



Original article

New methodologies to derive discharge limits considering operational flexibility of radioactive effluents from Korean nuclear power plants based on historical discharge data

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ABSTRACT

The new methodologies to derive discharge limits considering operational flexibility according to international safety standards were developed to help reduce the environmental releases of radioactive effluents from nuclear power plants (NPPs). To overcome the limitations of the two existing methods to set up discharge limits assuming a specific statistical distribution of the effluent discharge, two modified equations were newly proposed to directly derive a particular discharge limits corresponding to the target 'compliance probability' based on the actual annual discharge data for a specific NPP and radionuclide groups. By applying these to the actual yearly discharge data of 14 Korean NPPs for 7 radionuclide groups for the past 20 years, the applicability of two new methodologies to actual cases was demonstrated. The 'characteristic value' with approximately a 90% compliance probability for each Korean NPP and radionuclide group was proposed based on the results. The new approaches for setting up the discharge limits and the characteristic values developed in this study are expected to be effectively utilized to foster operator's efforts to progressively reduce the environmental releases of radioactive effluents of NPPs relative to the previous discharge data considering operational flexibilities.

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1. Introduction

Liquid and gaseous radioactive materials are released into the environment during nuclear power plant (NPP) operation. To reasonably reduce the effect of radioactive materials released into the environment, each country endeavors to follow the As Low As Reasonably Achievable (ALARA) and Best Available Technique (BAT) principles as a concept of optimization [1–4]. The regulatory authority of each country sets up the discharge limits of radioactive effluents for facilities and activities to optimize public protection [2–6]. It is a general rule to set up the discharge limits of NPPs as the radiation dose constraints (e.g. mSv/y), which is lower than the dose limit of the public. In reality, various quantities are being applied, including the concentration limits of radionuclides (e.g. Bq/m³) in the exclusion area boundary (EAB), equivalent to the dose constraints and radioactivity limits being released to the environment (e.g. Bq/y) [2,7,8].

The International Atomic Energy Agency (IAEA) proposes the

concept of dose constraints and optimized discharge limits setup, an example of which is presented in Fig. 1 [2]. As shown in Fig. 1, the specific dose constraints (B), considering the characteristics of sites and specific facilities or activities, is a starting point for the optimization process to find the optimum discharge levels. To set up the optimized discharge limits, the BAT principle, a concept of optimization for the (C) region based on the specific dose constraints, is applied, and the operational flexibility (E) should be considered during this process [2]. (E) Includes the operational flexibility and anticipated operational occurrence (AOO), and historical data of similar facilities can provide helpful information about the minimum allowance for flexibility [2]. The OECD/NEA claims that the discharge of actual effluents from a NPP is below the dose constraints, defines (E) in Fig. 1 as a headroom, and claims that the reduction of discharge can be achieved by the application of BAT or the reduction of headroom [4]. The Environment Agency (EA) in England espouses the progressive reduction in the release of radioactive effluents. It proposes a rule to set up the discharge limits for minimizing the expected headroom during normal operation [3]. In other words, these agencies recommend deriving the discharge limits by reflecting operational flexibility considering

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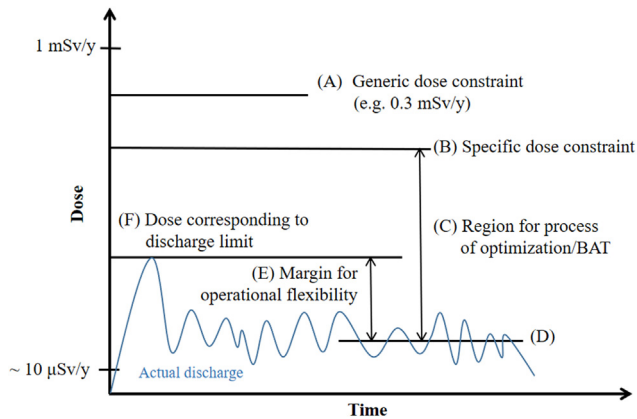


Fig. 1. Concept of setting up the discharge limit based on the specific dose constraint and operational flexibility. Specific dose constraint (B) can be larger than the generic dose constraint (A) [2].

the characteristics of radioactive effluents releases from actual NPPs.

The EA ratified all the standards proposed by the IAEA, shown in Fig. 1, into the setup for the discharge limit [3,9,10]. It considered the existing discharge limit based on dose constraints (B) alongside the BAT principle (C). It set up the discharge limits (F) reflecting (E) based on the actual discharge of the NPPs. However, some nations do not separately demand the (C) process and regulate using (B); hence, they do not comply with the IAEA standards. For instance, dose constraints are established on Appendix I to 10 Code of Federal Regulations (CFR) 50 and concentration limits of radionuclides are established on Appendix B to 10 CFR 20, part of the United States Nuclear Regulatory Commission (NRC) for setting up the discharge limits [5,6]. The Canadian Nuclear Safety Commission (CNSC) set up the derived discharge limits (Bq/y) deduced by the dose limit or dose constraints for the CANDU. Using a similar method, the radioactivity limits (Bq/y) are deduced by dose constraints for some NPPs in Hungary [11,12].

The discharge limits for all NPPs are the dose constraints (B) shown in Fig. 1 and the concentration limits of radionuclides of EAB in Korea [7]. As the Nuclear Safety Act was revised according to the recent trend of the international organization, the maximum permitted discharge radioactivity per radionuclide groups were proposed by preparing the discharge plan (in this study, the amount of radioactivity and radioactive effluents discharge were regarded as the same) [13]. In Korea, optimization (C) was carried out by presenting the values approved by the Safety Analysis Report [14]. However, the discharge limits considering the operational flexibility have not yet been set up, and there is no publicly available study on this subject.

Meanwhile, the Environmental Protection Agency (EPA) proposes a methodology for setting up the discharge limit to reduce or remove environmentally released contaminants outside the nuclear field [15–18]. The discharge limit is calculated based on the statistics of the pollutant concentration or effluent toxicity released from existing facilities. This is similar to setting up the discharge limit considering the operational flexibility mentioned by the IAEA. However, it has limitations in that this method is non-nuclear field-based and deduces the discharge limit assuming a statistical distribution.

The EA, applying standard of setting up the discharge limit proposed by the IAEA, has reported effluent discharge by including expected events (anticipated but unplanned events likely to happen during the lifetime of the reactor), which is the value of the design basis source term and is similar to the concept of anticipated

operational occurrences (AOO) in normal operation [19]. However, because the EA proposes an equation that separates the normal operation and expected events for setting up the discharge limit, it has limitations in its application to NPPs in the USA or Korea in which the discharge including AOO is reported [12,20–25].

Therefore, this study aimed to utilize the discharge data of actual operating NPPs under the international standards of the IAEA and developed new methodologies for setting up the discharge limits for the progressive reduction of radioactive effluents releases. To overcome the limitations of the existing methods to set up discharge limits (i.e., the assumption of a specific probability distribution such as normal distribution or log-normal distribution), two modified equations (namely EA-based and EPA-based equations) were proposed. Furthermore, the characteristic value set for each NPP and radionuclide group, which presents a specific compliance probability to the discharge limits, was deduced based on the distribution of actual annual discharge data of Korean NPPs. Furthermore, the effect of the discharge limits setup period (10 years or 1 year) on the specific compliance probability to the discharge limits was analyzed.

2. Materials and methods

2.1. Collection of radioactive effluents discharge data and selection of radionuclide groups

In this study, 21 NPPs located at Kori, Hanbit, Hanul, and Wolsong sites, operating in January 2021, were selected as subjects for evaluation (Shin Kori Unit 3, Shin Kori Unit 4, and Shin Wolsong Unit 2 started to operate in 2016, 2019, and 2015 and did not have enough discharge data; thus, they were excluded). Table 1 summarizes the basic information about NPPs subject to evaluation, along with the analysis types conducted in this study. First, the radioactive effluents discharge data of the NPPs were analyzed for the year following the commercial operation. In addition, the discharge data from 2000 to 2009 were collected from ‘Radiation management yearbook’ and the more recent data from 2010 to 2019 were obtained from ‘Survey of radiation environment and assessment of radiological impact on environment in vicinity of nuclear power facilities’ [24,25]. It is noted that discharge data collected from both sources are credible and comparable each other for the purpose of this study, since they were officially reported by the monopoly operator, Korea Hydro & Nuclear Power Co. Ltd. (a single operating organization of all Korean NPPs) and the level of detail of the data from the two sources are equivalent (i.e. radionuclide-specific annual discharge data for liquid and gaseous effluents from NPPs). For convenience, each NPP was assigned a random mark, from K01 to K21.

Table 1 shows the correlation with the electrical output or design capacity, which affects the discharge characteristics of the radioactive effluents of 21 NPPs (for Type 1 analysis, refer to Section 3.1). The discharge trend for each radionuclide group of 18 NPPs operating for more than 10 years was quantitatively analyzed using the Mann–Kendall Trend Test (for Type II, refer to Section 3.2). The discharge characteristics for each year, each NPP, and each radionuclide group of 14 NPPs were identified using 20-year data, and the applicability of the discharge limits proposed in this study was examined (for Type III, refer to Section 3.3).

There are various classifications for each radionuclide group depending on the characteristics of the radioactive effluents in each country [26]. In this study, the Korean classification standard for the radionuclide groups of radioactive effluents was applied. The radionuclide groups of radioactive gaseous effluents are classified into five radionuclide groups: (1) fission products (G), (2) radioactive iodines (I), (3) particulates (P), (4) gaseous tritium (T), and (5)

^{14}C (C), and those of liquid radioactive effluents are classified into two radionuclide groups, namely (1) fission and activation products (F), and (2) liquid tritium (H). The dissolved and entrained gases were not discharged from all 21 NPPs subjected to the analysis over 15 years; hence, they were excluded from the study. Furthermore, the radioactive effluent release records of liquid ^{14}C were only found in four plants; thus, it was excluded.

2.2. Mann–Kendall Trend Test

To set up the optimized discharge limits considering the operational flexibility recommended by the IAEA, an analysis of the discharge characteristics of the radioactive effluents from actual NPPs is required [2]. Therefore, for the quantitative analysis of the discharge characteristics of the radioactive effluents, the Mann–Kendall Trend Test (M – K Test), which is widely used for tendency analysis of time series functions, was utilized in this study [26,27].

A null hypothesis (H_0) of the M – K Test is that ‘there is no tendency’ in the time series function. In this study, the time series function $X = \{x_1, x_2, \dots, x_n\}$ is a function over time, x_i is the data value at time i , and n is the number of data samples, signifying the number of data values used for this study’s NPPs. The statistic S was calculated as shown in Eq. (1).

$$S = \sum_{i=1}^{n-1} \sum_{j=i+1}^n \text{sgn}(x_j - x_i), j > i \quad (1)$$

where x_i and x_j are data values at times i and j , respectively, and the sine function $\text{sgn}(x_j - x_i)$ is defined as shown in Eq. (2).

$$\text{sgn}(x_j - x_i) = \begin{cases} 1 & ((x_j - x_i) > 0) \\ 0 & ((x_j - x_i) = 0) \\ -1 & ((x_j - x_i) < 0) \end{cases} \quad (2)$$

If the sample size n is larger than 10, the statistic S is regarded as a normal distribution, and the Z value of the standard normal distribution is calculated as shown in Eq. (3).

$$Z = \begin{cases} \frac{S-1}{\sqrt{V(S)}} & (S > 0) \\ 0 & (S = 0) \\ \frac{S+1}{\sqrt{V(S)}} & (S < 0) \end{cases} \quad (3)$$

Eq. (3) is considered ‘decreasing tendency’ if the Z value is negative, ‘increasing tendency’ if the Z value is positive, and ‘no tendency’ if the Z value is 0. If the calculated Z value satisfies the condition of $|Z| > Z_{1-\alpha/2}$, the null hypothesis (H_0) is rejected, and it is judged that an increasing or decreasing tendency exists. If it does not satisfy the condition, the null hypothesis (H_0) is adopted, suggesting no tendency in the time series function. The value of $|Z| > Z_{1-\alpha/2}$ can be identified in the standard normal distribution table with a significance level of α . In this study, a confidence interval of $\alpha = 0.05$ was used. If $|Z| > 1.96$ is valid in the 5% confidence interval, the null hypothesis (H_0) is rejected.

2.3. Methodology for setting up the discharge limits of radioactive effluents

In Korea, the discharge limits for regulation are radiation dose constraints and concentration limits of radionuclides in EAB [7]. The radiation dose constraints of Korea follow the USA dose constraint standards for the radioactive effluents—10 CFR 50, Appendix I [5]. The concentration of radionuclides in EAB was derived based on the public dose limit, and the discharge management standards for exhaust/drainage for each radionuclide group were applied [7]. As a new policy for submitting the discharge plan was introduced in Korea in 2015, it is necessary to rationally set up the maximum permitted discharge radioactivity per each radionuclide group (discharge limit) alongside the existing discharge limits [13]. However, the method for setting the discharge limits that will be introduced is under review. Accordingly, in this study, new methodologies for deriving discharge limits that improved the limitations of the existing methods was developed. It was confirmed that

Table 1
Basic information about the Korean operating NPPs subjected to evaluation and the analysis types.

No.	Reactor	Type*	Design capacity (MWe)	Commercial operation date	Period of used data	Types of analysis**		
						I	II	III
1	Kori-2	PWR	650	Jul 1983	2000–2019	✓	✓	✓
2	Kori-3	PWR	950	Sep 1985		✓	✓	✓
3	Kori-4	PWR	950	Apr 1986		✓	✓	✓
4	Shin Kori-1	PWR	1000	Feb 2011		✓	–	–
5	Shin Kori-2	PWR	1000	Jul 2012	2013–2019	✓	–	–
6	Hanbit-1	PWR	950	Aug 1986	2000–2019	✓	✓	✓
7	Hanbit-2	PWR	950	Jun 1987		✓	✓	✓
8	Hanbit-3	PWR	1000	Mar 1995		✓	✓	✓
9	Hanbit-4	PWR	1000	Jan 1996		✓	✓	✓
10	Hanbit-5	PWR	1000	May 2002	2003–2019	✓	✓	–
11	Hanbit-6	PWR	1000	Dec 2002		✓	✓	–
12	Hanul-1	PWR	950	Sep 1988	2000–2019	✓	✓	✓
13	Hanul-2	PWR	950	Sep 1989		✓	✓	✓
14	Hanul-3	PWR	1000	Aug 1998		✓	✓	✓
15	Hanul-4	PWR	1000	Dec 1999		✓	✓	✓
16	Hanul-5	PWR	1000	Jul 2004	2005–2019	✓	✓	–
17	Hanul-6	PWR	1000	Apr 2005	2006–2019	✓	✓	–
18	Wolsong-2	PHWR	700	Jul 1997	2000–2019	✓	✓	✓
19	Wolsong-3	PHWR	700	Jul 1998		✓	✓	✓
20	Wolsong-4	PHWR	700	Oct 1999		✓	✓	✓
21	Shin Wolsong-1	PWR	1000	Jul 2012	2013–2019	✓	–	–

* PWR and PHWR represent pressurized water reactor and pressurized heavy water reactor, respectively.

** Types of analysis are described in Section 3, and Type I, Type II, and Type III are the correlation analysis between the discharge and electrical output or design capacity, quantitative analysis utilizing the Mann–Kendall trend test, and identification of application of the discharge limits proposed in this study, respectively.

the derived discharge limits apply to Korean NPPs.

Although IAEA explains the reasonable concept of setting up the discharge limits considering operational flexibility, as presented in Fig. 1, a detailed method for deriving the flexibility has not been proposed [2]. However, in EA, the discharge limit setting up a method that satisfies the conditions in Fig. 1 was proposed and applied to setting up the discharge limits for new NPPs [9,10]. Therefore, Eq. (4) presents a method for setting up the discharge limits for the latest UK HPR1000 and UK ABWR NPPs proposed by EA in England:

$$A_i^{\text{proposed}} = A_i \cdot \frac{\mu_i + k \cdot \sigma_i}{\mu_i} + COEE_i \quad (4)$$

where A_i^{proposed} is the proposed discharge limit of radionuclide i (TBq), A_i is the expected discharge of radionuclide i for a new NPP (TBq), μ_i is the average discharge of radionuclide i of a similar NPP (TBq/y), and k , is a coverage factor that is the real number in the normal distribution curve [9,10]. For the normal distribution curve, in the one-sided normal distribution, k was 1.282 at a confidence interval of 90%. σ_i is the standard deviation for the discharge of radionuclide, i , of a similar NPP (TBq/y). $COEE_i$ represents the discharge of radionuclide i due to the expected event (TBq).

Eq. (4) proposed by EA has the limitation that it sets up the discharge limits assuming that the discharge characteristics of the radioactive effluents follow a normal distribution. Furthermore, in countries where the annual discharge data for normal operation and AOO are reported, it is difficult to derive the discharge limits by separating the anticipated/expected event proposed by EA [21–25]. Therefore, a new methodology for setting up the discharge limits was proposed utilizing the statistics of the radioactive effluents discharge of the actual operating NPPs and actual radioactive effluents discharge data for both normal operations and AOO by modifying these limitations. Eq. (5) presents the proposed method for setting up discharge limit:

$$DL_{i,t} = \mu_{i,t} + l \cdot \sigma_{i,t} \quad (5)$$

where $DL_{i,t}$ is the discharge limit of the radionuclide groups i for the past t years (TBq/y), $\mu_{i,t}$ is the average discharge of the radionuclide groups i for the past t years (TBq/y), and l is the real number, characteristic value which can be set up by the operator or regulator depending on the characteristics of each NPP and radionuclide group. $\sigma_{i,t}$ is the standard deviation for the discharge of radionuclide groups i for the past t years (TBq/y).

Meanwhile, the EPA proposes a methodology for setting up the discharge limits for the contaminants released to the environment of the fields outside of nuclear energy [15–18]. The method for setting the discharge limit for a contaminant is shown in Eq. (6):

$$L = LTA \cdot VF \quad (6)$$

where L is the discharge limit of the contaminant (mg/L), LTA is the long-term average of the contaminant concentration (mg/L), and VF is the variability factor. In the EPA, the 99% and 95% percentiles were used to express the daily maximum VF and average monthly VF, respectively [18]. Eq. (6) has the limitation that it is used in fields outside of nuclear energy and assumes that the discharge of the contaminant follows a log-normal distribution. Therefore, the discharge limit for each radionuclide group was derived based on the actual annual radioactive effluents discharge for each NPP, rather than assuming a specific distribution. Eq. (7) presents the proposed method for setting up the discharge limit:

$$DL_{i,t} = m \cdot \mu_{i,t} \quad (7)$$

where $DL_{i,t}$ is the discharge limit of the radionuclide groups i for the past t years (TBq/y), m is the real number that indicates characteristic value, and $\mu_{i,t}$ is the average discharge of radionuclide groups i for the past t years (TBq/y).

3. Results and discussion

3.1. Discharge characteristics of the liquid and gaseous radioactive effluents

The amount of discharged radioactive effluents for each nuclear facility has been reported [19,21,24,25]. The discharge of the radioactive effluents for each radionuclide group was analyzed and reported using various methods, including the raw (non-normalized) annual discharge (TBq/y) [28,29], normalization of design capacity (TBq/y per MWe) [30,31], and normalization of annual electrical output (TBq/y per GWh or TBq/y per MWh) [4,32–34]. There is a case that the analysis was carried out with the unit of annual electrical output (electrical energy generated) at the EA to identify the correlation between the electrical output of the nuclear reactor and discharge of the radioactive effluents to the environment [32]. The EA report claimed that most NPPs subjected to the survey show two mixed characteristics of ‘no correlation’ or ‘proportional relationship.’ However, a normalization method has not been proposed considering publicly available evidence on the correlation between the design capacity of a facility and radioactive effluents discharge. Therefore, the correlation between the radioactive effluents discharge and the design capacity of the facility or electrical output was analyzed to identify the availability of the discharge limits (Eq. (5) based in EA and Eq. (7) based in EPA) derived for the Korean NPPs and to select the unit suitable for the discharge characteristics of the radioactive effluents of the Korean NPPs. The normalized discharge of the radioactive effluents can be presented using Eq. (8) and (9):

$$N_1 = \frac{N_0}{P} \quad (8)$$

$$N_2 = \frac{N_0}{P \cdot t} \quad (9)$$

where N_0 is the raw (non-normalized) value of the actual radioactive effluents discharge (TBq/y), N_1 is the discharge normalized by the design capacity (TBq/y per MWe), and P is the design capacity of the NPP (MWe). N_2 represents the discharge normalized by the electrical output (TBq/y per MWh), and t is the operating hours of the NPP for 1 year (h).

3.1.1. Correlation between the electrical output of the NPPs and the radioactive effluents discharge

Of the three units expressed for the annual radioactive effluents discharge, the correlation of electrical output with the radioactive effluents discharge of the Korean NPP was investigated using two normalized methods (normalized by the design capacity of the facility and electrical output). First, to identify the correlation between the annual electrical output and the annual discharge, which is one of the methods, the correlation coefficient (R) between the radioactive effluents discharge of 21 Korean NPPs and the annual electrical output for the past 20 years was compared. The R -value is a numerical value representing the degree of correlation between the two variables. It is interpreted as a weak correlation if the R -value is 0.35 or below, a moderate correlation between 0.36 and

0.67, and a strong correlation between 0.68 and 1 [35]. Fig. 2 shows the R-value between the annual electrical output and annual radioactive effluents discharge of the PWRs and PHWRs.

In Fig. 2(a), the correlation coefficient for each radionuclide group shows that the upper limit of $(\mu \pm \sigma)$ is 0.35, and the maximum value is below 0.67, suggesting a weak correlation between the discharge and electrical output [35]. Furthermore, among seven radionuclide groups, the average value of R for four of the radionuclide groups was negative, indicating no correlation between these four groups. In Fig. 2(b), the correlation shows that the upper limit of $(\mu \pm \sigma)$ is 0.36, or higher in T, F, and H nuclides group, but the maximum value is below 0.67; hence, it can be expected that there is a moderate correlation between the discharge and electrical output compared with the PWRs [35]. However, it was found that the R-value of the C nuclides group was negative in all NPPs, and it showed a negative value at a rate of 33% in the F nuclides group. Furthermore, there was no correlation between the discharge and electrical output. In other words, the correlation between the radioactive effluents discharge for each radionuclide group and the electrical output was low in all Korean NPPs.

The low correlation is in line with the fact that radioactive effluents is discharged from PWRs after permanent shutdown [26]. For instance, after the permanent shutdown, the liquid and gaseous tritium are released at a level similar to that of the operating period. Moreover, although the K09 NPP has been permanently closed since 2017, I, T, C, and H nuclides group were discharged in 2018, and T, C, and H were released in 2019 [24,36]. Compared with the operating period, the average discharge of I, C, and H was reduced, but T doubled, showing an inversely proportional relationship with the electrical output. The low correlation between the electrical output and the radioactive effluents discharge can be explained by the effect of the moderator and coolant already present in the system and the storage water of the spent fuel pool on the radioactive effluents discharge. To explain this correlation, modeling was performed using Eq. (10). It can be divided into two terms that is newly generated during the power operation and that is present in the existing systems:

$$A_i = (f_{N,1} + f_{E,1}) \cdot T \cdot P \cdot \delta_i + (f_{N,2} + f_{E,2}) \cdot V_i \quad (10)$$

where A_i is the annual radioactive effluents discharge of radionuclide i (TBq/y), $f_{N,1}$ is the discharge rate of the radioactive effluents newly generated during the power operation in the case of normal operation (1/y), $f_{E,1}$ is the discharge rate of the radioactive effluents newly produced due to the event (1/y), where the event is an unexpected circumstance. T is the power generation time (h), P is the design capacity of the NPP (MWe), δ_i is the discharge of the radioactive effluents generated per electrical output of the radionuclide i (TBq/MWh), $f_{N,2}$ is the discharge rate of the radioactive effluents already present in the normal operation (1/y), $f_{E,2}$ is the discharge rate of the radioactive effluents due to the radioactive material already present (1/y), and V_i includes the discharged amount of radioactive materials already present during the design [26,37]. All factors are interconnected and can have a complicated effect on radioactive effluents discharge.

Fig. 2(b) shows that the correlation between the liquid and gaseous tritium discharge and the electrical output is higher than that of the PWRs. This is because the electrical output was affected by T and P in Eq. (10) and the operating conditions of the NPPs. In addition, because PHWRs use deuterium oxide as the moderator and coolant, more tritium is generated owing to the neutron activation when the nuclear fuel is burned out compared with the PWRs. Hence, there is relatively higher correlation with the electrical output [38].

The above findings can be summarized that the correlation between the radioactive effluents discharge and the electrical output of Korean NPPs could be not so high and varied with various factors including reactor types, designs, and radionuclide groups. It should be noted that Harris et al. derived quite similar conclusion that radioactive releases are dictated not only by electrical generation but also by other factors such as design of radioactive waste treatment systems, etc. [39].

3.1.2. Correlation between the design capacity of the NPPs and the radioactive effluents discharge

As the discharge and electrical output are variable for each year, linear correlation analysis can be conducted using the correlation coefficient (R). However, the design capacity is unchangeable annually for each NPP; hence, its correlation with the

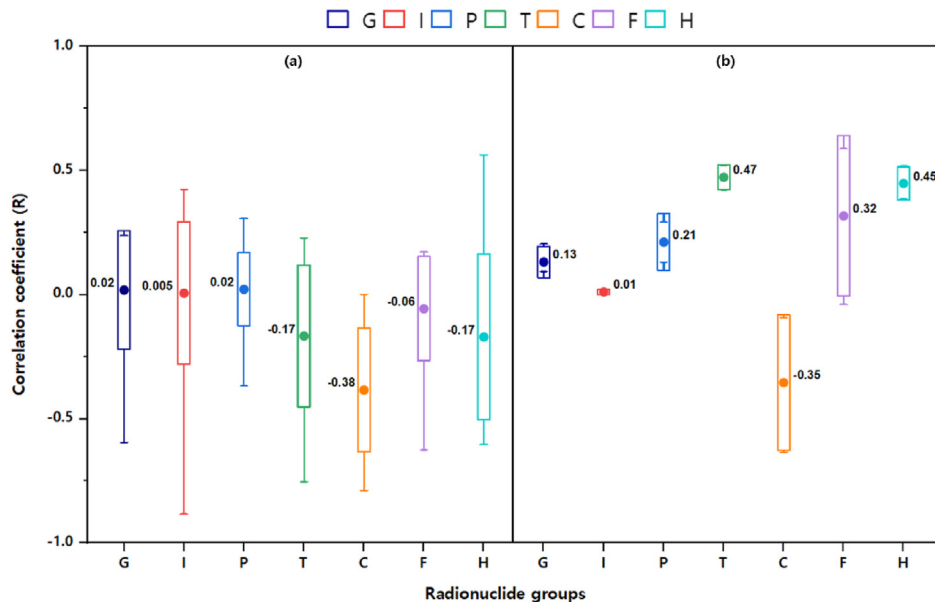


Fig. 2. Correlation coefficient (R) between the annual radioactive effluents discharge of the Korean NPPs and the annual electrical output: (a) is for the PWRs, and (b) is for the PHWRs. \square is $\mu \pm \sigma$, I is minimum-maximum, and \bullet signifies the average.

radioactive effluents discharge cannot be carried out. For this reason, the correlation between the discharge and design capacity was analyzed using the normalized value obtained by dividing the discharge by the design capacity (TBq/y per MWe), a similar approach as the one in which EA used the normalized value (TBq/y per MWth) for the correlation analysis of the discharge and thermal output [32]. Fig. 3 presents the discharged amount of radioactive effluents from the PWRs and PHWRs normalized to the design capacity.

Except for the NPPs with minimum radioactive effluents releases of 0, the range by the radionuclide groups shows a 36–296 times difference in Fig. 3(a) and 18–276 times difference in Fig. 3(b). Therefore, it is expected that there is a low correlation between design capacity and discharge in all NPPs. To analyze the characteristics of each radionuclide group, the coefficient of variation (CV) of the normalized values of each NPP were compared. CV is the value obtained by dividing the standard deviation by the average and determining the dispersion of datasets with different measurement units. A larger CV value indicates a relatively large difference [40]. In I of Fig. 3(a), the maximum CV value is 6.86 (F: 5.69, P: 5.68, G: 2.84, T: 1.1, C: 0.7, H: 0.64, T + H: 0.54), and in the same radionuclide groups of Fig. 3(b), the maximum CV value is 4.7 (P: 3.69, G: 1.43, C: 0.87, H: 0.82, F: 0.81, T + H: 0.553, T: 0.551). This indicates a relatively strong correlation between the design capacity and the radionuclide groups, except for C and H in the PHWRs, compared with the PWRs.

Although there was no significant correlation between the radioactive effluents discharge and the design capacity, as shown in Fig. 3, it was found that the CV value for each NPP was low in T and H, suggesting a higher correlation compared with other radionuclide groups. NRC proposes a fixed value of 0.4 Ci/y per MWth (1.5×10^{-2} TBq/y per MWth), obtained by dividing the sum of liquid and gaseous tritium discharges by the thermal output, to calculate the release of radioactive materials in radioactive effluents from PWRs [37]. The PWRs in Korea show an average of 1.4×10^{-3} – 5.8×10^{-3} TBq/y per MWth, which is 10% lower than the value proposed by NRC. Meanwhile, because the thermal output is calculated by multiplying the design capacity and other factors, its correlation between the radioactive effluents discharge and the design capacity can be analyzed [32]. For example, in the PWR shown in Fig. 3, the CV of the values normalized to the design capacity is 1.1, 0.64, and 0.54 for T, H, and T + H (the sum of the liquid and gaseous tritium), and because the CV is low in the T + H, the correlation with the design capacity is expected to be high compared with T and H. Such results can validate the suggestion of obtaining values by normalizing the tritium to the sum of the liquid and gaseous in the NRC. However, the correlation between the discharge of the liquid and gaseous tritium and the design capacity is relatively higher than that of other radionuclide groups; thus, there is no significant relationship. Therefore, it would be more suitable to reflect the raw discharge than the values normalized to the design capacity or thermal output when setting up the discharge limits.

In Fig. 3 (a), the CV value of ^{14}C is 0.7, the second minimum value after the liquid tritium. It was identified that there is a higher correlation compared with other radionuclide groups except for the liquid and gaseous tritium. In NRC, the discharge of gaseous ^{14}C was set to 7.3 Ci/y (0.27 TBq/y) to calculate the radioactive effluents discharge [37]. It can be expected that this does not reflect the correlation with the thermal output or design capacity, unlike tritium. In the Korean PWRs, the average discharge of gaseous ^{14}C is 0.045–0.13 TBq/y, which is 16% lower than the value presented by NRC. Therefore, in Korean PWRs, the CV value for raw discharge is

lower than the CV value of the discharge normalized to the design capacity, and the same unit as the one proposed by NRC can be used.

The discharge normalized to the sum of liquid and gaseous tritium, and the average discharge of gaseous ^{14}C was higher in the PHWR than in the PWR. In the PHWRs, the discharge normalized to the thermal output is in the range of 2.8×10^{-2} to 4.3×10^{-2} TBq/y per MWth. In contrast, the gaseous ^{14}C has an average discharge in the range of 0.15–0.25 TBq/y. The reason that the gaseous ^{14}C discharge is higher in the PHWR than in the PWR is that the effect of operating conditions on the first term in Eq. (10) is dominant, and deuterium oxide (including ^{17}O) is used as a moderator in PHWRs [41].

Based on the results presented in Section 3.1.1 and Section 3.1.2, it was found that there is no correlation between the discharge characteristics of the radioactive effluents of the Korean NPPs and the annual electrical output or design capacity. Therefore, in this study, raw (non-normalized) annual discharge (TBq/y) was applied to set up the discharge limit.

3.2. Analysis of the radioactive effluents discharge trend of Korean NPPs through the Mann–Kendall trend test

The radioactive effluents discharge trends of Korean PWR and PHWRs were analyzed for the past 20 years. The analysis was carried out on 18 NPPs, for which the discharge data ($n > 10$) could be used in the Mann–Kendall trend test (refer to Table 1). To analyze the annual discharge trend of the radioactive effluents for each NPP, the Z value of Eq. (3) is presented in Fig. 4, (a) presents the Z values from 2000 to 2019 (period A), (b) shows the Z values from 2000 to 2009 (period B), and (c) presents the Z values from 2010 to 2019 (period C). In the 95% confidence interval, the Z value higher than +1.96 means the increasing tendency (region I), a value lower than −1.96 means the decreasing tendency (region III), and that between −1.96 and +1.96 means it is impossible to decide the tendency (region II).

During the period A (2000–2019) as shown in Fig. 4 (a), the NPPs with undecided tendency accounted for 64%. Those with decreasing tendency accounted for 24%, and those with increasing tendency accounted for 12% of the PWRs. In the PHWRs, NPPs with an undecided tendency account for 67%, and those with a decreasing tendency account for 33%. The Z values of G and I were negative in all NPPs, and that of P was negative in all NPPs, except for two plants, suggesting a decreasing tendency. For G, I, and P, the effect of the first term in Eq. (10) is dominant. The correlation with nuclear fission in the nuclear fuel in the operating NPPs and the decreasing tendency is likely attributed to improvement of fuel integrity and waste management system [42,43].

During the period A as shown in Fig. 4(a), the tendency of the ^{14}C discharge can be determined only in the PHWRs. In the period A, there was an increasing tendency of 66.7% (none: 33.3%). Although it was difficult to determine the tendency during the period B (2000–2009) as shown in Fig. 4(b), it showed an increasing tendency of 66.7% during the period C (2010–2019) as shown in Fig. 4(c). In PHWRs, ^{14}C is produced in the moderator, heat transfer cooling system, and nuclear fuel, and only very few remain in the fuel. The remainder was removed by the ion exchange resin of the moderator and the primary heat transfer purification system [41]. Significant amounts of this resin accumulate until the lifetime of nuclear reactor, and hence, the discharge is estimated to increase. It was identified that the ^{14}C newly generated from the fuel in the first term of Eq. (10) and ^{14}C already present and accumulated in the second term of Eq. (10) have a complex effect on the discharge.

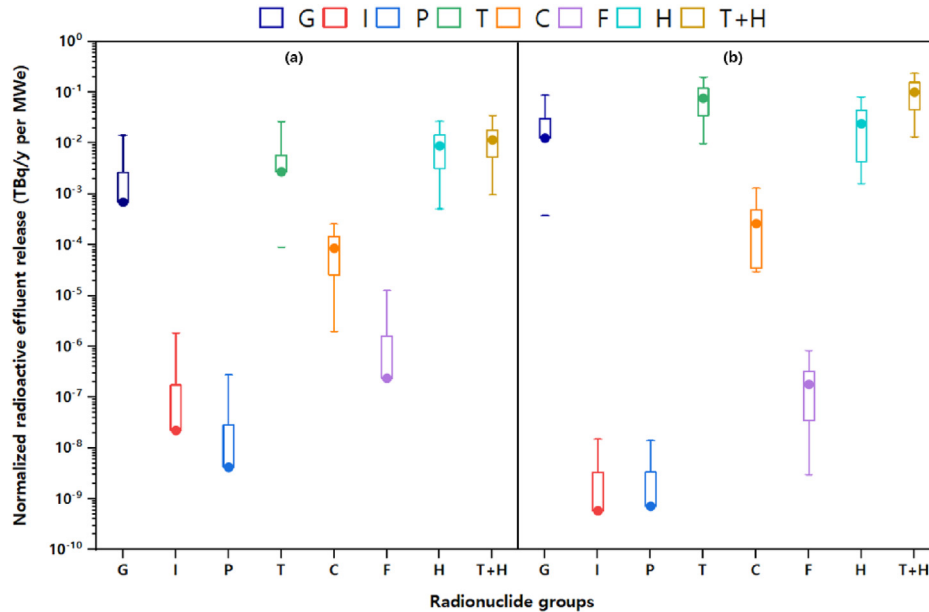


Fig. 3. Radioactive effluents release normalized to the design capacity of each NPP: (a) is for the PWRs, and (b) is for the PHWRs. \square is $\mu \pm \sigma$, I is minimum-maximum, and \bullet is the average. T + H indicates the sum of the liquid and gaseous tritium.

During the period A as shown in Fig. 4 (a), the PWRs show a higher increasing tendency ratio for liquid and gaseous tritium compared with other radionuclide groups. The gaseous tritium has an increasing tendency of 60% (none: 40%), and the liquid tritium has a rising tendency of 26.7% (none: 73.3%). The tritium is generated in the nuclear fuel during the normal operation of the first term of Eq. (10). It is produced and discharged by activating boron and lithium in the stock of tritiated water corresponding to V_i of the second term in Eq. (10), the coolant present in the system [37]. Boron is used as a neutron absorber in the primary coolant; hence, it is expected to increase as tritium is generated through the reaction with neutrons, and the NPPs in operation [37]. With the operation of the NPP, boron is removed to control the generation of tritium, but the remaining boron is reused through the boron recycle system (BRS) [37]. Therefore, with the increase in the concentration of reused boron, the concentration of the gaseous tritium is expected to increase.

During the period B as shown in Fig. 4(b), in the 95% confidence interval, T showed an increasing tendency of 66.7% in the PHWRs K20 and K21. Still, during the period C as shown in Fig. 4(c), T and H showed a decreasing tendency (33.3% for both, none: 66.7%). In the 90% confidence interval, the increasing tendencies of T and H were both 50% (none: 50%) during the period B, and the decreasing tendencies of T and H were both 66.7% (none: 33.3%) in period C. The reason that the decreasing tendency of the liquid and gaseous tritium discharge is dominant in the PHWRs in period C is probably because the discharge of tritium was reduced by installing the tritium removal facility (TRF) in 2007 [44,45].

3.3. Reasonable setup of the discharge limits considering the operational flexibility

3.3.1. Analysis method for the discharge limits applicability

By utilizing the actual annual discharge data for each radionuclide group of 14 Korean NPPs over the past 20 years, Eq. (5) and (7), the equations for the discharge limits proposed in Section 2.3, is applicable. Furthermore, the setup period of proposed discharge limits was set to 10 years (M1) and 1 year (M2). The applicability of

the two derived methodologies for setting up the discharge limits was investigated. Eq. (11) is the equation to determine if the annual radioactive effluents discharge for each NPP and radionuclide group exceeds the discharge limits:

$$DL_{i,t} < D_{i,t} \quad (11)$$

where $DL_{i,t}$ is the discharge limits of the radionuclide groups i for the past t years (TBq/y), and $D_{i,t}$ represents the actual discharge of radionuclide groups i in year t (TBq/y). Once Eq. (11) holds, the failure rate (unit: %) was estimated, and the failure rate for each NPP and each radionuclide group was analyzed. If the failure rate is 10%, this suggests that the compliance probability is 90%. The compliance probability is based on the actual annual discharge data for a specific NPP and radionuclide groups and can be expressed as (1-failure rate). The change in the failure rate according to the l value in Eq. (5) proposed in this study, along with the m value in Eq. (7), was analyzed. Furthermore, the l and m values for each NPP and each radionuclide group were proposed for application to a specific NPP.

In Fig. 4, based on the decreasing tendency and increasing tendency of the (b) period B (2000–2009) and (c) period C (2010–2019), the satisfaction of the discharge limits can be identified. If there is an increasing tendency in period B and a decreasing tendency in period C, the failure rate according to Eq. (11), decreases. Moreover, if there is a decreasing tendency in period B and an increasing tendency in period C, the probability of the failure rate increases. If there is an increasing or decreasing tendency in both B and C periods, it will be impossible to compare the failure rates. Furthermore, if the average radioactive effluents discharge during period B is lower than the average discharge during period C, regardless of the tendency, the M1 failure rate will be higher than the M2 failure rate. In contrast, the M1 failure rate was lower than the M2 failure rate.

3.3.2. Analysis of the discharge limits failure rate

When the discharge limits are set as that of the EA in England, Eq. (4) assumes a normal or log-normal distribution. In addition, Eq.

(6), proposed by the EPA, takes a log-normal distribution and sets up the discharge limits [9,10,15]. The Korean radioactive effluents discharge for each NPP and radionuclide group showed a 54% log-normal distribution and 16% beta distribution (the remaining 30% shows various probability distributions such as extreme value and logistic) when predicted using the distribution fit function of the Crystal Ball software, indicating that it differs from the normal distribution [46]. Fig. 5 presents the failure rates of the discharge limits derived based on the discharge in 2000–2009 for 14 NPPs and six radionuclide groups (^{14}C is excluded because it is used only in the deduction of the discharge limits of PHWRs) during 2010–2019. The discharge limits were analyzed using two methods: one with a 10-year period (M1) and the other with a 1-

year period (M2).

As shown in Fig. 5(a), the failure rate of M1 is 1–1.6 times higher than that of M2. In the one-sided normal distribution, with 1σ , the failure rate was 15.9%, and when the characteristic value l was 1, the failure rate was 16.3% in M1 and 14.2% in M2. In Fig. 5(b), when the characteristic value m is 1, the failure rate is 24.4% in M1 and 24.6% in M2, indicating that it does not follow the normal distribution in both Fig. 5(a) and (b). The failure rates of the discharge limit for each radionuclide group (G, I, P, and F) in 14 NPPs (11 PWRs and 3 PHWRs) are presented in Fig. 6.

In Fig. 6 (a), (b), (c), and (d), the M1 method resulted in lower failure rates for the G and I nuclides groups compared with the M2 method. Furthermore, the average radioactive effluents discharge of the period C (2010–2019) was lower than that of the period B (2000–2009) in all 14 NPPs for the G nuclides group and nine NPPs for the I nuclides group, indicating that the M1 failure rate is lower. Based on these results, it can be estimated that if the setup period is 1 year, the compliance probability is relatively more vulnerable to the discharge change trend compared with the 10-year period. Moreover, because the I nuclides group was not discharged for 9–20 years, or its data includes that of the NPPs below lower limit of detection, its failure rate is lower than that of other radionuclide groups.

In the P nuclides group, the failure rate was lower in M2 than in M1. Among the 11 PWRs, in five NPPs, the average radioactive effluents discharge of the period C was 6.6–280 times higher than that of the period B, which allows for the expectation of a lower M2 failure rate than M1. However, among the remaining 11 PWRs, in 6 plants, the average radioactive effluents discharge in the period B was 5–126 times higher than that of the period C. For the PHWRs, in all two NPPs of which the values can be compared, the average discharge of the period B was 1.9–102 times higher than period C, suggesting the M1 failure rate is lower than the M2 failure rate. Although these results do not provide valid evidence on why the M2 failure rate is lower than the M1 failure rate in the P nuclides group, the different discharge characteristics for each NPP can be identified.

For F nuclides group, the failure rate did not show any regularity in the M1 and M2 methods. During the period B as shown in Fig. 4 (b), the NPPs with a decreasing tendency accounted for 44.4% (none: 33.3%, increasing tendency: 22.2%), and during the period C, none accounted for 88.9% (decreasing tendency: 11.1%). Because various discharge limits for each NPP are applied owing to such increasing and decreasing tendencies, it is expected that the failure rate does not show any regularity in the M1 and M2 methods. Fig. 7 shows the failure rate for each radionuclide group (T, C, and H) for the past 10 years in 14 NPPs (11 PWRs and 3 PHWRs).

Fig. 7 (a) and (c) present the failure rates of the liquid and gaseous tritium in the PWRs. As shown in Fig. 7 (a), T and H show a maximum of 56 and 40 times higher failure rates, respectively, compared with other radionuclide groups, as shown in Fig. 6. In addition, in (c), tritium offers a maximum of 7 times higher than that of the other radionuclide groups. The average discharge of T for the period C was 1.01–2.4 times higher compared with the period B in 10 NPPs among 11 PWR plants, and that of H for the period C is 1.1–2.7 times higher compared with the period B in 7 NPPs among 11 PWR plants, indicating higher failure rate compared with other radionuclide groups. In all three PHWRs, the average discharge of the T nuclides group for the period C was lower than that for the period B, which indicates that the failure rate is lower than that of the PWRs. In PWRs, the regularity of M1 and M2 could not be found for the H nuclides group. During the period B shown in Fig. 4 (b), the H nuclides group showed an increasing tendency of 11.1% (none: 77.8%, decreasing tendency: 11.1%), and during the period C, it showed a decreasing tendency of 27.8% (none: 72.2%). Therefore,

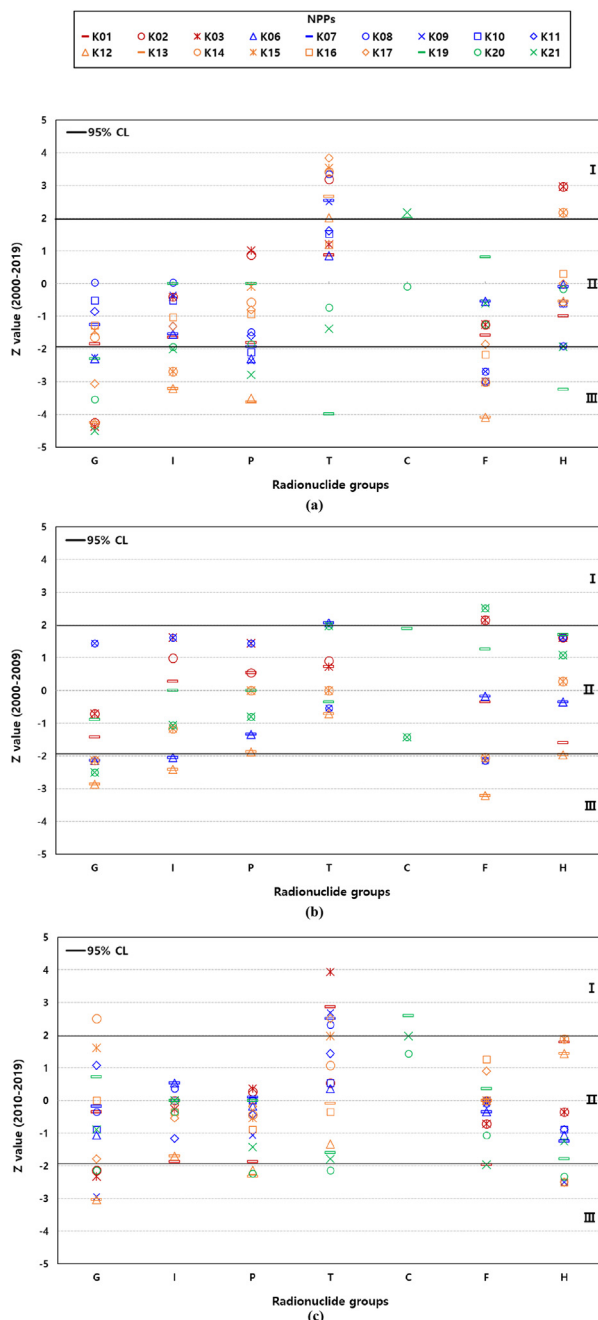


Fig. 4. Z value of the Mann–Kendall trend test for each radionuclide group of Korean NPPs. K01 to K21 signifies the NPPs in legend.

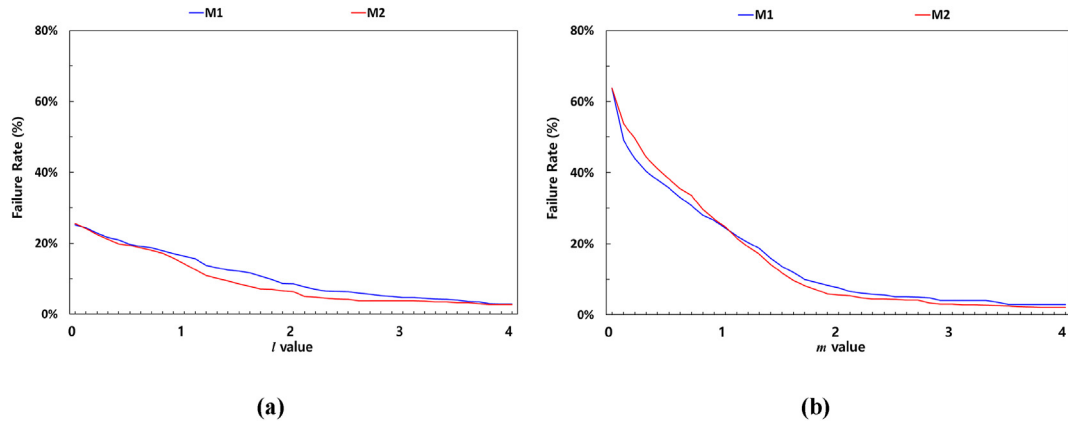


Fig. 5. Failure rates for all NPPs when the discharge limits derived using M1 and M2 are applied: (a) presents the failure rates with different characteristic value l of Eq. (5) and (b) shows the failure rates with different characteristic value m of Eq. (7).

H has more variable increasing and decreasing tendencies than T, and hence, it is difficult to find the regularity of H.

Fig. 7 (b) and (d) present the failure rate of ^{14}C , alongside the liquid and gaseous tritium in three PHWRs. In (b), the failure rate of ^{14}C was approximately 40% higher than that of liquid and gaseous tritium. For C nuclides group, the failure rate did not show any regularity in the M1 and M2 methods.

3.3.3. The characteristic value for each NPP and each radionuclide group presenting a specific compliance probability to the discharge limits

Based on the failure rates for each NPP and each radionuclide group shown in Section 3.3.2, the failure rate decreases with an increase in the characteristic values l and m . For example, if the compliance probability was set at 90% (failure rate of 10%), the corresponding characteristic values l and m were set up, and

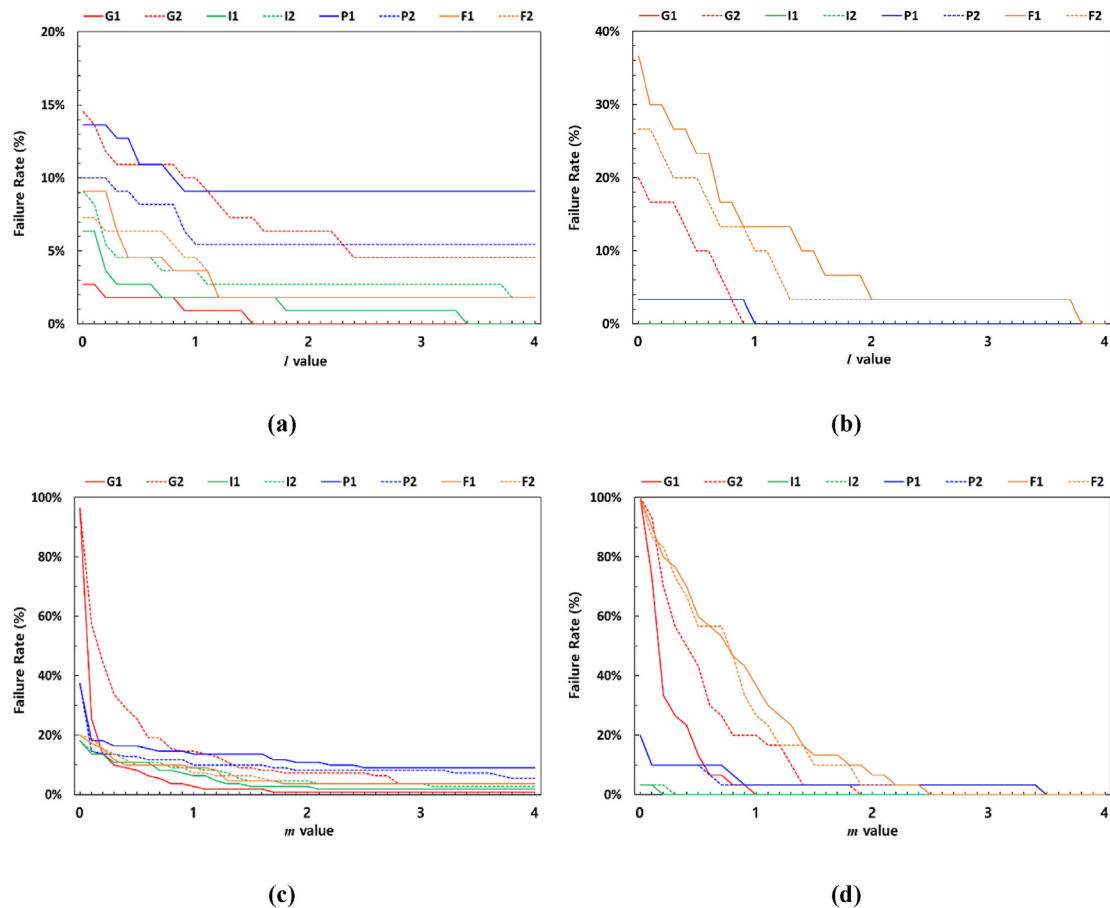


Fig. 6. Failure rates for each radionuclide group (G, I, P, and F) when the discharge limits derived using M1 and M2 are applied: (a) and (b) present the discharge limits of Eq. (5) and the failure rates with different characteristic value l , (c) and (d) present the discharge limits of Eq. (7) and the failure rates with different characteristic value m , (a) and (c) correspond to the PWRs, and (b) and (d) correspond to the PHWRs. The number 1 signifies the method of M1, and the number 2 signifies the method of M2 in legends.

suitable discharge limits were established. Table 2 shows the characteristic value for each NPP and each radionuclide group, corresponding to a specific compliance probability of 90% when the discharge limit is set up using Eq. (5) based on the distribution of the actual annual discharge data of Korean NPPs. By dividing methods into M1 and M2, the minimum characteristic value of l with a compliance probability of over 90% was tabulated for each NPP and radionuclide group.

In Table 2, the compliance probability for most NPPs is 90% when the characteristic value l is below 4. However, some NPPs exceeded 4. Comparing the M1 and M2 methods, the case in which the characteristic value l exceeds 4, resulting in a compliance probability of 90%, accounting for 4.8% in M1 and 1.2% in M2, showing a lower compliance probability in the M1 method. For M1 and M2, the ratio of NPPs, for which the compliance probability falls short of 90% until the l value exceeds 4, is high in the P nuclides group. The K01 NPP showed a negative value during the period B (2000–2009) and a positive value during the period C (2010–2019) for H, allowing us to expect a low compliance probability. In K02, K14, and K15 NPPs, it was not possible to determine the increasing and decreasing tendencies during both periods B and C for the P nuclides group, indicating a higher l value compared with the other radionuclide groups. In the K15 NPP, the increasing and decreasing tendency of the T nuclides group could not be determined during the period B. In contrast, this radionuclide groups showed an increasing tendency during period C, suggesting a low compliance

probability.

Using ^{14}C , the compliance probability of discharge limit for PHWRs can be calculated. Using the M1 method, in the K19, K20, and K21 NPPs, a 90% compliance probability was achieved at 0.6, 0.7, and 4+, respectively. The M2 method resulted in a compliance probability of 90% at 1, 0.6, and 2.4, respectively. Table 3 lists the characteristic values for each NPP and radionuclide group corresponding to a specific compliance probability of 90% when the discharge limit was set up using Eq. (7) based on the distribution of the actual annual discharge data of past Korean NPPs. By dividing into M1 and M2 methods, the minimum characteristic value of m with a compliance probability of over 90% was tabulated by the NPPs and radionuclide groups.

Comparing the M1 and M2 methods in Table 3, the case in which the m value exceeds 4, resulting in a compliance probability of 90%, accounting for 8.3% in M1 and 2.4% in M2, presents a lower compliance probability in the M1 method. When the compliance probability fell short of 10%, the ratio with the m value of over 4 was higher in the order of H, P, and I in the M1 method. In the M2 method, the G and P nuclides groups showed the same ratio of the m value over 4. Using ^{14}C , the compliance probability of discharge limit in PHWRs can be calculated. Using the M1 method, in K19, K20, and K21 NPPs, a 90% compliance probability was achieved at 1.4, 1.5, and 4+, respectively. The M2 method resulted in a compliance probability of 90% at 1.6, 1.6, and 2.4.

The range of characteristic value l using M1 with the discharge

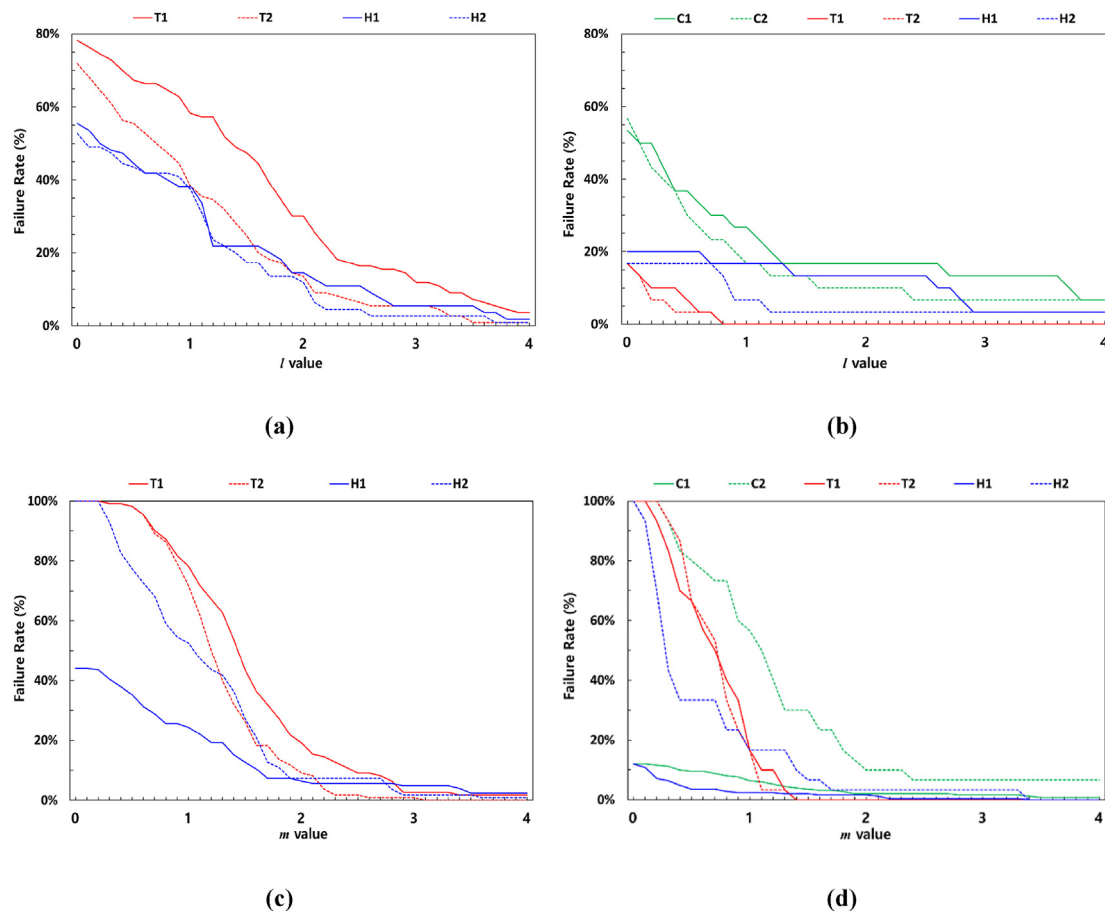


Fig. 7. Failure rates of the liquid and gaseous tritium and ^{14}C when the discharge limits derived using M1 and M2 are applied: (a) and (b) present the discharge limits of Eq. (5) and the failure rates with different characteristic value l , (c) and (d) present the discharge limits of Eq. (7) and the failure rates with different characteristic value m , (a) and (c) correspond to the PWRs, and (b) and (d) correspond to the PHWRs. The number 1 signifies the method of M1, and the number 2 signifies the method of M2 in legends.

Table 2

Characteristic value l for each NPP and each radionuclide group presenting a specific compliance probability of 90% to the discharge limit when the discharge limit was set up using Eq. (5).

NPPs	M1						M2					
	G	I	P	T	F	H	G	I	P	T	F	H
K01	0.2	0	0	0	0.3	4 ⁺ (11.3)	1.1	0	0	1.5	0.9	1.4
K02	–*	–	4 ⁺ ** (18.8)	1.9	–	3.6	–	0	0.9	1.3	–	2.1
K03	–	0	0.9	1	–	3.6	–	0	0	0.9	–	2.1
K06	–	0	0	1.8	0	1.2	0.2	0	0	1.1	0	1.5
K07	–	0	–	2.2	0	1.2	4	0	–	1.4	0	1.4
K08	0	1.8	–	3.9	0.4	1.2	0.1	1.1	–	2.6	0	1.2
K09	–	0	–	3.6	0.4	1.2	–	0	–	3.2	0	1.2
K12	–	–	–	1.9	–	1.1	–	0	–	1.7	–	2
K13	–	–	–	2.1	–	1.1	–	0	–	1.6	–	2
K14	–	–	4 ⁺ (5.2)	2.9	–	1.8	1.3	–	0	2	–	–
K15	–	–	4 ⁺ (86.5)	4 ⁺ (4.5)	–	1.8	2.3	–	4 ⁺ (17.6)	2.4	–	–
K19	–	–	–	–	1.4	–	0.4	–	–	–	–	–
K20	–	–	0	0.6	0.1	2.9	0.5	–	0	0.4	0	1.2
K21	–	–	–	0.1	2	0	0	–	–	0	1	0

* indicates the case in which the compliance probability is 100%, even with an l of less than 0.

** indicates the case in which the compliance probability is under 90% even at 4+. The value in parentheses is the l value at 90% of compliance probability.

Table 3

Characteristic value m for each NPP and each radionuclide group presenting a specific compliance probability of 90% to the discharge limit when the discharge limit was set up using Eq. (7).

NPPs	M1						M2					
	G	I	P	T	F	H	G	I	P	T	F	H
K01	1.1	0.1	0.1	1	1.3	4 ⁺ (4.8)	1.6	0.1	0.1	1.6	1.7	3.6
K02	0.2	0.3	4 ⁺ ** (33.2)	1.9	0	4 ⁺ (4.2)	0.4	0.3	3.3	1.6	0	2.8
K03	0.3	0.9	2.5	1.5	0	4 ⁺ (4.2)	0.4	0.8	1	1.4	0	2.8
K06	0.1	–*	0	1.4	0	1.7	1.1	0	0	1.4	0	1.7
K07	0.1	–	0.3	1.5	0	1.7	4 ⁺ (4.6)	0	1	1.4	0	1.7
K08	1	4 ⁺ (4.1)	0.1	2.9	1.3	1.5	1.2	3.1	0.1	2.2	1	1.5
K09	0.1	0.7	0.1	2.8	1.3	1.5	0.1	0.8	0.1	2.6	1	1.5
K12	0.2	0.3	0.1	1.6	0	1.4	0.3	0.3	0.1	1.5	0	1.7
K13	0.1	0.3	0.1	1.6	0	1.4	0.2	0.3	0.1	1.5	0	1.7
K14	0.1	–	4 ⁺ (9.4)	2.3	–	1.6	1.8	–	0.1	1.9	–	1.6
K15	0.1	–	4 ⁺ (141.5)	2.9	–	1.6	2.6	–	4 ⁺ (29.5)	2.1	–	1.6
K19	0.8	–	–	0.4	2	0.2	1.4	–	–	0.6	1.9	1.3
K20	0.5	0	0.9	1.3	1.1	2.2	1.3	0	0.7	1.1	1	1.7
K21	0.2	–	0.1	1.1	1.8	0.9	0.8	–	0.1	1	1.5	1

* indicates the case in which the compliance probability is 100%, even with an m of less than 0.

** indicates the case in which the compliance probability is under 90% even at 4+. The value in parentheses is the m value at 90% of compliance probability.

limit of Eq. (5) averages at 0.7 to 1.5, except for 4+ NPPs, and that using M2 averages from 0.3 to 1.36. Based on the discharge limit in Eq. (7), the M1 method results in 0.6–0.82, and the M2 method results in 0.63–1.52. If the discharge of Korean NPPs follows the normal distribution curve, the characteristic values l and m should be 1.282 when the compliance probability is 90%. However, the discharge characteristics of the Korean NPPs did not follow a specific statistical distribution. Therefore, both methods of Eq. (5) and (7) can be applied to set up the discharge limits. However, the superiority of these two methods cannot be determined. This is because the characteristics of the NPPs and the radionuclide groups have a more significant effect on the compliance probability of the discharge limits compared with the impact of the methodologies. When the discharge limit of Eq. (5) is set up, the M2 method rather than the M1 method can increase the compliance probability, and when the discharge limit of Eq. (7) is used, the M1 method, rather than the M2 method, can increase the compliance probability.

Between the two methods considering different discharge limits setup periods, the advantage of M1 is that the discharge limits are updated every 10 years; hence, the NPP officer's issues for setting up the discharge limits are not significant, and the advantage of M2 is that the change in discharge performance of a specific radionuclide groups for the short term can be dynamically reflected.

However, in the case of M2, the limitation that the compliance probability is relatively more sensitive to the discharge change trend was identified. Therefore, the characteristic value set with the past discharge data of Korean NPPs (Tables 2 and 3) can be applied to a specific NPP in the future.

4. Conclusions

In this study, two methodologies for setting up the discharge limits were proposed considering the operational flexibility based on the actual radioactive effluents discharge. The existing studies claimed that the correlation between the electrical output and the radioactive effluents discharge has 'no correlation' or a 'proportional relationship.' However, this study identified no correlation between radioactive effluents discharge and electrical output in Korean NPPs. Furthermore, it was determined that there is no correlation between the discharge and the design capacity of the NPPs, which was not revealed in the existing studies.

This study newly proposed two approaches to set up the discharge limits by directly using the statistics of the actual past discharge data of an NPP, rather than simply assuming a conventional normal or log-normal distribution of the radioactive discharge data which was adopted in existing studies, and deriving the

characteristic value equivalent to the probability of compliance to the discharge limits. Though the applicability of the both new methodologies was shown through case studies for operating Korean NPPs, their relative superiority could not be determined since the characteristics of an NPP and radionuclide groups are more dominant than that of the specific methodology. It was also identified that the discharge limits setup period of 1 year has the advantage that the varying trend of annual discharge data of the corresponding NPP can be quickly reflected compared with the period of 10 years.

In addition, the radioactive effluents discharge trend of 18 Korean NPPs was analyzed using the Mann–Kendall trend test. Based on the results, the fission products, radioactive iodines and particulates in gaseous radioactive effluents showed a decreasing tendency over the past 20 years. This can be explained by improvement of the nuclear fuel integrity and waste management systems, and the effort of operators to minimize radioactive effluents. On the other hand, the gaseous tritium shows an increasing tendency in Korean PWRs, which can be attributed to the much more active recycling of soluble poison (boron) in the BRS in Korean PWRs resulting in enhanced concentration of tritium in the primary coolant and subsequently in the gaseous effluent streams. Both liquid and gaseous tritium showed a decreasing tendency in Korean PHWRs since 2010, which can be ascribed to deployment of the dedicated facility to remove tritium from the primary coolant and moderator.

The new methodologies for setting up the discharge limits developed in this study and the characteristic value for each radionuclide group for Korean NPPs are expected to be effectively utilized for the progressive reduction of the environmental releases of radioactive effluents of the NPPs considering the operational flexibilities from historical discharge data.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

- [1] International Atomic Energy Agency, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, General Safety Requirements No. GSR Part 3, IAEA, Vienna, 2014.
- [2] International Atomic Energy Agency, Regulatory Control of Radioactive Discharges to the Environment, General Safety Guide No. GSG-9, IAEA, Vienna, 2018.
- [3] Environment Agency, Developing Guidance for Setting Limits on Radioactive Discharges to the Environment from Nuclear Licensed Sites, Science Report SC0010034/SR, EA, London, 2005.
- [4] Organisation for Economic Co-operation and Development/Nuclear Energy Agency, Effluent Release Options from Nuclear Installations, OECD/NEA, MA, USA, 2003, ISBN 92-64-02146-9.
- [5] United States Nuclear Regulatory Commission, Domestic Licensing of Production and Utilization Facilities; Numerical Guides for Design Objectives and Limiting Conditions for Operation to Meet the Criterion "As Low as Is Reasonably Achievable" for Radioactive Material in Light-Water-Cooled Nuclear Power Reactor Effluents, Code of Federal Regulations Title 10, Part 50, Appendix I, USNRC, Washington, 2021.
- [6] United States Nuclear Regulatory Commission, Standards for Protection against Radiation; Annual Limits on Intake (ALIs) and Derived Air Concentrations (DACs) of Radionuclides for Occupational Exposure; Effluent Concentrations; Concentrations for Release to Sewerage, Code of Federal Regulations Title 10, Part 20, Appendix B, USNRC, Washington, 2021.
- [7] Nuclear Safety and Security Commission, Standards for Radiation Protection, Etc., 2019. Notice No. 2019-10.
- [8] J.H. Cheong, Calculation of risk-based detection limits for radionuclides in the liquid effluents from Korean nuclear power plants, *J. Nucl. Sci. Technol.* 54 (9) (2017) 957–968.
- [9] Environment Agency, Assessing New Nuclear Power Station Designs Generic Design Assessment of General Nuclear System Limited's UK HPR1000, Consultation Document version 1, EA, London, 2021.
- [10] Hitachi-Ge Nuclear Energy Ltd, UK ABWR Generic Design Assessment Quantification of Discharges and Limits, HE-GD-0004 Revision G, Hitachi, Tokyo, Japan, 2017.
- [11] Canadian Nuclear Safety Commission, Process for Establishing Release Limits and Action Levels at Nuclear Facilities, Discussion Paper DIS-12-02, CNSC, Ottawa, 2012.
- [12] European Commission, Verifications under the Terms of Article 35 of the Euratom Treaty-PAKS Nuclear Power Plant Hungary, Technical Report HU-04/4, EC, Luxembourg, 2005.
- [13] Nuclear Safety and Security Commission, Standard Format and Content of the Discharge Plan of Liquid and Gaseous Radioactive Materials from Nuclear Power Plant and Related Facilities, 2017. Notice No. 2017-4.
- [14] Korea Hydro and Nuclear Power Co. Ltd, Final Safety Analysis Report of Kori Unit 2, KHNP, Gyeongju, 2008.
- [15] Environmental Protection Agency, Understanding and Accounting for Method Variability in Whole Effluent Toxicity Applications under the National Pollutant Discharge Elimination System, EPA 833-R-00-003, EPA, Washington, 2000.
- [16] Y. Zhou, N. Duan, X. Wu, H. Fang, COD discharge limits for urban wastewater treatment plants in China based on statistical methods, *Water* 10 (6) (2018) 777.
- [17] Y. Zhou, J. Lei, Y. Zhang, J. Zhu, Y. Lu, X. Wu, H. Fang, Determining discharge characteristics and limits of heavy metals and metalloids for wastewater treatment plants (WWTPs) in China based on statistical methods, *Water* 10 (9) (2018) 1248.
- [18] Environmental Protection Agency, Development Document for Effluent Limitations Guidelines New Source Performance Standards and Pretreatment Standards for the Organic Chemicals and the Plastics and Synthetic Fibers Point Source Category, ume 1, EPA, Washington, 1987.
- [19] International Atomic Energy Agency, Radioactive discharges databases (RADD), Available online: <https://europa.eu/radd/> (accessed on 8 June 2021).
- [20] United States Nuclear Regulatory Commission, Domestic Licensing of Production and Utilization Facilities; General Design Criteria for Nuclear Power Plants, Code of Federal Regulations Title 10, Part 50, Appendix A, USNRC, Washington, 2021.
- [21] Entergy, Arkansas Nuclear One Unit 1 and Unit 2-2019 Annual Radioactive Effluent Release Report, USNRC, Washington, 2020.
- [22] Korean Institute of Nuclear Safety, Waste System and Environmental Discharges Radiation Source, KINS/RG-N12.01, 2015.
- [23] Korea Hydro and Nuclear Power Co Ltd, APR 1400 Design Control Document Tire 2, Radioactive Waste Management, Revision 3, APR1400-K-X-FS-14002-NP, USNRC, Washington, 2018.
- [24] Korea Hydro and Nuclear Power Co Ltd, Survey of Radiation Environment and Assessment of Radiological Impact on Environment in Vicinity of Nuclear Power Facilities, KHNP, Gyeongju, 2018.
- [25] Korea Hydro and Nuclear Power Co, Ltd, Radiation management yearbook, KHNP, Gyeongju, 2009.
- [26] J.S. Kang, J.H. Cheong, Characteristics of radioactive effluent releases from pressurized water reactors after permanent shutdown, *Energies* 13 (10) (2020) 2436.
- [27] Environmental Protection Agency, Guidance for Data Quality Assessment: Practical Methods for Data Analysis, EPA QA/G-9, EPA, Washington, 2000.
- [28] Canadian Nuclear Safety Commission, Radioactive Release Data from Canadian Nuclear Power Plants 2001-10, INFO-0210 Revision 14, CNSC, Ottawa, 2012.
- [29] Oslo, Paris Commissions, Liquid Discharges from Nuclear Installations in 2016, OSPAR, London, UK, 2018.
- [30] General Nuclear System Limited, Chapter 6 Quantification of Discharges Limits, Pre Construction Environmental Report HPR-GDA-PCER-0006, Revision 1, UK HPR1000 GDA Requesting Party, 2020.
- [31] Environment Agency, Criteria for Setting Limits on the Discharge of Radioactive Waste from Nuclear Sites, Environmental Permitting Regulations (England and Wales), EA, London, UK, 2012.
- [32] Environment Agency, Study of Historical Nuclear Reactor Discharge Data, Better Regulation Science Programme, Science Report SC070015/SR1, EA, London, UK, 2009.
- [33] European Commission, Radioactive Effluents from Nuclear Power Stations and Nuclear Fuel Reprocessing Plants in European Community (1976-1980), EC, Luxembourg, 1983.
- [34] The United Nations Scientific Committee on the Effects of Atomic Radiation, Sources and Effects of Ionizing Radiation, ume 1, UNSCEAR, Vienna, Austria, 2000.
- [35] Richard Taylor, Interpretation of the correlation coefficient: a basic review, *J. Diagn. Med. Sonogr.* 6 (1) (1990) 35–39.
- [36] Nuclear Safety and Security Commission, Regarding Concrete Voids of Containment Buildings of Hanbit Unit 3 and 4, the NSSC Will Verify Structural

- Integrity Objectively and Transparently, Press Releases, NSSC, Seoul, Korea, 2020.
- [37] United States Nuclear Regulatory Commission, Calculation of Releases of Radioactive Materials in Gaseous and Liquid Effluents from Pressurized Water Reactors, NUREG-0017, USNRC, Washington, DC, USA, 2020. Revision 2.
 - [38] T.Y. Kong, S.Y. Kim, Y.J. Lee, J.K. Son, S.J. Maeng, Radioactive effluent release from Korean nuclear power plants and the resulting radiation doses to members of the public, *Nuclear Engineering and Technology* 49 (8) (2017) 1772–1777.
 - [39] J.T. Harris, D.W. Miller, Radiological effluents released by U.S. commercial NPP from 1995–2005, *Health Phys.* 95 (6) (2008) 734–743.
 - [40] European Commission, Word definition, coefficient of variation, Available online, <https://datacollection.jrc.ec.europa.eu/worddef/coefficient-of-variation>. (Accessed 30 June 2021).
 - [41] International Atomic Energy Agency, Management of Waste Containing Tritium and Carbon-14, Technical Reports Series No. 421, IAEA, Vienna, 2004.
 - [42] United States Nuclear Regulatory Commission, Radioactive Effluents from Nuclear Power Plants-Annual Report 2018, NUREG/CR-2907, 24, USNRC, Washington, DC, USA, 2018.
 - [43] J.T. Harris, Radiological releases and environmental monitoring at commercial nuclear power plants, in: T. Pavel (Ed.), *Nuclear Power –Operation, Safety and Environment*, InTech, London, 2011, pp. 237–260.
 - [44] J.U. Cheong, S.W. Kim, H.G. Cheong, J. Lim, S.I. Cheong, J.U. Kim, Comparison of tritium concentration in the environment around PHWR, in: *Korean Radioactive Waste Society Autumn Meeting*, Busan, Korea, October 14–16, 2015.
 - [45] K.M. Song, S.H. Sohn, D.W. Kang, H.S. Chung, Introduction to Wolsong tritium removal facility (WTRF), in: *Transaction of the Korean Nuclear Society Autumn Meeting*, Busan, Korea, October 27–28, 2005.
 - [46] Oracle And/or its Affiliate, Oracle Crystal Ball Software User Manual, Version 11.2.4.