 4, Hanbit Units 1 to 6, Hanul Units 1 to 6, Shin Kori Units 1 to 4, and Shin Wolsong Units 1 to 2, and data from 2000 to 2020 were utilized. Kori Unit 1 was permanently shut down in 2017, so data from 2000 to 2016 were used for this unit, and for newly constructed NPPs after the 2000s, data from the year following their commissioning were utilized (IAEA 2023; Kim, N.H. et al. 2021). Reports of accidents and failures for each NPP can be found in the OPIS, but there are limitations in estimating abnormal releases, so data on the safety regulatory status of each plant were collected (KINS 2024; NSSC 2024).

Based on the preliminary analysis of the integrated database, abnormal releases in US PWRs consist of 210 RYs for gaseous, 73 RYs for liquid, and 36 RYs for simultaneous

gaseous and liquid releases. When counting the number of individual events per NPP in a year where multiple abnormal releases occurred, there were a total of 705 events, with 519 events for gaseous, 93 events for liquid, and 93 events for simultaneous gaseous and liquid releases. Out of 1,300 RYs, abnormal releases occurred in 319 RYs, accounting for a ratio of 24.54 %, implying that there is approximately a 1/4 chance of abnormal releases occurring in a specific year. In contrast, among the 310 RYs of Korean PWRs, a total of 31 RYs are suspected of abnormal release, with a ratio of 10 %, indicating a relatively lower occurrence compared to the US, at about 2.3 %.

Figure 1 illustrates the annual status of normal and abnormal releases for the relatively high number of reported abnormal release events in US PWRs with 1,300 RYs from the integrated database. Unlike the years of normal discharge (white cells), Figure 1 indicates the years of abnormal releases in color, with six colors (red, orange, green, yellow, blue, purple) distinguishing the physical state of discharged effluents (liquid, gaseous, simultaneous liquid and gaseous) and discharged NPPs (single unit, multiple units). The horizontal axis denotes abbreviations for the 62 US PWRs, while the vertical axis represents the years from 2000 to 2020. The numbers in the cells represent the occurrences of abnormal releases in the respective years. When abnormal releases result in the simultaneous discharge of liquid and gaseous, they are distinguished as “L” (Liquid) and “G” (Gaseous) in the figure.

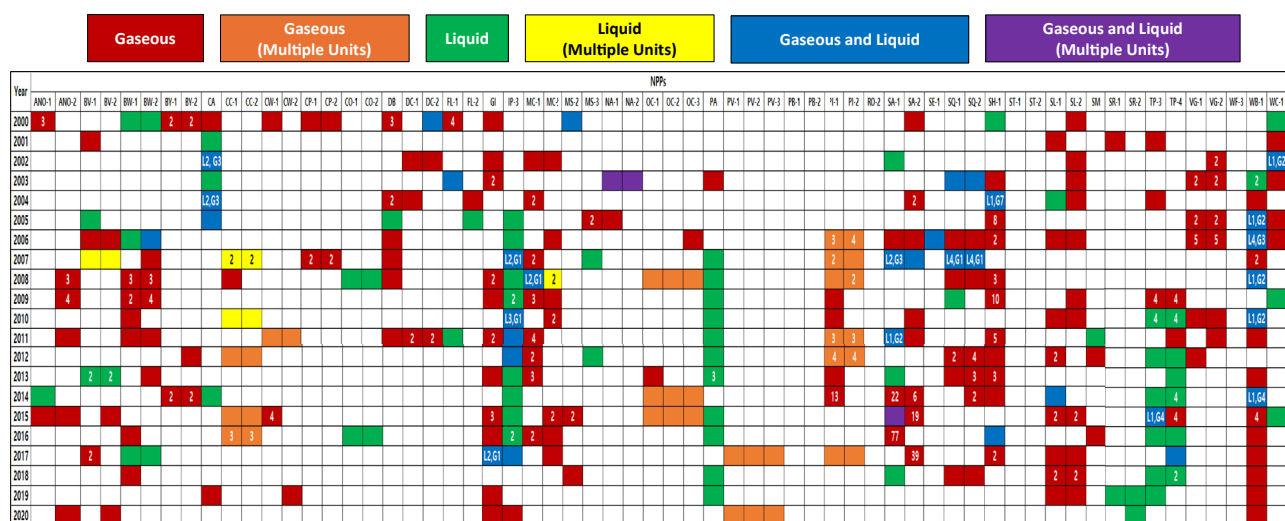


Figure 1: The status of normal and abnormal releases of radioactive materials, both liquid and gaseous, from US PWRs (1,300 RYs) during the period of 1990–2020, as compiled in the integrated radioactive effluent database established in this study. The numbers within each cell represent the occurrence of abnormal releases in the corresponding year. Colored cells without numbers indicate a single abnormal release in that year, while cells with numbers indicate multiple abnormal releases in the same year. Red, green, and blue denote gaseous, liquid, and simultaneous liquid and gaseous release, respectively, from single unit, while orange, yellow, and purple denote gaseous, liquid, and simultaneous liquid and gaseous release, respectively, from multiple units.

2.2 Proposal of the release correlation factor as an indicator for estimating abnormal releases

In research within the non-nuclear fields, based on the historical discharge data of chemical pollutants by type, attempts were undertaken to identify factors leading to increases in the abnormal discharge of pollutants into the environment and their causes, and to compare the activities of discharge by factor (Luan et al. 2022). However, there are limitations in that the correlation between the increase of discharge and abnormal release was analyzed qualitatively (such as issues with facilities or policy changes). In the nuclear fields, cases of estimating abnormal release using the historical discharge data of radioactive materials are not publicly available in general literature. Therefore, in this study, two candidate equations for the release correlation factor (RCF) were proposed as indicators of the correlations between the discharge data and abnormal release, and between the increase in activities of discharge and abnormal release by radionuclides (groups) based on integrated radioactive effluent database.

The first candidate equation for RCF was defined as the reciprocal of the ratio of the activities of radionuclide (group) discharged in the specific year to the activities of radionuclide (group) discharged in the previous year ($s_{n,i}^{-1}$), as shown in Equation (1). In Equation (1), $s_{n,i}$ represents the activities for year n by radionuclide (group) i relative to the annual discharge for the previous year ($n - 1$). In this study, the reciprocal of $s_{n,i}$, denoted as $s_{n,i}^{-1}$, was proposed as the first candidate RCF. Cases where there was no discharge for a specific radionuclide (group) (i.e., $A_{n,i} = 0$) were not considered as they were not related to abnormal release.

$$s_{n,i}^{-1} = \frac{A_{n-1,i}}{A_{n,i}} \quad (A_{n,i} \neq 0) \quad (1)$$

where, $A_{n,i}$ represents the annual activities (TBq/y) of radionuclide (group) i at a specific NPP in year n , where n refers to a specific year within the period from 2001 to 2020.

Equation (1) employs the ratio of the activities of radionuclide (group) discharged between a specific point (i.e., a particular year) and the previous point (i.e., the preceding year). However, this equation has inherent limitations in detecting increases in activities of radionuclides discharged due to abnormal release in cases where there is a continuous increase in the activities of radionuclides (groups) discharged over a certain period (i.e., several years). To address such limitations as observed in Equation (1), the

second candidate equation for RCF was derived, as shown in Equation (2).

$$f_{n,i} = \frac{A_{n,i}}{\overline{A_{n-1,i}}} \quad (2)$$

where, $\overline{A_{n-1,i}}$ represents the average activities of radionuclide (group) i over the period up to year $(n - 1)$, starting from the year 2000 (TBq/y).

In Equations (1) and (2) presented above, if the value of $s_{n,i}^{-1}$ is less than 1, and the value of $f_{n,i}$ is greater than 1, it can be considered that activities of radionuclide (group) discharged for that year has increased compared to the annual average activities of the previous year or the preceding years. In this case, the likelihood of abnormal release is higher, implying a greater probability that the estimation of abnormal releases is true.

To assess the correlation between the data and abnormal release, the estimation ratio of abnormal release according to each radionuclide (group)'s RCF value can be calculated to determine the degree of correlation. The estimation ratio of abnormal release for radionuclide (group) i for a period of k years based on the RCF value is defined as $ER_{RCF,i,k}$, as shown in Equation (3).

$$ER_{RCF,i,k} = \frac{RY_{(s^{-1} < RCF), ab, i, k}}{RY_{(s^{-1} < RCF), tot, i, k}} \text{ or } \frac{RY_{(f > RCF), ab, i, k}}{RY_{(f > RCF), tot, i, k}} \quad (3)$$

$RY_{(s^{-1} < RCF), ab, i, k}$ and $RY_{(f > RCF), ab, i, k}$ represent the number of reactor years (RYs) corresponding to abnormal release for radionuclide (group) i for a period of k years when s^{-1} is less than RCF and when f is greater than RCF, respectively. $RY_{(s^{-1} < RCF), tot, i, k}$ and $RY_{(f > RCF), tot, i, k}$ denote the total number of RYs for radionuclide (group) i for a period of k years when s^{-1} is less than RCF and when f is greater than RCF, respectively.

The frequency of abnormal release caused by any events j , regardless of RCF, for a period of k years, $Freq_{ab,j,k}$ is defined as shown in Equation (4).

$$Freq_{ab,j,k} = \frac{RY_{ab,j,k}}{RY_{tot,k}} \quad (4)$$

where, $RY_{ab,j,k}$ represents the number of RYs for a period of k years affected by the abnormal release due to the event causing j , and $RY_{tot,k}$ denotes the total RYs for a period of k years. When $ER_{RCF,i,k}$ is higher than $Freq_{ab,j,k}$, stronger correlation with abnormal release is expected. For events where types are not distinguished, excluding j , it is denoted as $Freq_{ab,k}$, while for distinct event types j , they will be addressed in Section 3.2.

2.3 Correlation strength between increases the activities of specific radionuclides (groups) discharged and causal events

Causal events, j , that could lead to abnormal releases as described in Equations (3) and (4), can be broadly classified into primary system leakages, secondary system leakages, leakages of liquid and gaseous radioactive waste management system, leakage from defected spent fuel (SF) during handling, and spent fuel pool (SFP) leakages (ASME 2012, USNRC 2023b).

In the coolant of PWRs, tritium is generated through the neutron irradiation of boron and lithium, while ^{14}C , irradiation products, and Chalk River unidentified deposits (CRUD) are generated through the neutron irradiation of oxygen and nitrogen within the coolant (ORNL 1977, 1980). These generated radionuclides can be released externally in the event of primary system leakages (USNRC 2020).

In the secondary system of PWRs, even during normal operation, the tritium generated in the primary system may exist at slightly elevated levels compared to natural levels, and in the secondary system leakages, tritium can be released in liquid or gaseous. Within the secondary system, the power operated relief valve (PORV) of the steam generator (SG) plays a role in supplying generated steam to the turbine, and in the event of valve leakage, gaseous tritium corresponding to steam (H_2O) can be released (USNRC 2016).

In the gaseous radioactive waste management system (GRWMS) of the radioactive waste management system (RWMS), if a leak occurs, the primary component within this system, noble gas, which is a fission product, may be released (Caron and Yim 2006). Among these components, the gas decay tank (GDT) is designed to store compressed waste gases (i.e., fission gases), noble gas may be released due to piping and valve leaks (American Society of Mechanical Engineers Design 2012, IAEA 2016, USNRC 2012).

Within the nuclear fuel pellets of UO_2 in the reactor core, in addition to gaseous and fission products, tritium and ^{14}C can also be generated (Kim and Lee 2021). Tritium is produced through ternary fission, while ^{14}C can be generated through $^{17}_8\text{O}(\text{n}, \alpha)^{14}_6\text{C}$ neutron irradiation (ASME 2012; ORNL 1977, 1980; USNRC 2023b; Caron and Yim 2006). In the leakage from defected SF during handling, these fission products, tritium, and ^{14}C can be released (Jeong and Cho 2020).

Additionally, tritium is released from the fuel building through the evaporation of tritiated water from the SFP, and fission products are released from damaged fuel cladding in SF that has been cooled over an extended period. Therefore, when handling SF for storage, transportation, etc.,

consideration must be given to the effects of fission products (IAEA 2020; KHNP 2007).

As a result of significant events, specific radionuclides may be abnormally released, leading to an increase in the activities of radionuclides (groups) discharged. Therefore, there is a need to identify sensitive radionuclides (groups) that can assess the correlation between the activities of radionuclides (groups) and the causal events. Sensitive radionuclides (groups) expected for specific events can be classified as follows: noble gases, tritium, and ^{14}C (leakage from defected SF during handling), gaseous tritium (leakage from SG's PORV), and noble gases (leakage from GDT).

To assess the correlation between the increase in the activities of specific radionuclides (groups) discharged and the causal events, preliminary regression analysis was conducted. Among linear regression analyses, the basic equation of the linear equation was transformed into Equation (5) and utilized in the analysis of the results (Jabbari and Rezaei 2022).

$$\text{ER}_{\text{RCF}, i, j, k} = a \cdot \text{RCF}_{i, j, k} + b \quad (5)$$

where, $\text{ER}_{\text{RCF}, i, j, k}$ represents the dependent variable and refers to the estimation ratio of abnormal release caused by event j for a period of k years, for radionuclide (group) i , while $\text{RCF}_{i, j, k}$ represents as the independent variable denoting the RCF value when main causal event j occurs for radionuclide (group) i for a period of k years, with a representing the slope and b the constant term.

When the value of s^{-1} is less than 1, if the value of $\text{RCF}_{i, j, k}$ increases, the value of $\text{ER}_{\text{RCF}, i, j, k}$ decreases, resulting in a negative sign; conversely, when f is greater than 1, if the value of $\text{RCF}_{i, j, k}$ increases, the value of $\text{ER}_{\text{RCF}, i, j, k}$ increases, resulting in a positive sign. For events where the type is not specified, $\text{ER}_{\text{RCF}, i, k}$ is represented without j and is calculated similarly to Equation (3). Similarly, RCF_{i, j, k_0} events without a specified typed are represented as $\text{RCF}_{i, k}$, excluding j .

To identify sensitive radionuclides with a high correlation between the increase the activities of radionuclide (group) i discharged and major causal event j , both for events where the type is not specified and for events where the type is specified, three criteria for judgment were proposed. These criteria are based on Equation (1), where if the value of s^{-1} is less than 1, a decrease in the value of s^{-1} indicates an increase in the estimation ratio of abnormal release (when the sign of a in Equation (5) is negative), and based on Equation (2), where if the value of f is greater than 1, an increase in the value of f indicates an increase in the estimation ratio of abnormal release (when the sign of a in Equation (5) is positive), representing an increase the activities of radionuclides (groups) discharged due to causal

Table 1: The judgement criteria for deriving sensitive radionuclides (groups) with high correlation between the increase of the activities of radionuclides (groups) discharged and causal events.

No.	Judgement criteria
1	The sign of the slope a in Equation (5) is negative when the value of s^{-1} is less than 1, and positive when the value of f is greater than 1.
2	When the coefficient of determination (R^2) is 0.46 or higher ^a
3	When the $ER_{RCF,i,k}$ in Equation (3) exceeding the value of $Freq_{ab,k}$ in Equation (4) by 90 % or more

^aThe coefficient of determination R^2 is the square of the correlation coefficient R . The correlation coefficient R indicates a strong correlation between variables when it ranges from 0.68 to 1. R^2 ranges from 0 to 1, with values closer to 1 indicating a higher correlation between the variables (Nishiuchi 2023; Taylor 1990).

events. By establishing a meaningful range for RCF values in this way, three judgment criteria for deriving sensitive radionuclides (groups) were summarized in Table 1.

'Judgement Criterion 1' (C-①) is mandatory, and if both 'Judgement Criterion 2' (C-②) and 'Judgement Criterion 3' (C-③) are met simultaneously, it indicates the highest estimation accuracy for the strong correlation between the increase of the activities of radionuclides (groups) discharged and causal events. C-① must be satisfied, and at least one of C-② or C-③ must be met to derive sensitive radionuclides (groups).

3 Results and discussion

3.1 Derivation of RCF for discharge data from US PWRs and its applicability

3.1.1 Calculation and distribution characteristics of RCF for US PWRs

In Section 2.2, RCF values were derived for US PWRs with 1,300 RYs mentioned. Using Equations (1) and (2), the values of s^{-1} and f were calculated for radionuclides (groups) for every RY, and the results were plotted in ascending order in Figure 2. In Figure 2, the y-axis represents RCF values, where (a) indicates the range where the value of s^{-1} is less than 1, and (b) indicates the range where the value of f is greater than 1, which is the significant range.

In Figure 2, the reason for the relatively low number of data points for ^{14}C compared to other radionuclides (groups) is that Regulatory Guide 1.21 was revised in 2009, adding the record item of ^{14}C to the ARERR in US, leading to the recording of ^{14}C discharge data from US NPPs starting in 2010 (USNRC 2009).

Fundamentally, Figure 2 represents a curve of the 'upward and downward parabola' type, where the narrower the gap between the exponential growth and logarithmic growth parts and the steeper the slope, the greater the variability of increases or decreases in annual activities during the respective periods. From Figure 2A, it can be inferred that radionuclides (groups) with relatively large variations in annual activities are radioiodine (I), particulates (P), and dissolved gases (D) in the range where s^{-1} values are less than 1, and from Figure 2B, it can be inferred that radionuclides (groups) with f values greater than 1 are I , P , D , and fission and activation products (F).

To assess the relative distribution of annual activities by radionuclides (groups), the coefficient of variation (CV), which is the standard deviation divided by the mean, was calculated (Nair and Rao 2003). Table 2 shows the statistical values (mean and CV) of annual activities by radionuclides (groups) and the ratio of RCF values distributed within the range of 0.1–10 in Figure 2.

In this study, the ratio of RCF values distributed between 0.1 and 10 was used to differentiate between relatively large or small differences in annual activities during normal and abnormal releases due to events. As indicated in Table 2, ^{14}C (C) and liquid and gaseous tritium (H and T) have the highest proportion, with both s^{-1} and f values falling within the range of 0.1–10, exceeding 95 %. Therefore, radionuclides (groups) with a high proportion of RCF values distributed between 0.1 and 10 are considered that their abnormal releases contribute less to the annual activities. The analysis results indicating that I , P , and D show relatively large variations in annual activities in Figure 2 align with Table 2, where the proportion of RCF values for these radionuclides (groups) falling between 0.1 and 10 is relatively low.

The CV values for the annual activities of C , T , and H are 0.74, 1.18, and 2.08, respectively, indicating very small variations in annual activities. This aligns with the fact that both s^{-1} and f values fall within the range of 0.1–10, which is the highest proportion among the three radionuclides (groups). These results suggest that the relative magnitude of variations in annual activities between normal and abnormal releases can be reasonably represented by the distribution of s^{-1} and f values.

3.1.2 Estimation ratio of abnormal releases according to RCF

As stated in Equation (5), s^{-1} less than 1 or f greater than 1 implies that the discharges for the current year compared to the previous year or average from the reference year (i.e., 2000) up to the previous year has increased. In this

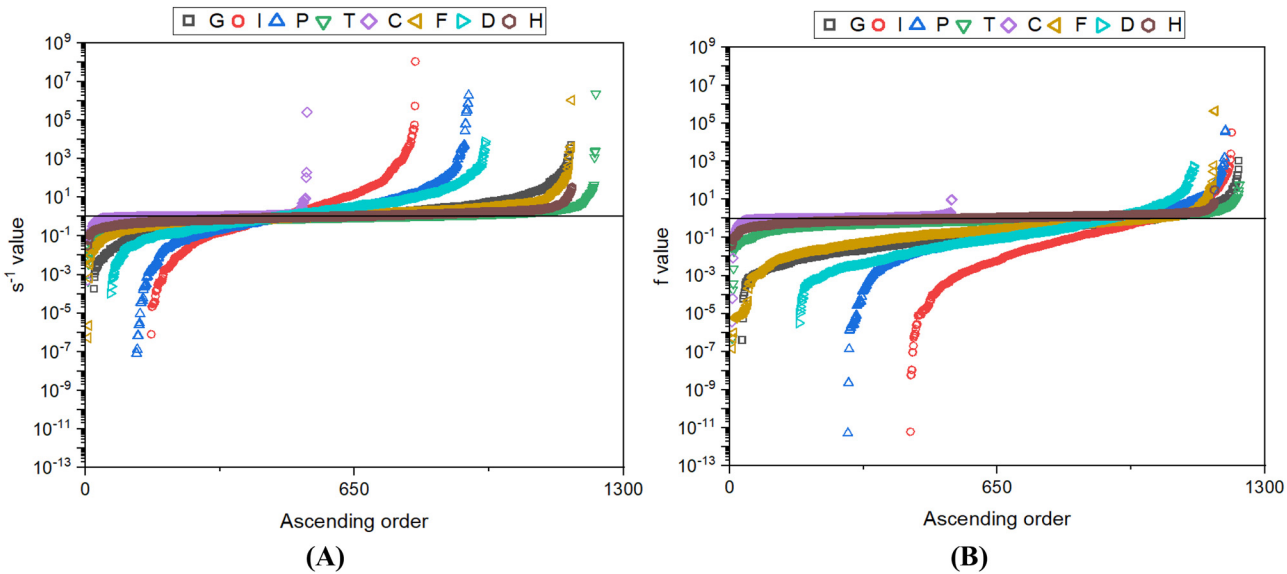


Figure 2: Distribution curves of RCF by radionuclides (groups) for 1,300 RYs in US PWRs. (A) and (B) represent the values of s^{-1} and f , respectively. The legend of the graph: G , I , P , T , and C represent gaseous radionuclides (groups) denoting fission products, radioiodine, particulates, gaseous tritium, and gaseous ^{14}C , respectively. F , D , and H represent liquid radionuclides (groups) denoting fission and activation products, dissolved gases, and liquid tritium, respectively.

Table 2: The distribution ratio of RCF values and statistical values of annual activities by radionuclides (groups) for 1,300 RYs of US PWRs.

Radionuclides (groups) ^a		Distribution ratio within the range of 0.1–10		Statistical values of annual activities	
		RCF		Mean	CV
		s^{-1}	f		
Gaseous	G	78 %	51 %	1.39	4.37
	I	42 %	26 %	9.44×10^{-4}	19.2
	P	54 %	54 %	4.08×10^{-4}	25.2
	T	96 %	95 %	3.01	2.08
	C	98 %	99 %	0.33	0.74
Liquid	F	91 %	63 %	3.44	25.1
	D	65 %	36 %	4.72×10^{-3}	6.68
	H	98 %	99 %	25.9	1.18

^a G , I , P , T , and C represent gaseous radionuclides (groups) denoting fission products, radioiodine, particulates, gaseous tritium, and gaseous ^{14}C , respectively. F , D , and H represent liquid radionuclides (groups) denoting fission and activation products, dissolved gases, and liquid tritium, respectively.

study, it is assumed that as the values of s^{-1} decrease or f increase, the estimation ratio of abnormal releases, as presented in Equation (3), increases. In this case, it is expected that there would be a high correlation with abnormal releases.

Figure 3 depicts the estimation ratio of abnormal releases, $\text{ER}_{\text{RCF},i,00-20}$, by radionuclides (groups) according to

Equation (3) for the dataset of discharges from 1,300 RYs in the US from 2000 to 2020. Figures 3A and B represent the estimation ratio of abnormal releases where the values of s^{-1} is less than 1 and f is greater than 1 according to Equation (3), respectively. The horizontal dashed line in Figure 3 represents the $\text{Freq}_{ab,00-20}$ mentioned in Equation (4) for the entire dataset of discharges from 1,300 RYs in the US from 2000 to 2020.

In Figure 3B, there are a total of 29 RYs across all radionuclides (groups) except for the I and D where the y-axis $\text{ER}_{\text{RCF},i,00-20}$ is 0 %. This is expected when the average activities up to the year before abnormal releases are significantly low compared to the years when abnormal releases occur. As an example, at the Beaver Valley NPP, during maintenance activity from 2000 to 2002, discharges of H were observed to be approximately 0.1 times lower than in other years (FENOC 2002). To ensure meaningful result analysis in Sections 3.1.2 and 3.1.3, f values where $\text{ER}_{\text{RCF},i,00-20}$ is 0 % for each radionuclide (group) were excluded.

In Figure 3, for fission products (G), the estimation ratio $\text{ER}_{\text{RCF},G,00-20}$ is consistently lower than the $\text{Freq}_{ab,00-20}$ for all RCF values. Similarly, in Figure 3A, for I , the estimation ratio $\text{ER}_{\text{RCF},I,00-20}$ is consistently lower than the $\text{Freq}_{ab,00-20}$ for all RCF values as well. In Figure 3A, for T , C , H , and F , as the value of s^{-1} decreases, the estimation ratios $\text{ER}_{s^{-1},T,00-20}$, $\text{ER}_{s^{-1},C,00-20}$, $\text{ER}_{s^{-1},H,00-20}$, and $\text{ER}_{s^{-1},F,00-20}$ respectively increase, with most s^{-1} values having higher values than the $\text{Freq}_{ab,00-20}$. In Figure 3B, for P and D , as f increases, the

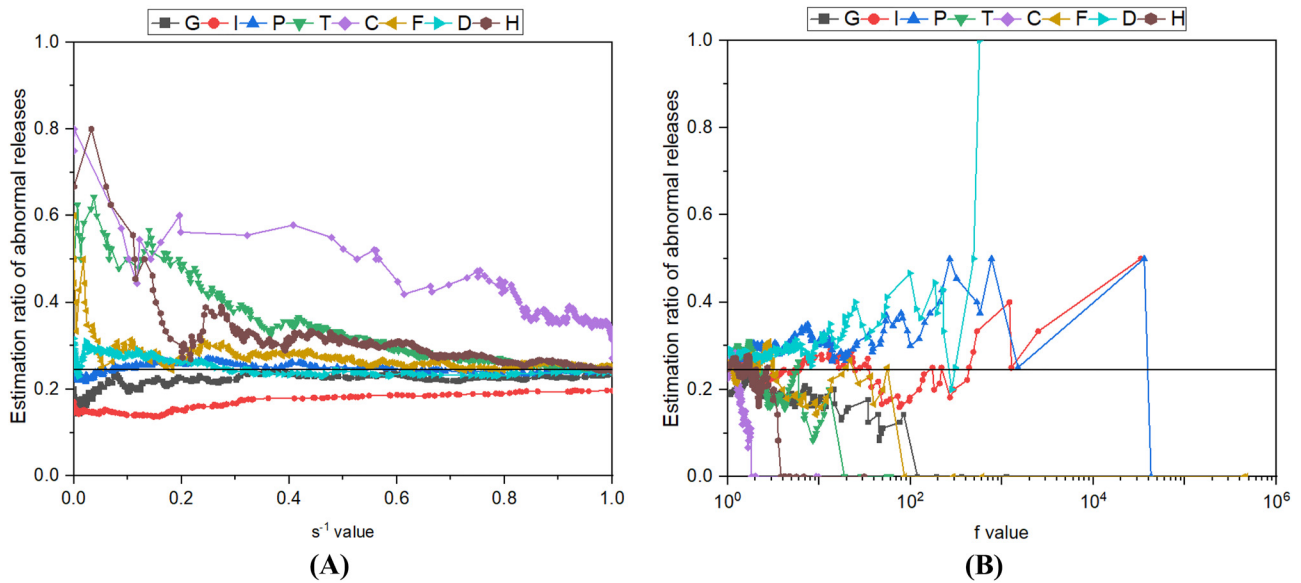


Figure 3: Estimation ratio of abnormal releases by radionuclides (groups), $ER_{RCF,00-20}$, based on RCF values for 1,300 RYs in US PWRs. (A) and (B) represent the estimation ratio of abnormal releases according to the values of s^{-1} and f , respectively.

estimation ratios $ER_{f,P,00-20}$ and $ER_{f,D,00-20}$ respectively increase, with most f values having higher values than the $Freq_{ab,00-20}$. Based on the above results, G and I were excluded from the analysis, and in the s^{-1} value, T , C , and H were analyzed, while in the f value, P and D were analyzed.

To assess the correlation between the increases of the activities of radionuclides (groups) discharged due to abnormal releases by radionuclides (groups) and the causal events, linear regression analysis based on Equation (5) was conducted on the dataset of discharges from 1,300 RYs in the US as depicted in Figure 3. Subsequently, the satisfaction status and values of the criteria for identifying sensitive radionuclides (groups) with high correlation between the

increases of the activities of radionuclides (groups) discharged and causal events, as criteria presented in Table 1, were summarized in Table 3.

In Table 3, T , C , and H satisfy all three criteria, indicating the highest correlation between the increases of the activities of radionuclides (groups) discharged and causal events in terms of s^{-1} values, while D satisfies all three criteria in terms of f values. Although not all criteria are satisfied, radionuclides (groups) that meet C-① and either C-② or C-③ are F and D in terms of s^{-1} values and P in terms of f values. Radionuclides (groups) that meet only C-① are I in terms of f values.

As mentioned in Section 2.3, T and C are confirmed to be generated whenever major events (primary system leakage,

Table 3: Assessment of compliance with criteria for determining sensitive radionuclides (groups) with high correlation between the increases of the activities of radionuclides (groups) discharged and causal events for 1,300 RYs in US PWRs. If the criteria are met, mark as Yes (Y); if not satisfied, mark as No (N).

Radionuclides (groups)		Verification of satisfaction status of the criteria (values)					
		s^{-1}			f		
		C-①	C-②	C-③	C-①	C-②	C-③
Gaseous	G	N (0.043)	N (0.38)	N (0 %)	N (-1.58×10^{-3})	Y (0.47)	N (0 %)
	I	N (0.058)	Y (0.82)	N (0 %)	Y (8.05×10^{-6})	N (0.33)	N (55.2 %)
	P	N (0.006)	N (0.02)	N (49.7 %)	Y (5.75×10^{-6})	N (0.15)	Y (99.5 %)
	T	Y (−0.299)	Y (0.86)	Y (99.4 %)	N (−0.02)	Y (0.56)	N (80.5 %)
	C	Y (−0.293)	Y (0.82)	Y (100 %)	N (−0.2)	Y (0.64)	N (52.5 %)
Liquid	F	Y (−0.079)	N (0.37)	Y (98 %)	N (-1.17×10^{-3})	N (0.09)	N (48.1 %)
	D	Y (−0.06)	Y (0.55)	N (56.5 %)	Y (6.58×10^{-4})	Y (0.51)	Y (99.3 %)
	H	Y (−0.173)	Y (0.55)	Y (96.3 %)	N (−0.039)	Y (0.51)	N (59.3 %)

secondary system leakage, leakage of RWMS, leakage from defected SF during handling). They can be classified as radionuclides (groups) with high estimation ratios of abnormal releases according to the results of Figure 3A. Moreover, based on the results of Table 3, it can be anticipated that T and C may experience an increase in activities due to abnormal releases when events occur, indicating a high correlation with events.

In Section 3.1.1, it was confirmed that the distribution of RCF in T , C , and H is predominantly between 0.1 and 10, with a proportion exceeding 95 %, and the CV values are low, ranging from 0 to 3. Based on these results, it was expected that the estimation ratio of abnormal releases would be low due to the small magnitude of the increases of activities of radionuclides (groups) discharged during abnormal releases. However, the results from Figure 3 and Table 3 indicate a very high correlation between the increases of activities of radionuclides (groups) discharged and abnormal releases, as well as a high correlation with causal events, for the same radionuclides (groups).

3.1.3 Derivation and verification of reference values of RCF

The RCF derived from a consistent set of discharge data can be utilized to estimate the correlation with abnormal releases for succeeding discharge datasets. In this study, to verify the applicability of the proposed RCF, discharge data from US PWRs from 2000 to 2020 were divided into two groups: Group 1 discharge data (i.e., preceding discharge

data) from 2000 to 2010 and Group 2 discharge data (i.e., succeeding discharge data) from 2011 to 2020.

Group 1 discharge data consists of a total of 681 RYs, with 163 RYs occurring abnormal releases. Group 2 discharge data comprises 619 RYs, with 156 RYs occurring abnormal releases. Therefore, the $\text{Freq}_{ab,11-20}$ calculated using Equation (4) is 25.2 % (=156 RYs/619 RYs). Figure 4 represents the estimation ratio of abnormal releases $\text{ER}_{\text{RCF},i,00-10}$ by radionuclides (groups) based on the RCF values from the Group 1 discharge dataset of 681 RYs from 2000 to 2010 in the US, following the same methodology as Figure 3. The horizontal dashed line in Figure 4 represents the $\text{Freq}_{ab,11-20}$ calculated for the entire Group 2 discharge dataset of 619 RYs from 2011 to 2020.

In Figure 4A, it is observed that for G , I , and P , the estimation ratios $\text{ER}_{s^{-1},G,00-10}$, $\text{ER}_{s^{-1},I,00-10}$, and $\text{ER}_{s^{-1},P,00-10}$ are lower than $\text{Freq}_{ab,11-20}$ for all s^{-1} values. Similarly, in Figure 4B, for G and T , the estimation ratios $\text{ER}_{f,G,00-10}$ and $\text{ER}_{f,T,00-10}$ are lower than $\text{Freq}_{ab,11-20}$ for all f values.

In Figure 4A, it is observed that for the T and H , as the value of s^{-1} decreases below 1, the estimation ratios $\text{ER}_{s^{-1},T,00-10}$ and $\text{ER}_{s^{-1},H,00-10}$ increase. For most s^{-1} values, these ratios are higher than $\text{Freq}_{ab,11-20}$. Similarly, in Figure 4B, it is observed that for the P and D , as f increase above 1, the estimation ratios $\text{ER}_{f,P,00-10}$ and $\text{ER}_{f,D,00-10}$ increase. For most f values, these ratios are higher than $\text{Freq}_{ab,11-20}$.

The C in Figure 4A, as mentioned in Section 3.1.1, has discharge records starting from 2010, thus limiting its usability for data analysis (USNRC 2009). In Figure 4A, it was

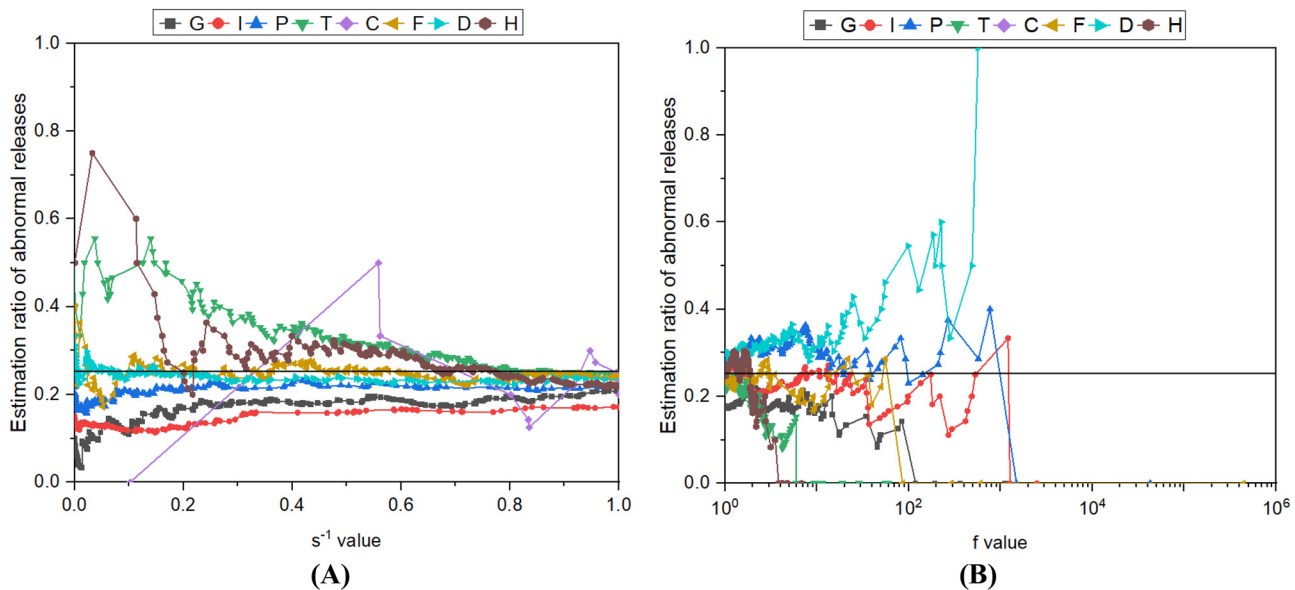


Figure 4: Estimation ratio of abnormal release by radionuclides (groups) based on RCF values for Group 1 discharge dataset of 681 RYs from 2000 to 2010 in US PWRs. (A) and (B) represent the estimation ratio of abnormal releases based on s^{-1} value and f value, respectively.

Table 4: Assessment of compliance with criteria for determining sensitive radionuclides (groups) with high correlation between the increases of the activities of radionuclides (groups) discharged and causal events for Group 1 discharge data for 681 RYs in US PWRs. If the criteria are met, mark as Yes (Y); if not satisfied, mark as No (N).

Radionuclides (groups)		Verification of satisfaction status of the criteria (values)					
		s^{-1}			f		
		C-①	C-②	C-③	C-①	C-②	C-③
Gaseous	<i>G</i>	N (0.102)	Y (0.58)	N (0 %)	N (−0.001)	N (0.44)	N (0 %)
	<i>I</i>	N (0.051)	Y (0.71)	N (0 %)	N (−2.01 × 10 ^{−5})	N (0.007)	N (31.4 %)
	<i>P</i>	N (0.037)	N (0.38)	N (0 %)	Y (5.91 × 10 ^{−5})	N (0.04)	Y (94.4 %)
	<i>T</i>	Y (−0.242)	Y (0.87)	N (74.6 %)	N (−0.003)	Y (0.77)	N (0 %)
	<i>C</i>	N (0.233)	N (0.28)	N (36.4 %)	–	–	–
Liquid	<i>F</i>	Y (−0.024)	N (0.11)	N (34.4 %)	N (−2.0 × 10 ^{−4})	N (0.007)	N (21.3 %)
	<i>D</i>	Y (−0.039)	N (0.40)	N (28.4 %)	Y (8.61 × 10 ^{−4})	Y (0.69)	Y (100 %)
	<i>H</i>	Y (−0.189)	Y (0.47)	N (62.1 %)	N (−0.007)	Y (0.58)	N (78.4 %)

confirmed that for s^{-1} values less than or equal to 0.1, the estimation ratio of abnormal releases $ER_{s^{-1}, C, 00-10}$ is 0 %, indicating that there are no NPPs corresponding to $RY_{(s^{-1} < RCF), ab, i, k}$ in Equation (3).

To ascertain the correlation between the increase of activities of radionuclides (groups) discharged due to abnormal releases in Figure 4 and the causal events, a linear regression analysis was conducted on the RCF values of Group 1 discharge data according to Equation (5). Subsequently, the satisfaction status and values of the criteria for identifying sensitive radionuclides (groups) with high correlation between the increases of activities of radionuclides (groups) discharged and events were summarized in Table 4.

In Table 4, the radionuclides (groups) that satisfy all criteria are *T* for the s^{-1} value and *D* for the f value. The radionuclides (groups) that satisfy C-① while also satisfying either C-② or C-③ are *H* for the s^{-1} value and *P* for the f value. The radionuclides (groups) that only satisfy C-① are *D* and *F* for the s^{-1} value. For all radionuclides (groups) that satisfy C-①, the reference values of RCF were determined. The radionuclides (groups) that meet these criteria are *T*, *F*, and *H* for the s^{-1} value, and *P* and *D* for the f value.

Based on the results in Table 4, *T*, *H*, *F* for the s^{-1} values, and *P*, *D* for the f values were prioritized as sensitive radionuclides (groups). The reference values for RCF selected from the Group 1 discharge data were determined based on the condition that $ER_{RCF, i, 00-10}$ is greater than $Freq_{ab, 11-20}$ (25.2 %) but not exceeding 100 %, and were chosen as the mean or median values of s^{-1} and f values. The reference values for RCF can be chosen by the user, and apart from the mean or median values, RCF values corresponding to estimation ratios of abnormal release $ER_{RCF, i, k}$ can also be selected according to the user's needs. The values of $ER_{RCF, i, 11-20}$ when applying the reference values to Group 2

Table 5: Estimation ratio of abnormal release when applying the reference values to Group 2 discharge data.

Radio-nuclides (groups)		Reference values ($ER_{RCF, i, 11-20}$)			
		s^{-1}		f	
		Median	Mean	Median	Mean
Gaseous	<i>P</i>	–	–	2.51 (26.79 %)	29.1 (35.71 %)
	<i>T</i>	0.603 (26.83 %)	0.541 (32.31 %)	–	–
Liquid	<i>F</i>	0.403 (29.41 %)	0.351 (31.17 %)	–	–
	<i>D</i>	–	–	3.99 (11.76 %)	23.3 (25 %)
	<i>H</i>	0.543 (30.77 %)	0.513 (30.99 %)	–	–

discharge data from 2011 to 2020 have been calculated and summarized in Table 5.

In Table 5, excluding *D*, it was observed that when the reference values were determined based on the mean and median values, they were respectively 1.1 to 1.2 times and 1.2 to 1.4 times higher than $Freq_{ab, 11-20}$, which is 25.2 %. For *D*, the estimation ratio of abnormal release was lower at 11.76 % when based on the median value and 25 % when based on the mean value. This discrepancy is likely due to the intermittent discharge of *D*, accounting for 121 RYs out of 691 RYs (19.55 %) during the period, suggesting insufficient discharge data for reliable analysis.

In Section 2.1, among the 705 events of multiple abnormal releases reported in US PWRs within a year, detailed information on the specific annual activities of radionuclides (groups) discharged attributed to actual abnormal releases was provided for 259 events. When examining the ratio of

the activities of radionuclides (groups) discharged during abnormal releases to the total annual activities of radionuclides (groups) discharged, it was found that the majority (accounting for 85 % of the total) had ratios of less than 20 %. The average ratio was 11.82 %, with a standard deviation of 28.06 %. This indicates that abnormal releases have a relatively minor impact on the total annual activities of radionuclides (groups) discharged, and there are many cases where a specific event of abnormal release does not dominantly influence the total annual activities of radionuclides (groups) discharged. Therefore, it may be challenging to establish a clear correlation between specific events and abnormal releases. However, by introducing the concept of RCF in this study, it was confirmed that abnormal release could be estimated at values exceeding approximately 1.1–1.5 times (26.79–35.71 %) the $\text{Freq}_{ab,00-20}$ value of 24.54 % for 1,300 RYs in US PWRs.

3.2 Reconstruction of major events related to abnormal releases from US PWRs using the database

From 2000 to 2020, a total of 319 RYs of abnormal releases occurred in the US, involving gaseous, liquids, and simultaneous gaseous and liquid releases. These events were categorized into six groups based on common characteristics identified in the integrated radioactive effluent database: primary system leakages, secondary system leakages, leakage of RWMS, leakage from defected SF during handling mentioned in Section 2.1 and additionally building (auxiliary building, containment building, fuel handling building, turbine building) leakages, and uncategorized events. Among these six categories, excluding building leakages and uncategorized events, analysis was conducted on four groups to examine the discharge characteristics. Building leakages, which primarily occurred at the Salem NPP in the US, involving abnormal releases through the containment equipment hatch, were excluded from the analysis due to their distinct characteristics. In 2014, 2015, and 2016, there were 28, 19, and 76 events, respectively, of the same event (PSEG 2014, 2015, 2016). This contrasts significantly with the maximum of 8 events per year occurring at other 61 NPPs excluding the Salem NPP. To ensure accurate result analysis, cases of abnormal release caused by building leakages were excluded due to their distinct characteristics.

Among the four groups, abnormal releases caused by leakage from GDT accounted for the highest proportion, with 97 RYs (30.4 %) out of the total 319 RYs. leakage from SG's PORV followed with 39 RYs (12.2 %) out of the total 319 RYs,

being the second highest proportion. Events where both leakage from GDT and leakage from SG's PORV occurred simultaneously in the same year totaled 3 RYs. In this study, detailed analysis was conducted on leakage from GDT, leakage from SG's PORV, and radionuclides (groups) involved in abnormal release clearly reported in leakage from defected SF during handling. In Equation (4), causal events j are denoted as GDT, SG, and Fuel, respectively.

3.2.1 Leakage from the gas decay tank

Out of the 1,300 RYs in the US, abnormal release due to leakage from GDT amounted to 97 RYs, resulting in an $\text{Freq}_{ab,GDT,00-20}$ of 7.46 % according to Equation (4). As mentioned in Section 2.3, abnormal release from leakage from GDT can result in the release of fission gases, including ^{85}Kr , ^{85m}Kr , ^{87}Kr , ^{88}Kr , ^{127}Xe , ^{131m}Xe , ^{133}Xe , ^{133m}Xe , ^{135}Xe , ^{135m}Xe , ^{137}Xe and ^{138}Xe . Figure 5 illustrates the RCF values when abnormal release occurs due to leakage from GDT. As mentioned in Section 2.2, if the s^{-1} value is less than 1 and the f value is greater than 1, it can be expected that the activities of radionuclides (groups) discharged will increase during abnormal release. While the average s^{-1} value in Figure 5A is not consistently less than 1, radionuclides with s^{-1} values less than 1 in over 50 % of cases include ^{138}Xe (85 %), ^{133m}Xe (59 %), ^{88}Kr (52 %), ^{135}Xe (51 %), ^{127}Xe (50 %), and ^{135m}Xe (50 %). The average value of f in Figure 5B is greater than 1 for all radionuclides, and the radionuclide with a ratio of over 50 % of cases where the actual f value exceeds 1 is ^{14}C (73 %).

When applying the criteria for determining the correlation between the increase of activities of radionuclides (groups) discharged and causal events as described in Equation (5) and Section 2.3, the radionuclides that satisfy all criteria (C-①, C-② and C-③) are ^{88}Kr , ^{137}Xe and ^{138}Xe in the f values. The radionuclides satisfying C-① while also meeting either C-② or C-③ are ^{85}Kr and ^{88}Kr in the s^{-1} values, and ^{85}Kr , ^{87}Kr , and ^{133m}Xe in the f values. Therefore, when considering the RCF values and the satisfaction of judgment criteria, radionuclides with a high correlation with the increase of the activities of radionuclides discharged and events are likely to be ^{88}Kr , ^{137}Xe and ^{138}Xe in the f values.

3.2.2 Leakage from the steam generator's power operated relief valve

Out of the 1,300 RYs in the US, abnormal release due to leakage from SG's PORV amounted to 39 RYs, resulting in an $\text{Freq}_{ab,SG,00-20}$ of 3 % as defined in Equation (4). This event could result in the leakage of gaseous tritium. While the average value of s^{-1} is not consistently less than 1, the

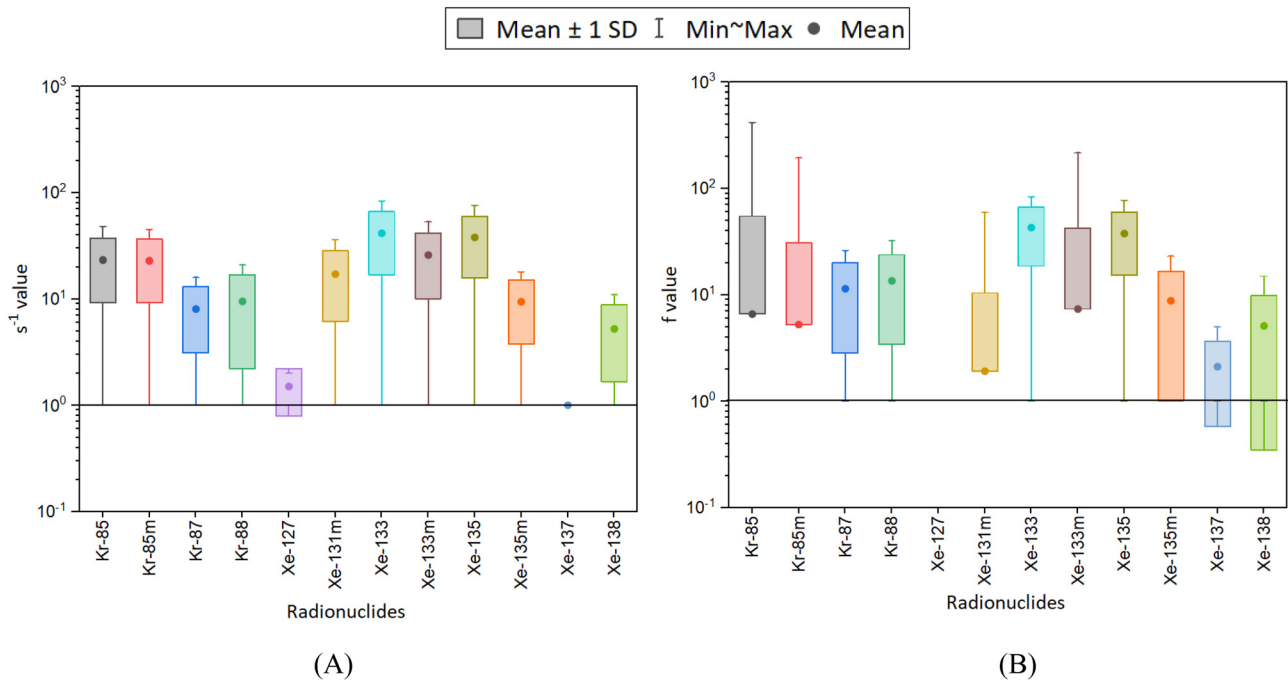


Figure 5: The RCF values of fission gases discharged abnormally due to leakage from the GDT in US PWRs. (A) and (B) represent the values of s^{-1} and f by radionuclides, respectively.

proportion of s^{-1} values less than 1 is 48.6 % when considering all s^{-1} values. Although the average value of f is greater than 1, the proportion of f values greater than 1 is 44.4 %, indicating that not all activities of radionuclides discharged increase during abnormal release causing events. While T does not satisfy all three criteria for determining the correlation between the increase of activities of radionuclides (groups) discharged and causal events as described in Equation (5) and Section 2.3, it does satisfy C-① and C-② for f values. Since C-①, which is essential in s^{-1} values, is not satisfied, it can be expected that the correlation between the increase of the activities of radionuclide discharged and causal events is higher in f values.

3.2.3 Leakage from defected spent fuel during handling

Out of the 1,300 RYs in the US, leakage from defected SF during handling amounted to 4 RYs, resulting in an $\text{Fre-}q_{ab, \text{Fuel}, 00-20}$ of 0.31 % as defined in Equation (4). As mentioned in Section 2.3, leakage from defected SF during handling results in the generation of T and C (ORNL 1977, 1980; USNRC 2020). It has been confirmed that fission gases emitted due to leakage from defected SF during handling include radionuclides such as ^{85}Kr , $^{85\text{m}}\text{Kr}$, ^{87}Kr , ^{88}Kr , $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$, and ^{135}Xe .

Figure 6 depicts the RCF values when there is abnormal release due to defected SF during handling. In Figure 6A, the average value of s^{-1} is less than 1 for the ^{14}C . The radionuclides ^{127}Xe and ^3H have proportions where 50 % are less than 1 of the s^{-1} value. The T and C, identified as radionuclides with a high estimation ratio of abnormal release in s^{-1} values in Section 3.1.2, correspond to radionuclides with high correlation between the increase of the activities of radionuclides and events. In Figure 6B, the f values for ^{85}Kr , ^{88}Kr , $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$ and ^{14}C have an average value greater than 1, and the ^{88}Kr corresponds to cases where all f values are greater than 1, with ^{14}C having a proportion of 75 % greater than 1. The radionuclide commonly associated with an increase in the activities when abnormal release occurs due to defected SF during handling is consistently ^{14}C .

Applying the criteria for determining correlation between the increase of the activities of radionuclides discharged and event in Equation (4) and Section 2.3, radionuclides that satisfy all three criteria are $^{133\text{m}}\text{Xe}$ for s^{-1} values, and ^{88}Kr and ^{14}C for f values. Isotopes that satisfy criterion ① while also meeting criterion ② are ^{85}Kr , $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$ and ^3H for f values. Among these radionuclides, ^{85}Kr , ^{133}Xe , $^{133\text{m}}\text{Xe}$ and ^3H have coefficient of determination, R^2 , all greater than 0.9, indicating a very high correlation between the increase of the activities or radionuclides discharge and leakage from defected SF during handling. Indeed,

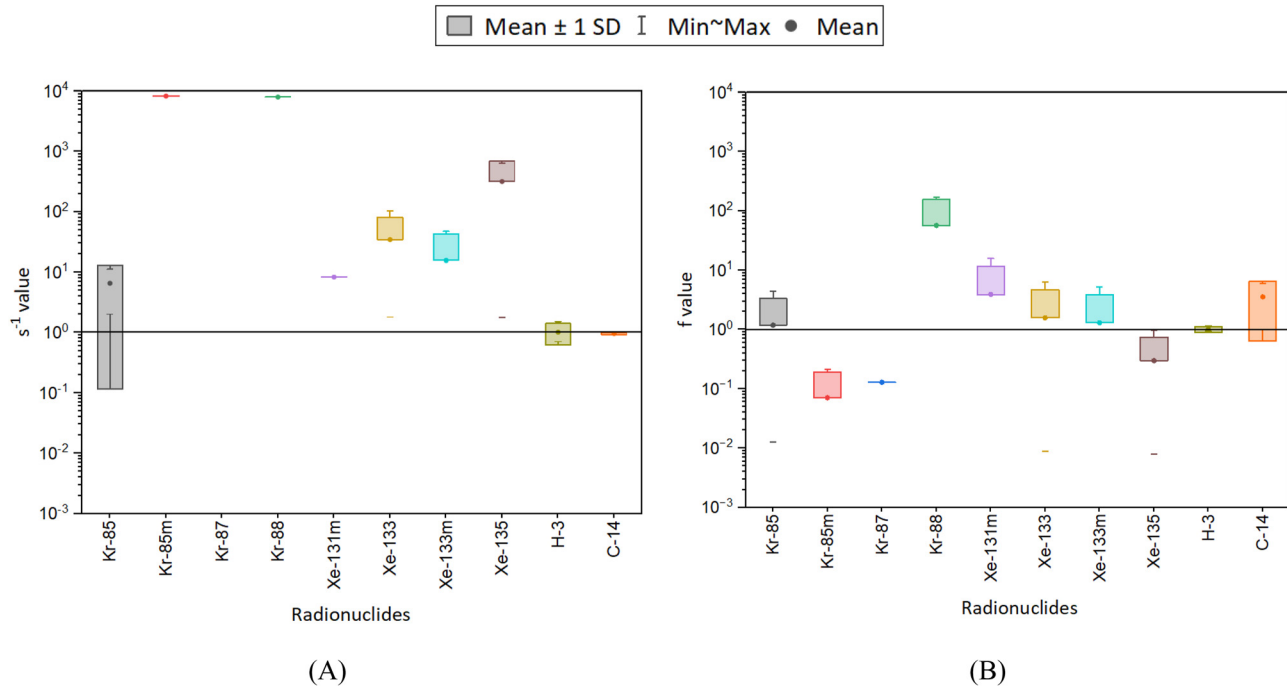


Figure 6: The RCF values of fission gases, ^3H , and ^{14}C discharged abnormally due to leakage from defected spent fuel during handling in US PWRs. (A) and (B) represent the values of s^{-1} and f by radionuclides, respectively.

abnormal releases of ^{85}Kr and ^{133}Xe were confirmed in cases of leakage from defected SF during handling at Calvert Cliffs Units 1 and 2, Catawba Unit 1, and Millstone Unit 2 (Dominion Energy 2015; Duke Energy 2015; Exelon 2016).

3.3 Study on the applicability of RCF to the discharge data of Korean PWRs

3.3.1 Derivation and analysis of RCF based on discharge data

RCF values for 310 RYs from 2000 to 2020 were derived for Korean PWRs, and their distribution characteristics were analyzed. Using Equations (1) and (2), s^{-1} and f values were calculated for radionuclides (groups) of each PWR for every RY, and the results were plotted in ascending order in Figure 7. This is comparable to the RCFs of US PWRs shown in Figure 2. On the y-axis of Figure 7, A represents the range where the value of s^{-1} is less than 1, and B indicates the range where f values are greater than 1, which is considered a significant range.

In Figure 7A and B, the radionuclides (groups) with steep slopes and narrow gaps between the exponential growth and logarithmic growth parts in the 'upward and downward parabola' type curve are ranked in descending order as *D*, *I*,

P, *F*, *G*, *C*, *T*, and *H*. Especially noteworthy are the top three ranked radionuclide groups, *D*, *I*, and *P*, which demonstrate results similar to those observed in Figure 2 for US PWRs. A steep slope indicates significant fluctuations in the annual activities of the corresponding radionuclides (groups). Reviewing the discharge data for Korean PWRs, it can be inferred that *D*, *I*, and *P* show relatively low frequencies of exceeding the Minimum Detectable Activity (MDA), with discharge frequencies of approximately 18 RYs (5.8 %), 92 RYs (30 %), and 111 RYs (36 %) out of 310 RYs, respectively. Consequently, it is anticipated that these radionuclide groups may exhibit larger fluctuations in the activities of radionuclides (groups) discharged due to their infrequent occurrences of exceeding the MDA.

Table 6 represents the distribution ratios of RCF values and statistical values of annual radioactive effluent release for 310 RYs from 2000 to 2020 for Korean PWRs. Comparing the statistical values of annual activities of radionuclides (groups) between Table 2 for US PWRs and Table 6 for Korean PWRs, it can be observed that Korean PWRs exhibit lower annual average activities, ranging from 6.14×10^{-5} to 0.82 times that of US PWRs. Furthermore, for radionuclides (groups) excluding *C* and *D*, the CV ranges from 0.23 to 0.67 times, indicating lower variability in annual activities compared to US PWRs. It is worth noting that discharge data for *C* has been recorded since 2012 in Korea, resulting in

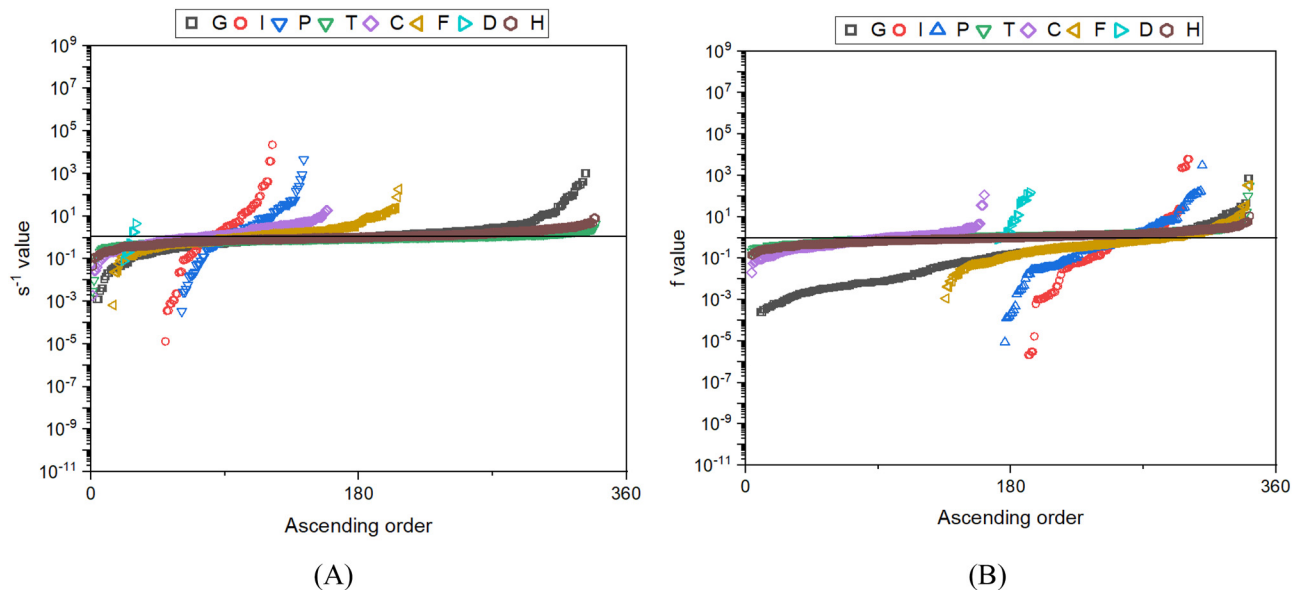


Figure 7: Distribution curves of RCF by radionuclides (groups) for 310 RYs in Korean PWRs. (A) and (B) represent the values of s^{-1} and f , respectively.

Table 6: The distribution ratio of RCF values and statistical values of annual activities by radionuclides (groups) for 310 RYs of Korean PWRs.

Radionuclides (groups)		Distribution ratio within the range of 0.1–10		Statistical values of annual activities	
		RCF		Mean	CV
		s^{-1}	f		
Gaseous	G	83 %	47 %	0.597	2.94
	I	30 %	18 %	1.93×10^{-5}	7.33
	P	34 %	23 %	1.53×10^{-5}	16.14
	T	99 %	98 %	2.46	1.02
	C	94 %	91 %	0.08	0.87
Liquid	F	82 %	47 %	2.11×10^{-4}	5.88
	D	27 %	7 %	2.72×10^{-7}	9.42
	H	100 %	99 %	8.11	0.66

limited available data. Additionally, D accounts for only 5.8 % of total discharge data among the 310 RYs, indicating a limited frequency of occurrence and suggesting limitations in comparison with statistical values from US PWRs.

Meanwhile, the proportion of radionuclides (groups) distributing within the range of 0.1–10 for s^{-1} values is highest in the order of H, T, C, G, F, P, I , and D , and this order remains the same for f values as well. In terms of CV, the order from lowest to highest is H, C, T, G, F, I, D, P , indicating a similarity to the order of proportions distributing within the range of 0.1–10. Similarly, in Table 2 for US PWRs, the top three radionuclides (groups) with the highest proportion of

distribution within the range of 0.1–10 are H, C , and T , parallel those of Korean PWRs, and the order of lowest CV is also similar to that of US PWRs.

In Figure 7, it can be observed that the distribution within the range of 0.1–10 is denser in Korean PWRs compared to that in US PWRs shown in Figure 2. Table 7 compares the statistical values of RCF corresponding to US PWRs in Figure 2 and Korean PWRs in Figure 7.

In Table 7, The σ value of RCF is smaller in all radionuclides (groups) for Korean PWRs compared to those for US PWRs in terms of s^{-1} values, and it is also smaller for all radionuclides (groups) except for T and C in terms of f values. When comparing the σ value with that of US PWRs, it can be observed that the annual activities of liquid and gaseous radionuclides (groups) in Korean PWRs exhibit less variability. Consequently, it is expected that Korean PWRs may exhibit less pronounced increases in activities of radionuclides (groups) during abnormal release causing events compared to US PWRs.

3.3.2 Estimation of correlation for specific event

In Korea, unlike the US, abnormal releases are not categorized and reported separately in annual radioactive effluent reports. Instead, the Korea Institute of Nuclear Safety (KINS) records accidents and failures in NPPs in OPIS (KINS 2024). However, identifying accidents or failures that may lead to abnormal releases individually is necessary. Nonetheless, cases that could be classified into the four groups of abnormal releases causes in Section 2.3 were not identifiable in OPIS.

Table 7: Comparison of statistical values of RCF between 1,300 RYs in the US and 310 RYs in Korean PWRs.

Radionuclides (groups)		Statistical values of RCF by countries									
		s^{-1}					f				
		USA ^a		Korea ^b		$\frac{\sigma_{U, s^{-1}}}{\sigma_{K, s^{-1}}}$	USA ^c		Korea ^d		$\frac{\sigma_{U, f}}{\sigma_{K, f}}$
		$\mu_{U, s^{-1}}$	$\sigma_{U, s^{-1}}$	$\mu_{K, s^{-1}}$	$\sigma_{K, s^{-1}}$		$\mu_{U, f}$	$\sigma_{U, f}$	$\mu_{K, f}$	$\sigma_{K, f}$	
Gaseous	<i>G</i>	30.1	272	15.9	90.5	3.0	7.55	60.2	6.01	57.3	1.1
	<i>I</i>	1.37×10^5	3.84×10^6	266	2,100	1.83×10^3	2,610	7.50×10^4	74.8	578	130
	<i>P</i>	5,350	7.69×10^4	50.5	388	198	2,270	6.90×10^4	14.8	175	394
	<i>T</i>	1,930	6.75×10^4	1.02	0.463	1.46×10^5	1.58	3.37	2.72	18	0.2
	<i>C</i>	96	1.14×10^4	2.18	2.92	3.90×10^3	1.09	0.637	2.08	9.7	0.1
Liquid	<i>F</i>	50	3.20×10^4	3.77	14.3	2.24×10^3	823	2.75×10^4	2.99	25.7	1,069
	<i>D</i>	9.7	352	0.33	0.91	388	18.2	215	3.73	18.3	11.7
	<i>H</i>	1.36	2.15	1.24	0.97	2.22	1.33	1.57	1.23	1.0	1.5

^a $\mu_{U, s^{-1}}$ and $\sigma_{U, s^{-1}}$ refer to the mean and standard deviation values of s^{-1} for the US PWRs. ^b $\mu_{K, s^{-1}}$ and $\sigma_{K, s^{-1}}$ refer to the mean and standard deviation values of s^{-1} for Korean PWRs. ^c $\mu_{U, f}$ and $\sigma_{U, f}$ refer to the mean and standard deviation values of f for the US PWRs. ^d $\mu_{K, f}$ and $\sigma_{K, f}$ refer to the mean and standard deviation values of f for Korean PWRs.

Therefore, a search for cases of abnormal release was conducted by referring to publicly available Safety Regulation Status for NPPs from 2000 to 2020 (NSSC 2024). From these data, events leading to abnormal release from 2000 to 2020 were expected to include the secondary system leakages and leakage from fuel defects. For these two groups of events, an analysis was conducted on the correlation between the increase of the activities of radionuclides (groups) discharged and causal events.

Additionally, using data recording the number of fuel defects from 1978 to 2003 for each NPP (Kori Units 1 to 4,

Hanbit Units 1 to 5, and Hanul Units 1 to 4), events were defined as fuel defects, and occurrences were counted based on records from 2000 to 2003. For Korean PWRs from 2000 to 2020, abnormal release occurred in 1 RY due to leakage from SG's valve and in 30 RYs due to leakage from fuel defects, resulting in an $\text{Freq}_{ab, 00-20}$ of 10 % (=31 RYs/310 RYs).

In Korean PWRs, the fission gases discharged during normal operation include ^{79}Kr , ^{85}Kr , $^{85\text{m}}\text{Kr}$, ^{87}Kr , ^{88}Kr , $^{129\text{m}}\text{Xe}$, $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$, ^{135}Xe , and $^{135\text{m}}\text{Xe}$. The fission gases discharged due to fuel defects in Korean PWRs are ^{85}Kr , $^{85\text{m}}\text{Kr}$,

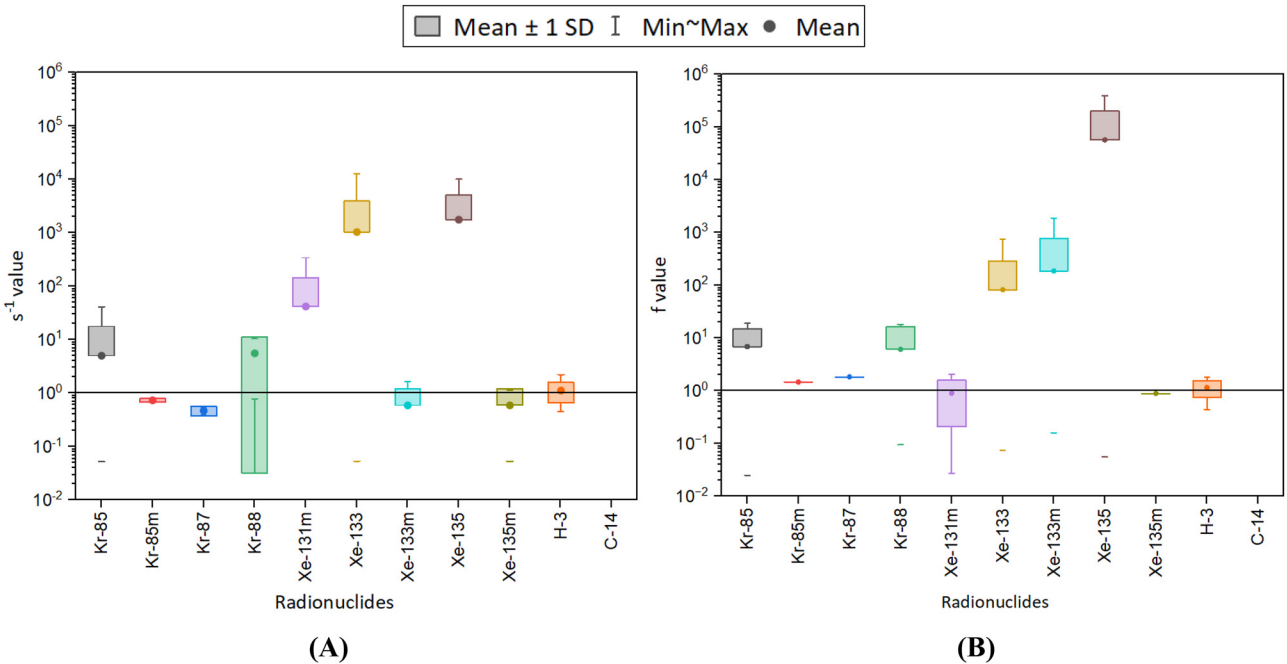


Figure 8: The RCF values for the abnormal release of fission gases, ^3H , and ^{14}C of leakage from fuel defects in Korean PWRs. (A) and (B) represent the values of s^{-1} and f , respectively.

^{87}Kr , ^{88}Kr , $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$, ^{135}Xe , and $^{135\text{m}}\text{Xe}$, as well as ^3H and ^{14}C .

Figure 8 represents the RCF values when abnormal release due to fuel defects in Korean PWRs. As mentioned in Section 3.2, it can be expected that the activities of radionuclides discharged will increase when the s^{-1} value is less than 1 and the f value is greater than 1. In Figure 8A, radionuclides with average values less than 1 include $^{85\text{m}}\text{Kr}$, ^{87}Kr , $^{133\text{m}}\text{Xe}$, and $^{135\text{m}}\text{Xe}$. Radionuclides with a ratio of s^{-1} values less than 1 exceeding 50 % include $^{85\text{m}}\text{Kr}$ (100 %), ^{87}Kr (100 %), $^{133\text{m}}\text{Xe}$ (80 %), $^{131\text{m}}\text{Xe}$ (73 %), ^{85}Kr (50 %), ^{88}Kr (50 %), and $^{135\text{m}}\text{Xe}$ (50 %). In Figure 8B, radionuclides with average f values greater than 1 include ^{85}Kr , $^{85\text{m}}\text{Kr}$, ^{87}Kr , ^{88}Kr , ^{133}Xe , $^{133\text{m}}\text{Xe}$, ^{135}Xe , and ^3H . Radionuclides with a ratio of f values greater than 1 exceeding 50 % include $^{85\text{m}}\text{Kr}$ (100 %), ^{133}Xe (89 %), ^3H (67 %), ^{85}Kr (63 %), and ^{135}Xe (57 %). The radionuclides commonly associated with an increase in the activities when abnormal release occurs due to fuel defects are consistently ^{85}Kr and $^{85\text{m}}\text{Kr}$.

When applying the three criteria to derive sensitive radionuclides highly correlated with an increase in activities in Korean PWRs using Equation (5), $\text{Freq}_{ab, \text{Fuel}, 00-20}$ is 9.68 % (=30 RYs/310 RYs). Radionuclides satisfying C-① for the s^{-1} value include ^{85}Kr and ^{133}Xe , with ^{133}Xe also meeting C-③. For the f value, radionuclides satisfying all criteria, C-①, C-②, and C-③, are ^{85}Kr and ^{135}Xe , while radionuclides satisfying C-① and C-② are $^{85\text{m}}\text{Kr}$, ^{87}Kr , and ^{88}Kr . Among these, ^{85}Kr is deemed to be the radionuclide with the highest correlation with an increase in the activities due to fuel defects.

When comparing the abnormal release due to leakage from defected SF during handling between US PWRs in Figure 6 and the Korean PWRs in Figure 8, it was observed that common radionuclides discharged abnormally include ^{85}Kr , $^{85\text{m}}\text{Kr}$, ^{87}Kr , ^{88}Kr , $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$, and ^{135}Xe , as well as ^3H , and ^{14}C . In both countries, the radionuclides most strongly correlated with abnormal releases and an increase in the activities due to leakage from defected SF during handling are ^{85}Kr and ^{88}Kr .

4 Conclusions

In this study, radioactive discharge data, including liquid and gaseous radionuclides (groups), from 62 US PWRs (1,300 RYs) and 22 Korean PWRs (310 RYs) were collected, processed, and integrated into a separate radioactive effluent database. Previous studies had limitations in simply documenting the increase of the quantity of radionuclides (groups) discharged due to causal events and failing to identify the correlation between events and abnormal releases. However, in this study, a new concept of RCF is

proposed to determine abnormality of radioactive discharges from PWRs. The applicability of RCF was demonstrated by calculating the estimation ratio of abnormal releases each radionuclide (group) based on actual discharge data from US and Korean PWRs. Through application of the RCF concept to US PWR discharge data, the probability of correct estimation of abnormality ranges 27–36 %, which is approximately 1.1–1.5 times higher than the frequency of abnormal releases. The relatively modest estimation power of this new methodology turned out to be because the proportion of radioactivity released due to a specific single event is not significant in the context of the annual radioactivity release.

By applying the concept of RCF to abnormal release data of specific causal events, the study systematically and comprehensively analyzed the correlation between the increase in the activities of radionuclides (groups) discharged and causal events, including leakage from GDT, leakage from SG's PORV, and leakage from defected SF during handling in US PWRs. Consequently, sensitive radionuclides were identified to comprehend the correlation between the increase of discharges and causal events in both countries. The sensitive radionuclides to each specific causal event were derived as ^{88}Kr , ^{137}Xe and ^{138}Xe for leakage from GDT and ^3H for leakage from SG's PORV. Regarding the leakage from defected SF during handling, ^{85}Kr , ^{88}Kr , ^{133}Xe , $^{133\text{m}}\text{Xe}$, ^3H and ^{14}C turned out to be sensitive to US PWRs, whereas ^{85}Kr and ^{88}Kr were sensitive to Korean PWRs.

As a result of this study, operators can identify previously unidentified events and changes in discharge trends through the correlation between discharge and event information, enabling them to take corrective actions and increase safety. Regulators may estimate the possible abnormality and potential correlation with specific causal event by reviewing a series of discharge data reported as normal discharge, and then may take appropriate measures such as detailed review or investigation from a safety perspective.

Abbreviations

ARERR	Annual Radioactive Effluent Release Report
ACR-1000	Advanced Candu Reactor 1000
AP1000	Advanced Passive 1000
BWR	boiling water reactor
CRUD	Chalk River unidentified deposits
CV	coefficient of variation
EA	Environment Agency
EPR	European Pressurized Reactor
ESBWR	Economic Simplified Boiling Water Reactor
GDT	gas decay tank
GRWMS	gaseous radioactive waste management system

IAEA	International Atomic Energy Agency
KINS	Korea Institute of Nuclear Safety
MDA	Minimum Detectable Activity
NPP	nuclear power plant
NSSC	Nuclear Safety and Security Commission
OPIS	Operational Performance Information System for Nuclear Power Plant
PORV	power operated relief valve
PWR	pressurized water reactor
RCF	release correlation factor
RWMS	radioactive waste management system
RY	reactor-year
SF	spent fuel
SFP	spent fuel pool
SG	steam generator
USNRC	United States Nuclear Regulatory Commission

Symbols

i	radionuclides (groups)
j	events
n	specific year within the period from 2001 to 2020
k	period
$A_{n,i}$	annual activities of radionuclide (group) i at a specific NPP in year n
$A_{n-1,i}$	annual activities of radionuclide (group) i at a specific NPP in year $(n - 1)$
$s_{n,i}$	activities for year n by radionuclide (group) i relative to the annual discharge for the previous year $(n - 1)$
$\frac{s_{n,i}^{-1}}{A_{n-1,i}}$	reciprocal of $s_{n,i}$
$\bar{A}_{n,i}$	average activities of radionuclide (group) i over the period up to year $(n - 1)$, starting from the year 2000
$f_{n,i}$	value of $A_{n,i}$ divided by $\bar{A}_{n-1,i}$
$ER_{RCF,i,k}$	estimation ratio of abnormal release for radionuclide (group) i for a period of k years based on the RCF value
$RY_{(s^{-1} < RCF), ab, i, k}$	the number of reactor years (RYs) corresponding to abnormal release for radionuclide (group) i for a period of k years when s^{-1} is less than RCF
$RY_{(f > RCF), ab, i, k}$	the number of reactor years (RYs) corresponding to abnormal release for radionuclide (group) i for a period of k years when f is greater than RCF
$RY_{(s^{-1} < RCF), tot, i, k}$	total number of RYs for radionuclide (group) i for a period of k years when s^{-1} is less than RCF
$RY_{(f > RCF), tot, i, k}$	the total number of RYs for radionuclide (group) i for a period of k years when f is greater than RCF
$Freq_{ab, j, k}$	frequency of abnormal release caused by any events j , regardless of RCF, for a period of k years
$RY_{ab, j, k}$	the number of RYs for a period of k years affected by the abnormal release due to the event causing j
$RY_{tot, k}$	total RYs for a period of k years
$ER_{RCF, i, j, k}$	estimation ratio of abnormal release caused by event j for a period of k years, for radionuclide (group) i ,
$RCF_{i, j, k}$	RCF value when main causal event j occurs for radionuclide (group) i for a period of k years

a	slope
b	constant
σ	standard deviation
G	fission products
I	radioiodine
P	particulates
T	gaseous tritium
C	^{14}C
F	fission and activation products
D	dissolved gases
H	liquid tritium

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