

Monitoring of Tritium Internal Exposure Doses of Heavy-Water Reactor Workers in Third Qinshan Nuclear Power Plant

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Abstract

To analyze the tritium internal exposure dose of workers in the Third Qinshan Nuclear Power Plant over the past 15 years. Urine samples provided by workers are tested directly to analyze the tritium concentrations and estimate internal exposure dose. Since 2004, an average of approximately 1600 workers have been monitored annually, with an average annual monitoring frequency of approximately 11 000. Since 2004, the average annual collective dose of tritium internal exposure was 149.62 person·mSv, accounting for 19.07% of the total annual collective dose. A total of 18 workers' annual individual internal tritium radiation doses exceeded 2 mSv, of which 5 workers' internal tritium radiation doses in a single intake exceeded 2 mSv. The occupational population with the largest total internal tritium radiation doses consists of maintenance personnel, fuel operators, and radiation protection personnel, whose collective doses of internal exposure account for 75.51% of the total collective doses within the plant. Over 15 years of operation, the internal tritium radiation doses of workers in the Third Qinshan Nuclear Power Plant have been strictly controlled within the national regulatory limit and power plant management target, ensuring the health and safety of the workers.

Keywords

Third Qinshan Nuclear Power Plant, heavy-water reactor, tritium, internal exposure dose monitoring

Introduction

The Third Qinshan Nuclear Power Plant (hereinafter referred to as plant) with 2 units put into commercial operation in 2002 and 2003, respectively, is the first and only Canadian Deuterium Uranium (CANDU6) heavy-water reactor nuclear power plant in China. The CANDU6 heavy-water reactor uses high-purity heavy water as a moderator and coolant. Deuterium is activated by neutrons in the reactor core to form tritium, which escapes from the system through leakage, maintenance, scavenging, and so on and then volatilizes in the air in the plant, resulting in internal exposure to plant workers. According to the operating experience of heavy-water reactor nuclear power plants around the world, the contribution of tritium internal exposure dose to the annual radiation dose of workers is approximately 20% to 30% of the total dose.¹

Tritium is a pure β -emitter with a half-life of 12.3 years. The β -rays emits have a maximum energy of 18.59 keV, but the average energy is only 5.68 keV. Based on low ray energy and weak penetrating power, the maximum range of the β -particles emitted by tritium is approximately 6 μm in human tissues,

which is much smaller than the depth of the skin cells under the epidermis (approximately 70 μm), meaning tritium cannot subject the human body to external radiation. Tritium in the moderators and coolants of heavy-water reactor nuclear power plants mainly exists in the form of tritiated water (HTO). The

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physicochemical properties of HTO are basically the same as those of plain water. It can easily volatilize in the air to form HTO vapor, which can be absorbed into the body through inhalation, skin absorption, and ingestion. The tritium is then evenly distributed through the human body by the circulatory system within approximately 2 to 3 hours, meaning all organs in the body are exposed evenly. During this process, 97% of HTO quickly enters the bloodstream and mixes with body water. The biological half-life of this tritium is approximately 10 days. The remaining 3% of the HTO infiltrates organic molecules. The biological half-life of this tritium is approximately 40 days.²

The low energy of tritium β decay limits its toxicity; thus, tritium poses a threat only from internal sources. Despite its low energy, the review showed that the relative biological effectiveness of chronic exposures to the β radiation of tritium is greater than that of γ - and X-rays.³ Furthermore, it has been demonstrated that the beta radiation of tritium is more effectively toxic when combined within the molecule.⁴ Especially, such genetic and cytogenetic impacts, over several generations, have the potential to significantly impact the population, with potential effects manifesting at ecosystem level.

The tritium concentration in the coolant and moderator of the plant will gradually increase with the running time of the units. According to the design, the tritium concentration in the system will be close to the saturation point after 40 years of operation. This article organizes and analyzes 15 years of tritium internal exposure dose monitoring of workers in the plant.

Materials and Methods

Monitoring Objects and Cycles

The plant conducts tritium internal exposure monitoring of all workers (including contractor personnel) based on tritium intake during on-site work. The routine monitoring period is 14 or 30 days depending on a worker's intake risk. During the overhaul of the reactor, based on a significant increase in the risk of tritium internal exposure, the regular monitoring period for all personnel will be shortened to 14 days. Special monitoring and mission monitoring are carried out according to specific needs.

Under normal circumstances, tritium internal exposure dose monitoring is not conducted for visitors and inspectors who only stay at the site for a short period. In special cases, special monitoring or mission monitoring will be carried out for these individuals according to specific needs.⁵

Main Reagents and Instruments

The scintillation solution used in this study was the Ultima GOLD LLT (10 L) solution produced by PerkinElmer. The scintillation counter was a PerkinElmer Tricarb 2900TR (Haiyan County, China).

Measurement Methods

The internal exposure monitoring of tritium mainly involves the analysis of biological samples from workers, typically urine samples. Monitoring methods include direct measurement of biological samples and oxidation distillation.⁶ Based on the large number of monitoring personnel and high frequency of monitoring, the plant measures the tritium concentrations in the urine samples by a direct measurement method. Tritium radiation doses are then estimated based on the results of these measurements.

Urine samples are provided and collected during a specified time period. For mission monitoring, urine samples are typically collected within 2 to 3 hours from the end of the workday. The samples consist of 2 mL of urine and 10 mL of scintillation solution. This mixture is shaken thoroughly and darkened for 30 minutes. A liquid scintillation counter is then used to measure the tritium content in the samples. The measurement mode of the liquid scintillation counter is set to decay per minute (DPM) and the ranges of the 3 windows of A, B, and C are 0 to 12, 18.6 to 156, and 156 to 2000 keV, respectively. The color correction, static control, and phase monitoring switches are all turned on. The measurement time is 3 minutes.

Estimation Method for Tritium Internal Exposure Dose

In the nuclear power plant, workers enter the radiation controlled areas frequently, which means worker may intake tritium several times in the same monitoring period. Therefore, simply taking the sum of 50-year committed doses of each tritium intake as the effective tritium internal exposure dose of the workers will lead to a significant overestimation of the actual effective dose. Additionally, workers may enter the radiation control area every day. Some workers even enter the radiation control area several times in a day. Using the method described above, each time a worker entered the radiation control area, a urine sample would have to be collected. This is not feasible in practice. Therefore, for routine monitoring, the tritium internal exposure dose of the workers in a heavy-water reactor nuclear power plant is estimated by adding segmented areas and adding the 50-year committed dose of the final sample.⁷

During the entire monitoring period (typically 2 weeks), if the tritium concentration in the urine detected on day t_i of the monitoring period is C_i (Bq/mL) and the concentration detected on day t_{i+1} is C_{i+1} (Bq/mL), then the effective dose (E) during the monitoring period can be estimated at a 95% confidence level using the following formula⁸:

$$E = \frac{(C_i + C_{i+1}) \cdot (t_{i+1} - t_i)}{2} \quad (D1)$$

where E is the effective dose (mSv), t_i and t_{i+1} are the dates of the 2 sample collections (days), and C_i and C_{i+1} are the tritium concentrations monitored in the 2 samples (Bq/mL).

The committed effective dose E (mSv) from cumulative exposure during a certain period after the last measurement can

Table 1. Annual Number of Workers at the Third Qinshan Nuclear Power Plant Under Tritium Internal Exposure Dose Monitoring From 2003 to 2017.

	Year														
	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017
Number of workers	550	1320	1818	1752	1593	1673	1808	1675	1717	1631	1371	1505	1585	1753	1742

be estimated using a simple estimation method. With this method, the committed effective dose can be derived from the parametric result of the last urine sample measurement C_n and default half-life when continuous deposition occurs. In the absence of other evidence, the half-life is assumed to be 10 days. The formula for estimation is written as follows⁸:

$$E = \frac{4.8 \times 10^{-5} C_n}{\ln 2/10} \quad (\text{D2})$$

where E is the effective dose (mSv) and C_n is the last urine sample measurement (Bq/mL).

During the working period, when workers are at risk of tritium intake, the effective dose of tritium internal exposure for each monitoring period is defined as D1. When workers are no longer engaged in work involving tritium exposure, the effective dose of tritium internal exposure is defined as the sum of D1 and D2.

Results

Number of Monitored Workers

Plant started the personnel dosimetry in 2002, but unit 1 was put into commercial operation at the end of 2002 and unit 2 was put into commercial operation in July 2003. Therefore, yearly monitoring data for both units only became available in 2004. This situation is reflected in the changes in the number of workers that were monitored each year. The number of workers monitored has been relatively stable since 2004, with an average of approximately 1600 workers per year, accounting for 74.88% of the total number of individuals monitored over the years considered in this study. The total number of measurements of tritium internal exposure is approximately 11 000 per year. Specific information is listed in Table 1.

Subject Population Demographics

In the year of 2017, there are 1742 workers who are involved into the risk of tritium internal exposure and monitored the tritium internal dose, including 1635 males and 107 females, listed in Table 2. The average age of the workers is 37.6, and the age range is from 22 to 63. The average number of years working at the plant is 10.3.

Annual Collective Dose of Tritium Internal Exposure and Its Proportion of the Total Collective Dose

Since 2004, the average annual collective dose of tritium internal exposure has been 149.62 person-mSv, accounting for

Table 2. Number of Workers on Age Range in 2017.

Age Range	20-30	31-40	41-50	51-60	>60
Number of workers	237	615	578	249	63

19.07% of the total annual collective dose. The collective dose of tritium internal exposure is closely related to the tritium concentration in the moderator and coolant, operating conditions of units, and tritium-related workload. At the initial stage of unit operation, the tritium concentrations in the moderator and coolant were relatively low, the collective dose of tritium internal exposure were low. As running time increases, the collective dose of tritium internal exposure grows with the tritium concentrations in the moderator and coolant. Since 2008, radiation protection technical and management measures in plant have been increasingly optimized to protect against tritium internal exposure. The trend of a continuous increase in tritium internal exposure collective dose has been controlled and is currently stable. Since the second half of 2012, plant started a special ALARA project to reduce the collective dose, focusing on the high radiation risk activities, encouraging the frontline workers' involvement to optimize the work process, to develop the more effective protection measures internal exposure, to implement timely detection and response procedures for heavy-water leakage. Based on these policies, the collective dose of tritium internal exposure has decreased each year. Specific results are presented in Figure 1. Based on abnormal operating conditions during the overhaul of units and an increase in workload in 2016, the annual collective dose of tritium internal exposure in this year reached 260.97 person-mSv, which is the highest value on record.

Maximum and Average Personal Tritium Internal Exposure Doses

Since 2003, the plant has not experienced any overexposure event in which personal exposure dose has exceeded the national regulation limit. In 2011, the maximum personal tritium internal exposure dose was 14.63 mSv, which is the highest value on record. This is because in February of that year, a welder was accidentally exposed to tritium during the cutting and welding of pipes in the moderator system. The plant promptly initiated a tritium-related emergency plan and sent the welder to a special hospital for medical decorporation. Following medical decorporation, the effective dose of tritium internal exposure that the welder received was determined to be 14.53 mSv. When the dose accumulated throughout the year was included, the total dose was 14.63 mSv, which is lower than the

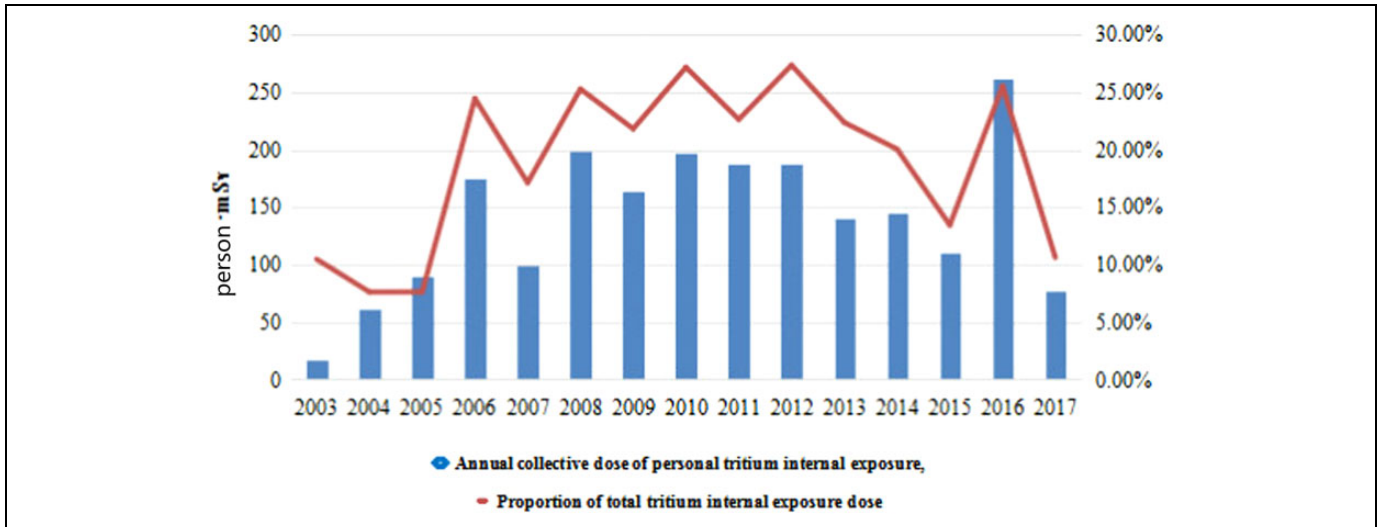


Figure 1. Annual collective dose of tritium internal exposure at Third Qinshan Nuclear Power Plant and its proportion of the total tritium internal exposure dose from 2003 to 2017.

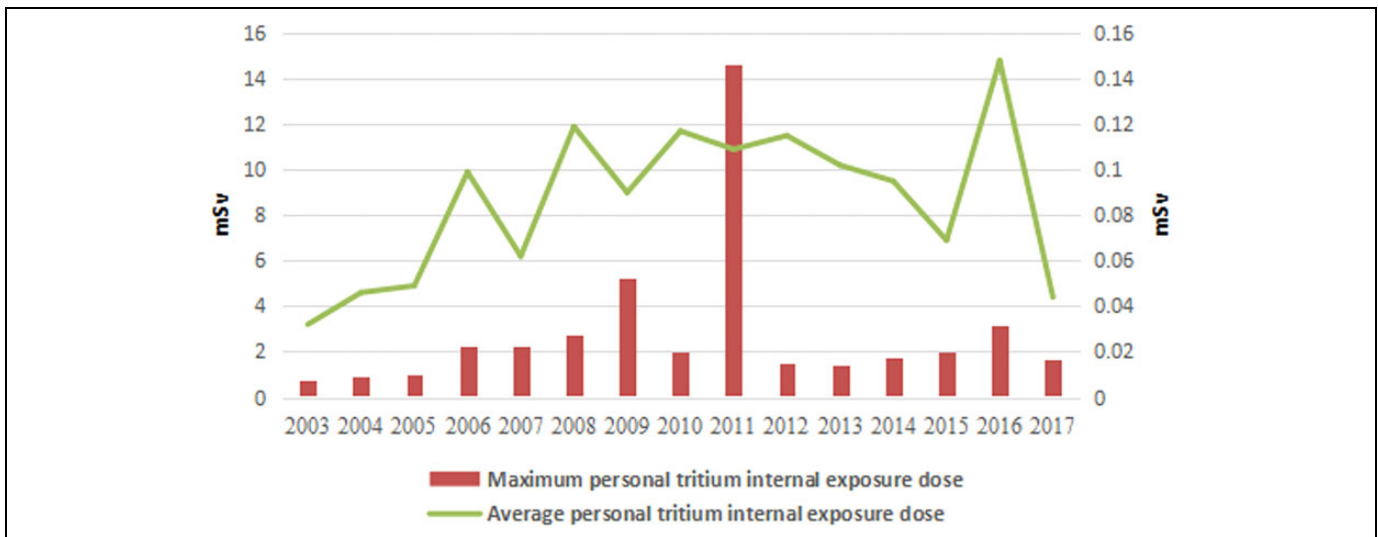


Figure 2. Maximum and average personal tritium internal exposure doses at Third Qinshan Nuclear Power Plant from 2003 to 2017.

national limit and the management target value of the plant. In the other years, the maximum personal tritium internal exposure doses were all less than 6 mSv (for details, see Figure 2).

Since 2003, the annual average personal internal exposure dose has been less than 0.16 mSv. Eighteen workers' annual personal internal exposure doses exceed 2 mSv, of which 5 workers' internal exposure doses in a single intake exceeded 2 mSv.

Statistics of Personnel Tritium Internal Exposure

Since 2003, the total number of the monitored workers is 23 493. The total number of workers with annual tritium internal exposure doses less than 0.10 mSv is 18 036, accounting for 76.77% of all workers. From the 15 years considered in this study, the total number of workers with annual tritium internal

exposure doses over 1 mSv is 185, accounting for 0.79% of all workers. The total number of workers whose annual tritium internal doses over 5 mSv is 2. Only 1 worker experienced a tritium internal exposure dose over 10 mSv with a dose of 14.63 mSv (for details, see Figure 3).

The workers who were monitored in 2011, 2016, and 2017 were normally distributed. Therefore, the statistics from these years are compared in Table 3. One can see that workers were distributed across all dose intervals in 2011 based on the occurrence of a tritium accident in that year. In this accident, a welder was exposed to a tritium internal exposure dose of 14.53 mSv. Another worker in the team was also exposed to tritium accidentally, with an annual internal tritium radiation dose of 3.26 mSv.

In 2016, the collective dose of tritium internal irradiation broke the previous record. Based on the distribution of

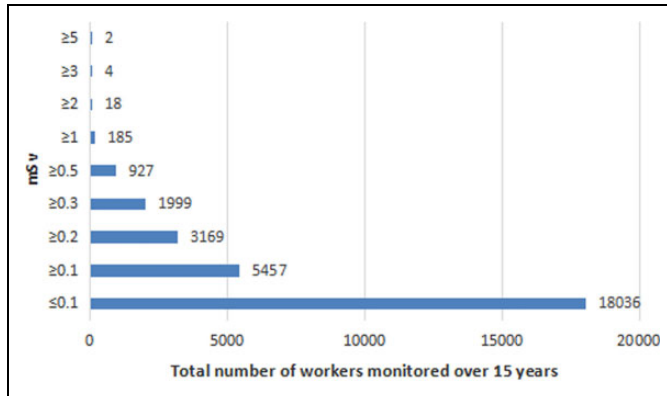


Figure 3. Total number of workers at Third Qinshan Nuclear Power Plant under tritium internal exposure monitoring from 2003 to 2017 (by dose).

Table 3. Distributions of Monitored Workers at Third Qinshan Nuclear Power Plant in 2011, 2016, and 2017 (by Dose).

Year	≤ 0.1	≥ 0.1	≥ 0.2	≥ 0.3	≥ 0.5	≥ 1	≥ 2	≥ 3	≥ 5
2017	1511	231	126	65	29	5	0	0	0
2016	1204	549	413	308	158	48	3	1	0
2011	1257	460	267	166	75	14	3	2	1

monitored workers, one can see that a meaningful number of workers are in the high-dose intervals. Notably, 48 workers experienced an annual internal exposure dose greater than 1 mSv, which is the highest value recorded for any year. In 2017, the collective dose of tritium internal exposure was the lowest since 2005. The workers are largely distributed in the low-dose intervals, with only 5 workers having doses over 1 mSv.

Statistics of Tritium Internal Exposure by Occupation

Unlike pressurized-water reactor nuclear power plants, heavy-water reactor nuclear power plants are designed to allow workers to enter most areas of reactor buildings during normal operation and to remain in most areas for extended periods.¹ During the overhaul of reactor units, open operation of the system will increase. The concentration of tritium in the air will increase sharply in certain areas, potentially in the entire reactor buildings, meaning workers in reactor buildings are exposed to a higher risk of tritium internal exposure. Maintenance personnel, fuel operators, radiation protection personnel, operating personnel, chemists, and technicians are the main groups exposed to occupational radiation in heavy-water reactor nuclear power plants. They are also the main populations with relatively high doses of tritium internal exposure. The monitoring statistics from the past 15 years show that the collective dose of tritium internal exposure of the 6 types of workers listed above accounted for 85.31% of the total dose of all workers in the power plant. The detailed data are listed in Table 4. One can see that the occupational populations with the highest doses of tritium radiation were maintenance

Table 4. Annual Average Tritium Internal Exposure at Third Qinshan Nuclear Power Plant From 2003 to 2017 (by Occupation).

Occupation	Annual Average Number of Monitored Workers	Annual Average Collective Tritium Dose (person-mSv)	Annual Per Capita Tritium Dose (mSv)
Maintenance	852	64.22	0.08
Fuel handling	122	29.37	0.24
Radiation protection	146	19.68	0.14
Operation	246	6.63	0.03
Technology	225	8.07	0.03

personnel, fuel operators, and radiation protection personnel. Their collective dose of tritium internal exposure accounted for 75.51% of the total dose of all workers in the power plant. The occupational populations with the highest per capita doses of tritium internal exposure were fuel operators and radiation protection personnel, whose doses were 0.24 and 0.14 mSv, respectively. The rest of the occupational populations all had doses below 0.10 mSv. As the main subjects under monitoring, maintenance personnel accounted for a significant proportion of tritium dosing. Their collective dose of tritium internal exposure accounted for nearly half of the average annual tritium internal exposure dose of the power plant. The per capita dose of the maintenance personnel was low because they experienced peak radiation during the 2 months of reactor unit overhauling, but experienced small doses of radiation during normal operation. Other occupational populations in the heavy-water reactor nuclear power plant had a lower risk of tritium internal exposure, with annual per capita tritium doses less than 0.03 mSv.

Fuel operators are unique to heavy-water reactor facilities. Because their work focuses on refueling systems, fuel operators have a relatively balanced workload of operation with tritium radiation risk throughout the year. They have the highest risk of tritium radiation among all occupational populations working on the heavy-water reactor. They have relatively balanced and high doses of tritium radiation, meaning they also have the highest per capita dose of tritium internal exposure. Heavy-water reactor nuclear power plants use natural uranium as a fuel. To maintain stable reactivity, refueling operations are required daily. Therefore, in contrast to the refueling personnel of a pressurized-water reactor nuclear power plant, the fuel operation personnel of a heavy-water reactor nuclear power plant occupy permanent positions. Furthermore, based on the complexity of heavy-water refueling systems, fuel operators are responsible not only for refueling but also for maintaining the normal operation of the entire refueling system. Specifically, they handle the technical management, operation, and maintenance of the refueling system. The refueling system is in the reactor core and the coolant of the main heat transfer system of the mobile power station runs through this area, meaning the risk of external and internal exposure is relatively

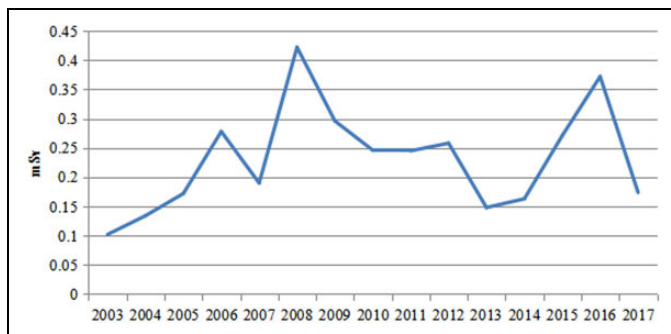


Figure 4. Annual average tritium internal exposure dose of fuel operators at Third Qinshan Nuclear Power Plant from 2003 to 2017.

high. Therefore, fuel operators had the highest per capita doses among all occupational populations at Third Qinshan Nuclear Power Plant, approximately 2.85 mSv total dose and approximately 0.24 mSv tritium internal dose. Detailed data are presented in Figure 4. These data reveal that the changes in the tritium internal exposure doses for fuel operators were largely consistent with the changes in the annual collective tritium internal exposure dose of the power plant. However, there are several differences in certain time periods. For example, the collective tritium internal exposure dose of the power plant decreased continuously in 2014 and 2015. In contrast, the tritium internal exposure doses of the fuel operators increased continuously during this period. This means that local working environments had a significant impact on fuel operator tritium internal exposure dosing, but had little impact on the rest of the workers at the power plant.

Radiation protection personnel are not directly involved in operations with a risk of tritium radiation, but their work requires them to participate in numerous activities that have a risk of tritium radiation. Regardless of whether the reactor units were being overhauled or operating normally, the radiation protection personnel had a relatively balanced workload throughout the year, but their dose of tritium radiation was less than that of the fuel operators. Radiation protection personnel may be exposed to plant-wide radiation because their coworkers and workplaces are distributed across the plant and their work is related to the entire plant. In a heavy-water reactor nuclear power plant, tritium internal exposure is not necessarily the most severe for radiation protection personnel, but they are very likely to be exposed. Specific results are presented in Figure 5. One can see that in general, the changes in the tritium internal exposure dosing for radiation protection personnel coincided with the changes in the annual collective tritium internal exposure dose of the power plant over the study period. This also indicates that the work of radiation protection personnel covers the entire plant.

Discussion

There is natural tritium in environment. Tritium is also produced as a by-product of the operation of nuclear reactors such

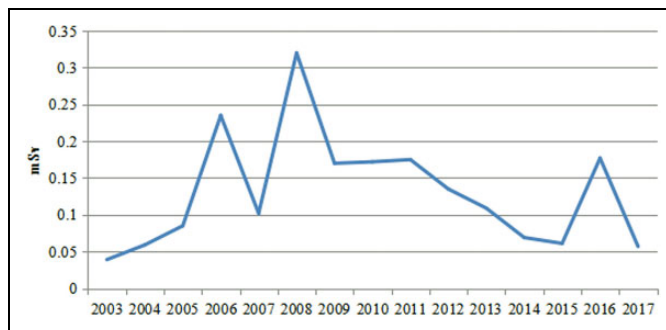


Figure 5. Trend of the annual average tritium internal exposure of radiation protection personnel at Third Qinshan Nuclear Power Plant from 2003 to 2017.

as various nuclear power plants.^{9,10} Tritium is mainly released in the form of HTO vapor from the nuclear power plants. Once released into the atmosphere, HTO can be incorporated into plants or soil moisture. A large number of papers have reported the tritium discharge of the nuclear power plants and its distribution in the surrounding environment.^{11,12} However, occupational exposure to tritium in nuclear power workers is rarely reported, occasionally seen in some internal annual report.

Since the 2 heavy-water reactor units have been put into commercial operation, the plant has fully absorbed knowledge from foreign heavy-water reactor nuclear power plants regarding radiation protection, and continuously explored and improved technical and management measures for protection against tritium internal exposure.¹³ The plant has also continuously optimized radiation protection and strengthened tritium internal exposure protection and dose reduction measures. It has achieved good results in terms of protection against tritium internal exposure, showing the lowest value of collective tritium internal exposure dosing and lowest proportion of tritium internal exposure out of collective dosing among the world's heavy-water reactor nuclear power (Figure 6).

Over the past 15 years of commercial operation, the tritium internal exposure protection and monitoring of workers has been effective, and the tritium internal exposure doses of workers have been strictly controlled within the limits of the state and plant. Although a tritium accident occurred in 2011, the plant responded and took proper emergency measures promptly and helped the victim remove the excess tritium medically, meaning the effective dose of the victim was effectively controlled. Complete and detailed records are kept during accident handling, which not only helps the power plant improve and optimize radiation protection to avoid future accidents but also provides a guide for other domestic tritium-related industries to handle similar incidents.

Querfeld et al¹⁴ reported that 10 surface water samples (collected on April 10, 2011) have been screened for their radionuclide content (³H, ⁹⁰Sr, ¹²⁹I, ¹³⁴Cs, and ¹³⁷Cs). The highest levels, 184 ± 2 Bq/L, ever reported in scientific literature after Fukushima were found in a puddle water sample from close to the Fukushima Daiichi Nuclear Power Plant (FNPP1). Matsu-moto et al¹⁵ measured the tritium concentrations in Japanese

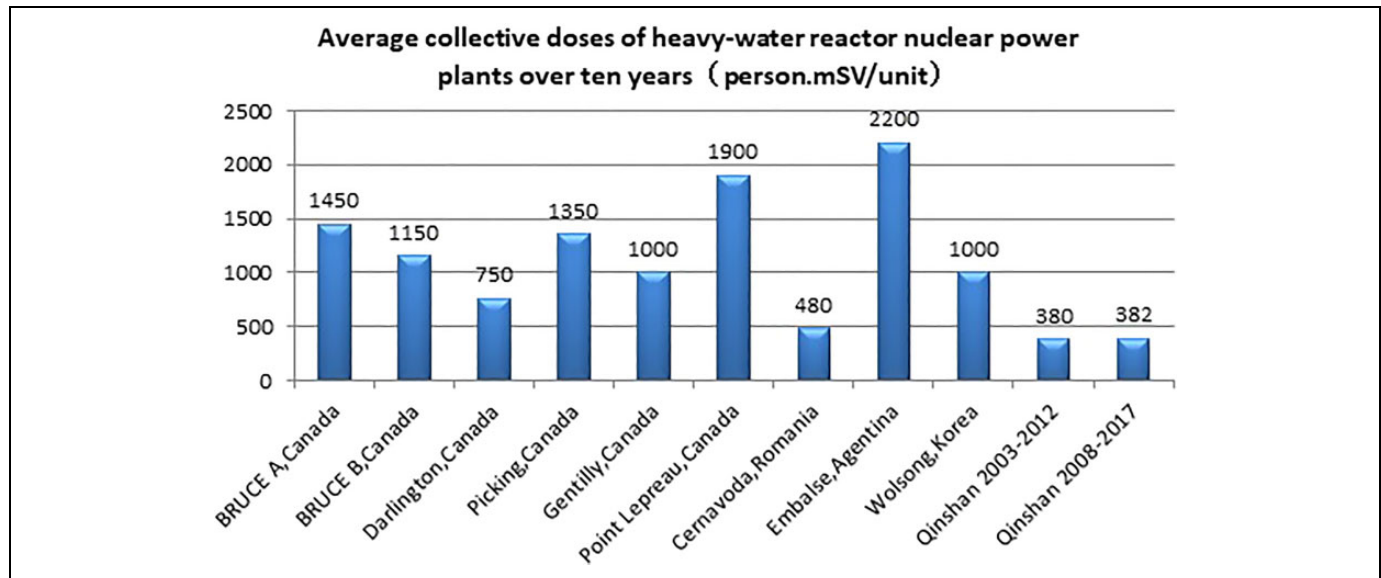


Figure 6. Comparison of the average collective doses of single units in domestic and foreign heavy-water reactor nuclear power plants over 10 years.¹³

precipitation samples collected after the March 2011 accident at the FNPP1. High levels exceeding the preaccident background were detected at 3 localities (Tsukuba, Kashiwa, and Hongo) southwest of the FNPP1 at distances varying between 170 and 220 km from the source. Although Chinese researcher detected the content of artificial radionuclides in snow water in a certain area of northwest China after the Fukushima nuclear accident, the results showed that the artificial radionuclide ^{131}I was contained in the snow water, but the contents of ^{137}Cs , ^{134}Cs , and tritium were not abnormal.¹⁶ The increasing maximum individual dose of 2011 is caused by an unplanned tritium intake event occurred in February 2011, earlier than Fukushima accident which happened in March. These data showed that the nuclear disaster in Fukushima Nuclear Plant in 2011 may have little impact on increasing dose of the Qinshan workers in the year of 2011.

With an increase in running time, the concentration of tritium in the moderator and coolant will increase and the challenges related to tritium internal exposure protection will become more difficult. The plant should carefully summarize operating experience, continuously improve the performance of tritium internal exposure protection, and strive to improve the radiation safety and health of workers. The real-time tritium-in-air monitoring system had been installed and operated to detect the tritium leakage and abnormal increase; the new tritium proof personal protective equipment had been developed to protect the workers from the risk of tritium intake. However, the technique to remove the tritium in the coolant and moderator, which will be put into operation in the future, will significantly decrease the risk of tritium internal exposure.

Authors' Note

Kongzhao Wang, Fengmei Cui, and Yulong Liu conceived and designed the experiment. Lei Sun, Kouhong Xiong, and Weibo Chen

performed the experiments. Youyou Wang and Huahui Bian analyzed the data. Kongzhao Wang and Fengmei Cui wrote the article.


Declaration of Conflicting Interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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