We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists



186,000

200M



Our authors are among the

TOP 1% most cited scientists





WEB OF SCIENCE

Selection of our books indexed in the Book Citation Index in Web of Science™ Core Collection (BKCI)

Interested in publishing with us? Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected. For more information visit www.intechopen.com



Radiological Releases and Environmental Monitoring at Commercial Nuclear Power Plants

Jason T. Harris Idaho State University United States of America

1. Introduction

The generation of electricity from nuclear power has become increasingly important due to the growing concerns of global climate change. Nuclear energy has long been recognized as a leading energy source that produces minimal pollution to the environment that can contribute to this phenomenon. In addition, nuclear power offers an attractive option for countries looking for energy source diversification. Currently there are 442 commercial nuclear power reactors operating in the world (International Atomic Energy Agency [IAEA], 2010, 2011). These power plants contribute about 19% of the electricity production today. The United States of America (U.S.) has the largest commercial nuclear reactor fleet in the world with 104 operating reactors (U.S. Nuclear Regulatory Commission [USNRC], 2010). Of these reactors, 69 are pressurized water reactors (PWRs) and 35 are boiling water reactors (BWRs), located on 65 sites around the country. These power plants contribute about 20% of the U.S. electricity production.

Although it is known that commercial nuclear power plants release small amounts of radioactivity into the environment, there is still the potential for these releases to impact public health. This is especially important today as changes are occurring in nuclear power plant operations including: higher electric generating capacities, increased power levels due to mechanical uprates, and plant life extensions. Public health effects must be reexamined as new light water reactor designs are being considered for construction. In addition, recent events at multiple nuclear power plants in the U.S. involving unplanned releases, especially tritium (³H), have led to increased scrutiny on monitoring and evaluating releases. Changes in radiation protection recommendations and regulations also warrant further and continued investigations in these matters. Although Harris (2007) and Harris & Miller (2008) have performed numerous studies of nuclear power effluent releases and environmental monitoring, data collection and analysis must continue to be performed for the entire nuclear industry.

This chapter focuses on recent research that has been conducted in the areas of commercial nuclear power radiological releases and environmental monitoring by the author. Although the emphasis will be on studies performed in the United States of America, international comparisons will be made where appropriate.

2. Background

Commercial nuclear power plants release small amounts of radiation into the environment under normal operating conditions. Many of the radioactive isotopes that are released are in the form of gaseous or liquid effluents and solid radioactive waste conditioned by the plant. These releases represent some of the by-products of electrical energy generation (Eisenbud & Gesell, 1997).

Three categories of radioactive by-products are produced during routine operation of a commercial light-water reactor: fission products, neutron activation products, and tritium (Glasstone & Jordan, 1980). Fission products are created as a result of the radioactive decay of the nuclear fuel. Approximately 300 different nuclides are formed in the operating reactor. Most of these nuclides are radioactive. Although there is a large quantity of fission products formed, many have little impact on the radioactive releases to the environment because of their extremely short half-lives (<1 day), small quantities, or biological insignificance. Gaseous fission products important to these releases include: ³H, ⁸⁵Kr, and ¹³³Xe. Iodine, solid at room temperature, is also released as a gaseous effluent due to vaporization. Important dose significant iodine isotopes include: ¹³¹I, ¹³³I, and ¹³⁵I. Other decay daughters of produced fission products may also appear in the gaseous effluents as particulate matter (USNRC, 1976a, 1976b).

Activation products are formed by neutron interactions with oxygen in water and air, with nitrogen and argon in air, and with impurity corrosion elements. Like fission products, many of the neutron activation products produced are insignificant in reactor effluents due to their short half-lives (<1 day) or small quantities. Relevant gaseous activation products include: ¹³N, ¹⁴C, ¹⁶N, and ⁴¹Ar (NCRP, 1985, 1987). Important liquid and solid waste activation products arising from interaction of neutrons with corrosion and erosion elements include: ⁵¹Cr, ⁵⁸Co, ⁶⁰Co, and ⁵⁹Fe (Kahn, 1980; USNRC, 1976a, 1976b).

Tritium (³H or T), is produced as a result of both nuclear fission (ternary fission) and neutron activation of deuterium (²H). Tritium is typically treated separately because it is produced in such large quantities compared to any other effluent nuclide and because it arises from other nuclear reactions. One significant source of tritium is the interaction of high energy neutrons with boron. Boron is used in PWRs for shim control (as boric acid) and BWRs as a burnable poison (Glasstone & Jordan, 1980). Tritium is also formed from the interaction of neutrons with ⁶Li (as lithium hydroxide in water treatment).

Typically, the radioactive emissions from operating nuclear power reactors result in insignificant doses to the general population. In 1988, when 110 nuclear power plants were operating at 70 sites in the United States, the mean collective effective dose commitment from all pathways ranged from a low of 1.1 x 10⁻⁵ person-Sv (0.0011 person-rem) to a high of 0.16 person-Sv (16 person-rem). The collective dose commitment for the 150 million persons living within the 2-80-km annuli was 0.75 person-Sv (75 person-rem) for that year (USNRC 1995). Other studies performed throughout the world have shown similar results for population doses around nuclear power plants (Walmsley et a.l., 1991; Ziqiang et al., 1996; Kim & Han, 1999; Nedveckaite et a.l., 2000; Liu et al., 2003; Quindos Poncela et al., 2003). Harris (2007) performed a study to look at the doses for maximally exposed individuals from all plants. A review of epidemiological studies of cancer in populations near nuclear facilities showed that in all scientific reports analyzing nuclear power plants, a cause and effect relationship between cancer risk and radiation exposure could not be found (Patrick, 1977; Jablon et al., 1990; Shleien et al., 1991; Lopez-Abente et al., 1999).

238

There has been a gradual reduction in both liquid and gaseous emissions from power reactors due to improvements in fuel performance and radioactive waste treatment system technology (Harris, 2002). However, the Electric Power Research Institute (EPRI) reports that although radioactive isotopes captured by these systems reduce effluent quantities, radioactive solid waste volumes increase (2003). Also, with longer operating times and license extensions, the accumulation of spent fuel is becoming more important. Many plants have begun storing spent fuel on-site in independent storage facilities. The ageing of existing nuclear power facilities and the increasing accumulation of radioactive wastes have led to an increased emphasis on solid radioactive waste disposal However, at this time doses to the public have not increased during the handling or transportation of radioactive waste shipments. Worldwide estimates also show that nuclear power will continue to grow and thus remain a source of radioactivity exposure to the public.

2.1 Regulatory criteria of releases

The principles that apply to U.S. nuclear power plant radiological releases include consensus scientific recommendations, governmental regulations (Code of Federal Regulations [CFR]), and specific criteria in each plant's operating license. Dose limits, concepts and models based on scientific agreement about radiation effects are recommended by the International Commission on Radiological Protection (ICRP) and the NCRP. Government radiation protection guidance is developed by the U.S. Environmental Protection Agency (USEPA) and approved by the President to assist federal agencies, such as the U.S. Nuclear Regulatory Commission (USNRC), in developing radiation protection regulations. This guidance is usually in agreement with the ICRP or the NCRP. The regulatory standards developed are then required to be incorporated into each nuclear power plant as radiological effluent technical specifications (RETS) that are to be followed through procedures and programs (Andersen, 1995).

Since the inception of nuclear power, federal radiation protection regulations have been based upon the recommendations of the ICRP. The initial ICRP recommendations, published as ICRP Publication 1 and ICRP Publication 2, provided dose limits, models, and radiation concepts. Subsequent to these initial recommendations, the ICRP issued three major revisions, ICRP Publication 26, ICRP Publication 60, and ICRP Publication 103. These recommendations lowered the annual dose limits for members of the public and revised dose models and concepts. ICRP Publication 26 (1977) recommended an annual dose limit of 5 mSv y⁻¹ (0.5 rem y⁻¹) to critical members of the general population (pregnant women and children). Critical members of the general population are those that are more susceptible to radiation effects. ICRP Publication 60 (1991) lowered recommended annual dose limits further to 1 mSv y⁻¹ (0.1 rem y⁻¹) for members of the general population. ICRP Publication 103 (2007) continues with this dose limit. Another important recommendation in terms of reactor releases is given in ICRP Publication 29. This document provides the Committee's recommendations for evaluating pathways between radioactive materials released into the environment and man (ICRP, 1978).

One important recommendation made by the NCRP, published as NCRP Report 92, is specifically concerned with public radiation exposure resulting from nuclear power (NCRP, 1987). The report outlines dose concepts, risks, and technical information regarding the nuclear fuel cycle. In 2011, NCRP will release another recommendation (Report 169) on effluent and environmental monitoring design. Other organizations, including the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), have had a

tremendous influence on the understanding of radiation concepts. The UNSCEAR reports yearly to the General Assembly and periodically issues (every four to five years) the important publication, Sources and Effects of Ionizing Radiation (2000). Another United Nations organization, the International Atomic Energy Agency (IAEA), also influences the practice of radiation protection and issues radiation related reports.

The updated regulations important to nuclear power plant radiological effluents are found in 40 CFR 190 (USEPA 1977). These regulations include limits on radiation doses received by members of the public off-site of the nuclear power plant. During normal operation, the annual dose to any member of the public shall be limited to: 0.025 mSv y⁻¹ (25 mrem y⁻¹) to the whole (total) body; 0.075 mSv y⁻¹ (75 mrem y⁻¹) to the thyroid; and 0.025 mSv y⁻¹ (25 mrem y⁻¹) to any other organ. The USEPA has also set forth guidelines for the maximum amount of radioactivity released into the environment (e.g. 5 mCi of ¹²⁹I per gigawatt-year of electrical energy produced).

The USNRC issues standards and regulations for radiation protection and nuclear plant operations. Standards for radiation protection are contained in 10 CFR 20 (USNRC, 1991). These standards incorporate the dose concepts and models from the older ICRP Publication 26 and 40 CFR 190. The criteria in 10 CFR 20 regarding dose limitations include: a public dose limit of 1 mSv y⁻¹ (0.1 rem y⁻¹), compliance with USEPA's 40 CFR 190 standards, and a requirement for a licensee survey of radiation levels in unrestricted areas, in controlled areas, and in effluent releases. Appendix B to 10 CFR 20 includes limits on effluent concentrations for radiological releases in air and water. These concentration limits are derived from occupational inhalation and ingestion annual limits on intake (ALIs) adjusted to reflect the dose limits set forth by the standards.

USNRC standards for nuclear power plant operations are contained in 10 CFR 50. These standards include criteria for radiological effluent technical specifications, effluent release design objectives and limits, and notification and reporting for events involving the release of radioactive materials. Technical specifications on effluents from nuclear power plants are listed in 10 CFR 50.36a (USNRC, 1996). The specifications require that the licensee comply with 10 CFR 20; that procedures be established and followed regarding the control of effluents; that a radioactive waste treatment system be installed, maintained, and used; that a report be submitted annually to the USNRC regarding effluent releases and the attributed estimated doses to the public; and that procedures be developed that comply with the principle of achieving radiation levels ALARA. Appendix I to 10 CFR 50 gives numerical guides for design objectives and limiting conditions for operation to meet the ALARA criterion for radiological effluents. Doses to members of the general public from radioactive material in liquid effluents released to unrestricted areas shall be limited to 0.003 mSv y-1 (3 mrem y⁻¹) to the whole (total) body, and 0.010 mSv y⁻¹ (10 mrem y⁻¹) to any other organ. The air dose due to the release of noble gases in gaseous effluents is restricted to 0.010 mGy y-1 (10 mrad y-1) for gamma radiation, and 0.020 mGy y-1 (20 mrad y-1) for beta radiation. The public dose from ¹³¹I, ³H, and all particulate radionuclides with half-lives greater than eight days in gaseous effluents is limited to 0.015 mSv y⁻¹ (15 mrem y⁻¹) to any organ. Standards in 10 CFR 50 also cover notification in the event of an abnormal radiological release.

Criteria for nuclear power plant effluents are contained in the radiological effluent technical specifications (RETS), which are part of the nuclear power plant operating license. The RETS include the Limiting Condition for Operation (LCO). The LCO is a description of the criteria that are to be met, the conditions under which the criteria apply, the actions to be taken if criteria are not met, and surveillance requirements to demonstrate that the criteria have

240

been met. The RETS must also contain a site specific Offsite Dose Calculation Manual (ODCM). The ODCM contains both the methodology and parameters used in calculating offsite doses resulting from radiological effluents and the REMP. The USNRC Regulatory Guide 4.1 outlines the programs for monitoring radioactivity in the environs of nuclear power plants (USNRC, 1975). The RETS and ODCM must be approved by the USNRC as part of the license application and approval process. Radiological effluent technical specifications guidelines are contained in NUREG-0133 (USNRC, 1978). The annual effluent report covers plant operations from the previous calendar year. The report includes a summary of the quantities of radiological effluents and solids discharged by the plant. USNRC Regulatory Guide 1.112 aids nuclear power plants in calculating effluent releases.

2.2 Environmental monitoring

Prior to the issuance of a construction permit or an operating license for a nuclear power station, federal agencies (i.e. USNRC) are required to assess the potential environmental effects of that facility to ensure that issuance of the permit or license will be consistent with the national environmental goals prescribed by the National Environmental Policy Act (NEPA) of 1969 and the Federal Water Pollution Control Act. In order to obtain information needed for this assessment, applicants are required to submit a report on the potential environmental impacts of the station and associated facilities. After the station becomes operational, an annual environmental report must be submitted to ensure continued compliance of the requirements set forth in the facility's license and of the Acts stated previously.

Radiological environmental monitoring programs at nuclear power plants are required in accordance with the Code of Federal Regulations. Development and maintenance of these programs are under the guidance of several federal documents. These radiological environmental monitoring programs are established to monitor the radiological impact of reactor operations on the environment. Objectives of these programs include: identification, measurement and evaluation of existing radionuclides in the environs of the facility and fluctuations in radioactivity levels which may occur; evaluation of the measurements to determine the impact of operations on the local radiation environment; collection of data to refine radiation transport models; verification that radioactive material containment systems are functioning to minimize environmental releases to levels that are as low as reasonably achievable (ALARA) and; demonstration of compliance with regulations. Implicit in these objectives are the requirements to trend and assess radiation exposure rates and radioactivity concentrations in the environment that may contribute to radiation exposures to the public. The results of the REMP are submitted as part of the plant's annual environmental report.

Each plant establishes their own, unique REMP program to reflect site-specific conditions and surrounding population characteristics. The program consists of preoperational and operational components. The preoperational program is conducted in part to measure background levels and their variations in environmental media in the area surrounding the plant. Environmental media include: milk produced from cows or goats, broadleaf vegetation, fish, fruits and vegetables, edible aquatic invertebrates, surface water, drinking water and ground water.

Each plant is to also make changes to its REMP program as conditions change. But, it has been reported recently that many plants are decreasing their programs due to budget constraints and lack of positive radioactivity measurements. This reduction can lead to decreased litigation protection, decreased public confidence, and potential unreported or undetected releases. Reduced REMP programs have led to recent public opinion and regulatory problems for several facilities due to unexpected and/or undetected tritium releases. Decreased lower limits of detection (LLDs) and minimal detectable activities (MDAs) reportedly have led to newly quantifiable low levels of many radionuclides in the environs around nuclear power stations. Changes in operating conditions may also lead to new radionuclide transport pathways being developed, as has been seen with precipitation scavenging and concentration in ice.

Ultimately, a nuclear power plant's REMP program is designed to assess the impact of radiological releases on the environment and the public. Public opinion of the nuclear power industry has traditionally been very troubled, especially with the accidents at Three-Mile Island and Chernobyl. Positive public opinion to nuclear power can only be achieved through truthfulness by the nuclear power company regarding operations and radiological releases and accurate and comprehensive monitoring of these releases.

3. Effluent release study

As commercial nuclear power electrical generation steadily increases in the U.S. and the rest of the world, it has become even more important to evaluate the release of radioactive materials into the environment. An easy way to track industry wide effluent releases is by performing trend analyses. Accumulated data may also be used for analysing reactor power up-rate consequences, protecting the nuclear power industry against litigation, and for assisting in new power plant siting. Most importantly, collecting and maintaining an effluent database is necessary in maintaining a favourable public perception regarding the low environmental and biological impact of nuclear power. This is especially important now as several recent, inadvertent releases of radioactive materials from nuclear power plants have occurred. Because of these circumstances, the author has compiled and analysed the effluent data for all U.S. commercial nuclear power plants since 1995. Presented here is also an update of the comprehensive study performed by Harris & Miller (2008).

The classification and monitoring of liquid and gaseous radiological releases is fairly uniform around the world. The classification is based on the nuclide, chemical or physical form, and dose or activity significance. In the U.S., gaseous effluents are divided into fission and activation gases, iodines, particulates (with half-lives greater than eight days), and tritium. Liquid effluents are divided into fission and activation products, dissolved and entrained gases, tritium, and gross alpha activity. International organizations and other nations use the same categories, but combine the fission and activation products and the dissolved and entrained gases in liquid effluents (UNSCEAR, 2000; Harris, 2002).

The classification of radioactive releases is important because dose calculations are based upon them. For example, the collective effective doses calculated by UNSCEAR (2000) use these effluent categories. The groupings also allow plants and nations to compare and benchmark with another. Unlike the simplified general UNSCEAR model, the USNRC model requires specific nuclide, meteorological, and site specific conditions. Hence, this model provides more accurate estimates of dose.

Other studies have been performed to assess the doses from nuclear power radiological releases. Vold (1984) determined the ratio of the collective effective dose equivalent (CEDE) via a specified ingestion pathway relative to that CEDE by inhalation per annual releases of a radionuclide. Kim & Han (1999) and Liu et al. (2003) assessed the impact of tritium

242

released from nuclear power plants in China. Both of these studies confirmed that the doses were less than 1% of the regulatory limits. Ziqiang et al. (1996) reported similar results not only for tritium, but for other radionuclides as well.

What is very common in the nuclear power industry is trending and benchmarking of data. This is done to improve plant operations and management. Many organizations that oversee different aspects of nuclear power plants use these methods for comparison. These comparisons may be advantageous or detrimental to a plant. For example, with radiological releases, high activities compared to other plants can lead to lower profits due to higher premiums from American Nuclear Insurers (ANI). It can also lead to scrutiny from the Institute of Nuclear Power Operations (INPO) and greater surveillance from USNRC. Thus, these comparisons are very important. Gilbert (1994) identifies statistical analyses suitable for detecting trends in environmental contamination data. Accurate trend analyses can aid plants in these aforementioned areas. Trend analyses were performed for the data over the 15 year period using the Mann-Kendall non-parametric test. Inspections of release trends over the fifteen year period help identify areas of concern with these releases. Future estimates of release radioactivity and public doses can then be made from these analyses.

3.1 Methodology

The data utilized for the effluent release study were taken from the annual radioactive release reports provided by the nuclear power plants to the USNRC as required in their operating license conditions. These reports were either provided directly to the author from the licensee or taken from the USNRC Agency wide Documents Access and Management System (ADAMS). The reports provide categorical effluent release data, nuclide specific radioactivity, and site specific data needed for dose calculations. Population information not provided by the licensees was taken from appropriate census reports (U.S. Census Bureau, 2010).

Data was analysed for those reactors that have operated for the 15 year period of 1994-2009. This length of time is long enough to allow plants to stabilize in the event of long shutdown periods and allows evaluation of plants for at least seven refuelling cycles. Events that may affect releases, such as power-uprates and failed fuel from defects, will also show up in this period. The beginning of this data set also coincides with the cessation of tracking radiological effluents by the U.S. in 1994. In this time frame, 103 reactors were operating. Browns Ferry Unit 3 began operation in 2006, to become the 104th operating reactor.

Effluent radioactivity was obtained from data reported by the nuclear power plants in their annual radioactive material release reports. The effluent data was compiled for all operating PWR and BWR plants from 1995 - 2005. The completeness of the data was 98%. In keeping with U.S. nuclear power effluent report formatting, data was compiled and analysed using the same categories as those listed in USNRC Regulatory Guide 1.21. The four gaseous effluent categories used were: fission and activation gases (F/A), total iodine (¹³¹I), particulate matter or particulates, and tritium. The three liquid effluent categories used were: fission and activation products and dissolved and entrained gases are several orders of magnitude smaller than tritium, those two categories were added together and listed as "F/D". This category replicates the reporting done by UNSCEAR. Gross alpha radioactivity was not included in this study.

Trend analyses were performed for the data over the time period using the Mann-Kendall non-parametric test. This procedure was used since missing values were allowed and the data need not conform to any particular distribution (Gilbert, 1994). Inspection of trends over the time period identifies the overall direction of industry effluent releases and can roughly be used to predict future releases.

For this updated study, one dose assessment methodology was used. The collective effective doses (CED) were calculated for the U.S. population using the UNSCEAR methodology. For these dose calculations, the effluent data was normalized. This was achieved by taking the amount of radionuclides released per unit of electrical energy generated each year. This method is the most common way to normalize effluent data. The electrical energy generated per year was obtained by multiplying the net electrical energy generated by the capacity factor. Capacity factor is defined as the gross electricity generated divided by the product of the licensed capacity and reference time. Normalizing data in this manner takes into account the operational performance of the nuclear power plant. However, it also assumes that effluent release amounts are a direct consequence of operation time. The author cautions against making simplistic comparisons of radioactive releases with the electrical energy generated because of the many factors which affect the amount of radioactive materials released, including the condition of the fuel, primary system integrity, design of effluent and radioactive waste treatment systems, maintenance activities, operations, and equipment performance.

3.2 Results and discussion

3.2.1 Radiological effluent releases

The annual variation of total nuclear power plant radioactivity released in gaseous effluents in PWRs and BWRs are shown in Figs. 1 and 2, respectively. As expected, the activity from PWR releases is higher than that from BWRs due to the greater number of plants. Regardless of this fact, the average tritium release from PWRs is also higher due to chemistry practices that create more tritium in the plant. Nearly every category from both reactor types is fairly level in terms of activity released for the entire time period. The evaluation of the data over the time period partly eliminates variations in annual values. The advantage of using 15 years of data is that operation anomalies, such as long shutdown times for maintenance, are averaged out. The one notable exception appears in the PWR particulate category. In 2003, one plant experienced an annual release over five orders of magnitude above the mean. This single event was significant enough to skew the entire industry release activity, especially since the annual radioactivity released in particulate matter is so low compared to tritium or fission and activation gases. The increase in 2005 was due to higher activity releases by several plants.

The annual variation of total nuclear power plant radioactivity released in liquid effluents in PWRs and BWRs are shown together in Fig. 3. As expected, and for reasons similar to that of the gaseous releases, PWR liquid radioactivity in releases is higher than in BWRs. Liquid releases have stayed very constant over the 11-year period. The most notable exception is the pronounced decline in BWR non-tritium (F/D) radioactivity from 2003 - 2007.

3.2.2 Radiological effluent trends

U.S. industry effluent trends were evaluated using the Mann-Kendall non-parametric test. The Mann-Kendall test was performed as follows: For any given release category, x, its feature vector consists of the release summation from all plants appearing in a given year i. These release activities are ordered from the first year, 1995, to the final year, 2009, that data was gathered.

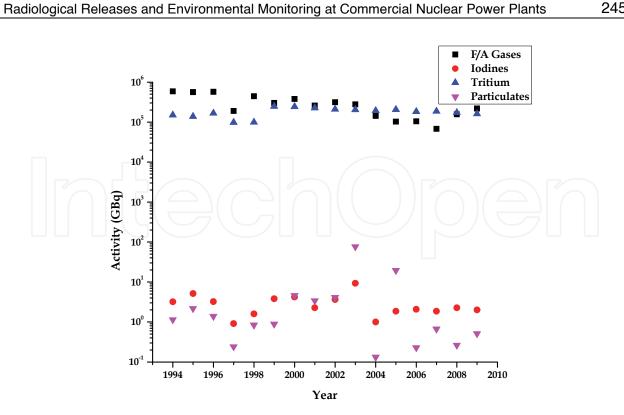


Fig. 1. Variation of total radionuclide activity released in gaseous effluents from PWR plants

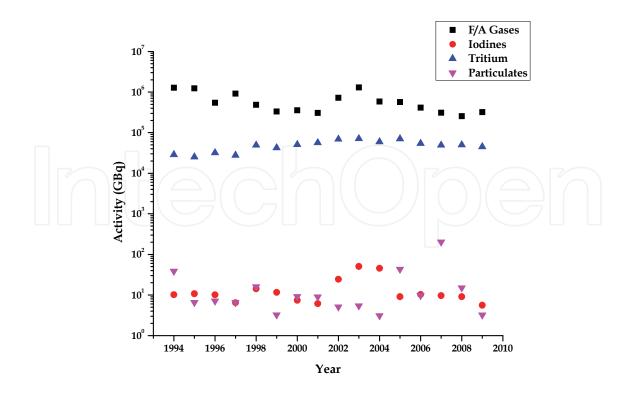


Fig. 2. Variation of total radionuclide activity released in gaseous effluents from BWR plants

www.intechopen.com

245

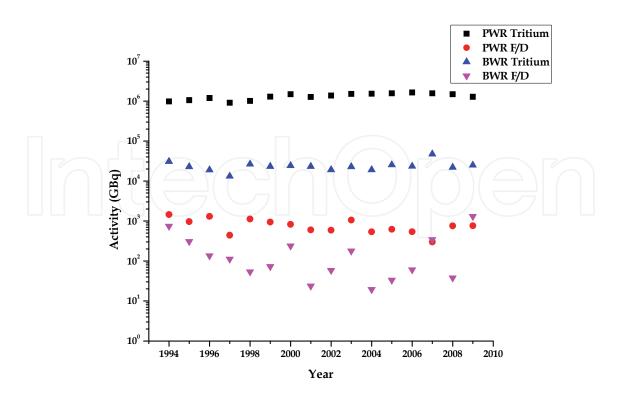


Fig. 3. Variation of total radionuclide activity released in liquid effluents from PWR and BWR plants

The x value for each year is compared to all other years greater than that year, and Kendall's statistic S is calculated as follows:

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^{n} \operatorname{sgn}(x_j - x_k)$$
(1)

where: $sgn(x_j - x_k) = \begin{cases} 1, & if x_j - x_k > 0 \\ 0, & if x_j - x_k = 0 \\ -1, & if x_j - x_k < 0 \end{cases}$

Generally, if a dataset displays a consistently increasing or decreasing trend, S will be positive or negative, respectively, with a larger magnitude indicating the trend is more consistent in its direction. By using the *sgn* function, the algorithm used was able to detect trends featuring either large or small increase steps from year to year equally. The S statistic is then compared to the corresponding P-value (Hollander & Wolfe, 1999).

Under the null hypothesis, H₀, that there is no trend displayed by the time series, the distribution of S is then expected to have a zero-mean and variance. The Mann-Kendall test was performed on BWR plant releases, PWR plant releases, and all plant releases (BWR and PWR combined) at a significance level of 0.05.

The results of the trend test are shown in Table 1. Over the past 15 years, it can be seen that for most effluent categories, the releases are level, meaning there is no increasing or decreasing trend. For these categories, improvements in radioactive waste treatment and

reactor operations are offset by increased power production, increased capacity factors, and power up-rates. Looking at PWRs only, gaseous fission and activation products and liquid F/D have decreased while liquid tritium has increased. Reduction in the fission and activation products over the years is probably a direct result of longer holdup times for radioactive decay. In addition lower fission and activation product activities may be due to improved fuel performance from better manufacturing methods. Also, longer operation times in recent years provide stability to the reactor, leading to less fuel shock and defects. The increasing trend for liquid tritium activity is due to an increase in liquid discharges by these plants. Formerly zero-discharge plants have begun to release liquids again to avoid build-up of their tritium inventory. Coupled with this practice, over the last fifteen years many plants have reduced recycling of boron for reactor control. This procedure contributes to increased tritium production.

| Release Category | S Statistic | Trend | |
|--------------------------|----------------|------------|--|
| PWR GaseousF/A gases | -61 Decreasing | | |
| PWR GaseousIodines | -15 None | | |
| PWR GaseousTritium | -3 | None | |
| PWR GaseousParticulate | -11 | None | |
| PWR LiquidsTritium | 57 | Increasing | |
| PWR Liquids – F/D | -35 | Decreasing | |
| BWR GaseousF/A gases | -41 | Decreasing | |
| BWR GaseousIodines | -13 | None | |
| BWR GaseousTritium | 31 | Increasing | |
| BWR GaseousParticulate | 7 | None | |
| BWR LiquidsTritium | 27 | Increasing | |
| BWR Liquids – F/D | -7 | None | |
| Total GaseousF/A gases | -65 | Decreasing | |
| Total GaseousIodines | -21 | Decreasing | |
| Total GaseousTritium | -3 | None | |
| Total GaseousParticulate | 13 | None | |
| Total LiquidsTritium | 57 | Increasing | |
| Total Liquids – F/D | -13 | None | |

Table 1. Mann-Kendall trend results for U.S. commercial nuclear power plant radiological effluent releases from 1995 – 2009

For BWRs, liquid tritium and gaseous releases have increased and gaseous fission and activation releases have decreased. The increase in gaseous and liquid tritium are relatively new phenomenon and is probably related more to the increased power production and capacity factors over the last several years than anything else. Looking at the entire industry (PWRs and BWRs combined), all effluent releases except gaseous fission and activation products and iodine and liquid tritium are level. Because PWRs make up 66% of the U.S. industry, their releases have a greater impact on the overall release trends, as is evident with the increasing liquid tritium.

3.2.3 Radiological impact of effluent releases

Tracking effluent release quantities is important in determining radioactivity levels in the environment. However, dose determination of the effluents must be performed to estimate the human effects of these radiation sources. The collective effective dose (CED) from radiological effluent releases was obtained using an average collective dose calculation method used by UNSCEAR. UNSCEAR (2000) calculates population dose by first calculating the CED per unit release of radionuclides released from reactors and then normalizes the value with the electrical energy generated. The CED is divided according to type of release (airborne or liquid), radionuclide category (noble gases, tritium, C-14, iodine, and particulate matter), and pathway (immersion, ingestion, inhalation, and external irradiation. The normalized collective effective dose model is given by:

$$D_{CE} = \sum_{i} \frac{A_i}{E} D_i \tag{2}$$

where: A_i =activity of release category i (GBq); D_{CE} = total CED (person-Sv GW⁻¹ y⁻¹); D_i = collective dose for release category i (person-Sv PBq⁻¹); and E =energy produced by the nuclear reactor (GW y⁻¹).

The dose assessment procedures for this model are applied to a model site with representative environmental conditions. The average population density used is 20 km⁻² within 2,000 km of the site. Within 50 km of the site, the population density is taken to be 400 km⁻². These parameters were obtained from previous UNSCEAR assessments and take into account the transport and dilution of released radionuclides from nuclear installations. The parameters used are assumed to not underestimate dose. Using this model site, the collective effective dose per unit release is obtained for the different release categories. Due to its lack of specificity, this model should be used for general comparisons only. The collective effective doses estimated from commercial nuclear power plant radiological effluent releases are very low especially when compared to other man-made sources of radiation. The doses only represent up to a few percent of the regulatory limits (Harris & Miller, 2008).

From the collective effective doses, effective doses were computed for the entire U.S. population to give average annual doses. This was done by taking the CEDs calculated for each release category, gaseous and liquid, and dividing them by the U.S. population for each year (U.S. Census Bureau, 2010). The effective doses were than summed to give a total dose for each person. The results of these effective doses are given in Table 2. For the 15-year period, total effective doses ranged from 5.42×10^{-8} mSv (5.42×10^{-6} mrem) to 1.68×10^{-7} mSv (1.68×10^{-5} mrem). The doses to an average person would be expected to be even lower since many do not live near a nuclear power plant. This is just one of several ways to calculate a very general effective dose for the population.

4. U.S. REMP evaluation

An evaluation of all U.S. nuclear power plant radiological environmental monitoring programs (REMP) was conducted from 1995-2007. An attempt was made to assess the significance of the radionuclides detected in the environment compared to natural and other man-made radiation sources. It is important to note that detected concentrations of radionuclides in the local environment as a result of nuclear power radiological releases

are very low. The percentage of plants sampling different pathways, the percent cumulative exposures, and differences between control and indicator measurements were determined.

4.1 Methods

Inspection and analysis of the industry REMP data was taken from the annual summary tables from each nuclear power plant's annual radiological environmental monitoring report. The summary data includes the following parameters: sample medium, type and number of analyses performed, required (LLD), the mean and range of the positive measured values of the indicator locations, the mean, range, and location of the highest indicator annual mean, the mean and range of the positive measured values of the number of non-routine reports sent to the USNRC. In this evaluation, the highest indicator values were compared to the control values.

The following environmental pathways and sample analyses (in parentheses) were investigated: bottom/shoreline sediment (gamma spectral analysis), fish (gamma spectral analysis), edible aquatic invertebrates (gamma spectral analysis), surface water (gamma spectral analysis, gross beta, and ³H), drinking or potable water (gamma spectral analysis, gross beta, and ³H), ground water (gamma spectral analysis, gross beta, and ³H), air particulate filters (gamma spectral analysis and gross beta), airborne radioiodine (¹³¹I), milk (⁹⁰Sr and ¹³¹I), food products such as fruits and vegetables (gamma spectral analysis), broadleaf vegetation (gamma spectral analysis), soil (gamma spectral analysis), grass (gamma spectral analysis), and direct radiation using thermoluminescent dosimeters (TLDs). In addition, pathways not normally sampled by most plants, like precipitation, were also studied.

For all data, only positive measured values were used in the statistical calculations. During the study period there were over 1.6×10^6 analyses performed on environmental media collected as part of the required monitoring programs. Broken down, this averages to about 1.27×10^5 analyses y⁻¹ and 2.0×10^3 analyses site⁻¹ y⁻¹.

4.2 Results and discussion

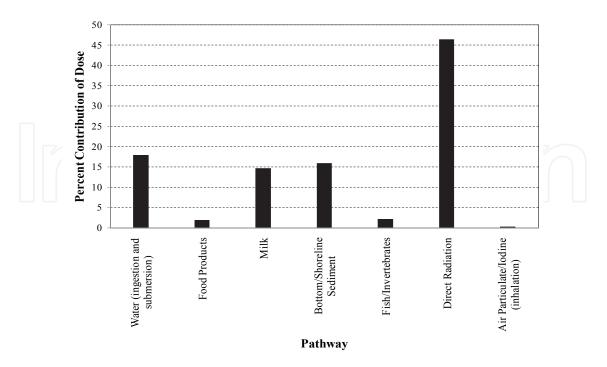
4.2.1 Cumulative dose contribution from effluent release pathways

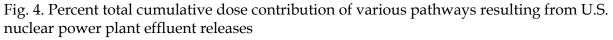
The percent contribution by each pathway to the public was determined using previously complied effluent data. The calculations provided by each plant from their REMP data identified the most important pathways for their respective sites. Fig. 4 shows the mean results for the seven most frequently sampled pathways or media for all plants. The greatest dose contributor comes from direct radiation released by the plant. This is especially true for BWR plants, which have no secondary loop for the reactor produced steam.

The water, milk, and sediment media, at about the same level, provide the next highest percent contributions to dose. Water media, which includes surface, ground, and drinking types, may contribute to dose to a much larger extent depending on the plant location. Those that are on saltwater sites may not have a known drinking water pathway, or dilution may be so great that submersion doses are low or non-existent. On the other hand, plants on freshwater sites typically have drinking water pathways and less dilution of their effluents (with the exception of sites located on the Great Lakes). All other pathways contribute to a much lesser extent.

| Year | Electrical Energy Produced (GW) | U.S. Population (x 104) | Annual Effective Dose (mSv GW-1 person-1) | | | | | | | |
|------|--|-------------------------------|---|--------------------------|-------------------------|--------------------------|-------------------------|-------------------------|-------------------------|--|
| | | | Gaseous Releases | | | Liquid Releases | | Total | | |
| | | | F/A Gases | Total I-131 | Tritium | Particulates | Tritium | F/D Gases | | |
| 1995 | 77.1 | 266,557 | 8.36 x 10-8 | 1.95 x 10 ⁻¹⁰ | 1.68 x 10-8 | 1.28 x 10-9 | 2.93 x 10-8 | 2.90 x 10-8 | 1.60 x 10-7 | |
| 1996 | 77.3 | 269,667 | 7.79 x 10 ⁻⁸ | 2.75 x 10 ⁻¹⁰ | 1.31 x 10-8 | 1.10 x 10-9 | 3.18 x 10-8 | 2.89 x 10-8 | 1.53 x 10-7 | |
| 1997 | 71.9 | 272,912 | 1.08 x 10-7 | 1.29 x 10 ⁻¹⁰ | 1.90 x 10-8 | 1.47 x 10-9 | 2.71 x 10-8 | 1.22 x 10 ⁻⁸ | 1.68 x 10-7 | |
| 1998 | 74.9 | 276,115 | 1.38 x 10-8 | 2.80 x 10-10 | 1.46 x 10-8 | 2.66 x 10-9 | 2.68 x 10-8 | 1.37 x 10-8 | 7.19 x 10 ⁻⁸ | |
| 1999 | 82.3 | 279,295 | 7.00 x 10-9 | 1.75 x 10 ⁻¹⁰ | 1.57 x 10-8 | 3.06 x 10-10 | 2.83 x 10-8 | 1.10 x 10-8 | 6.24 x 10 ⁻⁸ | |
| 2000 | 85.2 | 282,402 | 7.98 x 10 ⁻⁹ | 1.80 x 10 ⁻¹⁰ | $1.48 \ge 10^{-8}$ | 1.08 x 10 ⁻⁹ | 3.05 x 10 ⁻⁸ | 1.07 x 10 ⁻⁸ | 6.53 x 10 ⁻⁸ | |
| 2001 | 87.8 | 285,329 | 5.58 x 10-9 | 9.21 x 10-11 | 1.50 x 10-8 | 8.57 x 10 ⁻¹⁰ | 2.54 x 10-8 | 7.97 x 10-9 | 5.49 x 10 ⁻⁸ | |
| 2002 | 88.6 | 288,173 | 8.42 x 10-9 | 1.95 x 10 ⁻¹⁰ | 1.73 x 10-8 | 6.62 x 10 ⁻¹⁰ | 2.70 x 10-8 | 1.96 x 10-8 | 7.32 x 10 ⁻⁸ | |
| 2003 | 87.0 | 291,028 | 1.44 x 10-8 | 3.79 x 10 ⁻¹⁰ | 1.51 x 10-8 | 3.04 x 10-9 | 2.87 x 10-8 | 1.15 x 10 ⁻⁸ | 7.30 x 10-8 | |
| 2004 | 88.1 | 293,907 | 6.94 x 10-9 | 2.67 x 10 ⁻¹⁰ | 1.39 x 10-8 | 2.07 x 10-10 | 2.64 x 10-8 | 6.38 x 10-9 | 5.42 x 10 ⁻⁸ | |
| 2005 | 88.6 | 295,753 | 7.49 x 10-9 | 9.78 x 10 ⁻¹¹ | 1.59 x 10-8 | 5.11 x 10-9 | 2.77 x 10-8 | 5.99 x 10 ⁻⁹ | 6.23 x 10 ⁻⁸ | |
| 2006 | 89.3 | 298,593 | 5.40 x 10 ⁻⁹ | 9.84 x 10 ⁻¹¹ | 1.28 x 10 ⁻⁸ | 4.24 x 10 ⁻¹⁰ | 2.96 x 10 ⁻⁸ | 6.18 x 10 ⁻⁹ | 5.45 x 10 ⁻⁸ | |
| 2007 | 88.9 | 301,580 | 4.82 x 10-9 | 1.09 x 10 ⁻¹⁰ | 1.13 x 10-8 | 9.48 x 10-9 | 2.61 x 10-8 | 4.95 x 10-9 | 5.67 x 10 ⁻⁸ | |
| 2008 | 88.9 | 304,375 | 4.44 x 10-9 | 1.03 x 10 ⁻¹⁰ | 1.21 x 10-8 | 1.85 x 10-9 | 2.82 x 10-8 | 1.01 x 10-8 | 5.68 x 10 ⁻⁸ | |
| 2009 | 86.8 | 307,007 | 5.77 x 10-9 | 7.31 x 10-11 | 1.14 x 10-8 | 3.32 x 10 ⁻¹⁰ | 2.41 x 10-8 | 1.41 x 10-8 | 5.58 x 10-8 | |

Table 2. Average effective doses received by members of the public in the U.S. from commercial nuclear power plant radiological effluent releases





4.2.2 Plant pathway sampling

The investigation of the number of pathways sampled by each plant is important to gauge the diversity and scope of each plant's REMP in relation to each other. Used in conjunction with the results from Section 4.3.1, omissions of pathways can be reconsidered based on the overall percent contribution of dose. Obviously, programs will vary greatly since the power plant sites themselves also vary greatly. For example, plants may be located on coastal sites, in river valleys, in arid locations, or on small, man-made bodies of water. The level of human activity around the plant will also influence the degree of surveillance. The number of media sampled by plants ranged from five to more than 20, with a mean of 11.

The results of the percentage of plants sampling different media in their REMP are displayed in Fig. 5. As expected, 100% of plants sample for air particulate matter, ¹³¹I and direct radiation. Direct radiation is important as it is the greatest dose contributor to members of the public, especially from BWR plants. Milk, once sampled by nearly all plants, is now sampled by only 52% of plants. This is due to the reduction of the milk pathway (from cows or goats) in areas around plants. In lieu of milk sampling, plants may substitute broadleaf vegetation. One important characteristic of the data is that nearly 56% of plants sample groundwater. Recent unplanned tritium releases went unnoticed at several plants due to the lack of groundwater monitoring at those sites. In part due to this study and a national industry initiative, all plants began a groundwater monitoring program starting in 2006. At the time of this research and writing, updated data on nuclear power plant groundwater monitoring was not available. The dose important fish pathway is sampled by 88% of plants. Edible aquatic invertebrates are sampled by only 19% of the plants, mainly due to the lack of the pathway.

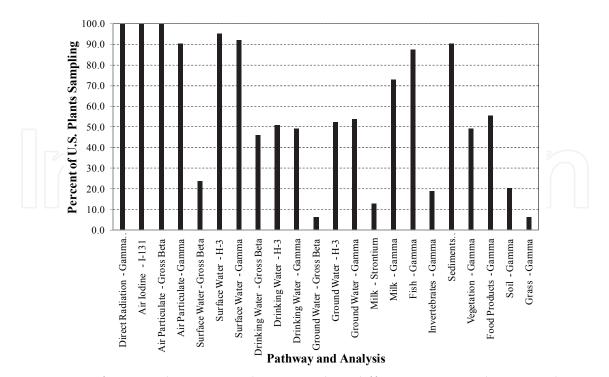


Fig. 5. Percent of U.S. nuclear power plants sampling different REMP pathways and performing specific analyses

4.2.3 REMP summary

Numerical results for the REMP summary are not listed here due to the breath of the study. Comparisons are only made between the control samples and the indicator or test measurements. Direct radiation pathways include radiation from buildings and plant structures, airborne material that might be released from the plant, cosmic radiation, "fallout", and the naturally occurring radioactive materials in soil, air and water. Analysis of the TLDs, which are used by all plants, indicated that there were no increased radiation levels attributable to plant operations. The mean BWR exposure was slightly higher than that for PWR plants, but the control locations actually gave higher mean values for all plant types.

Airborne pathway measurements include analyses of ¹³¹I, gross beta, and gamma spectra. For all ¹³¹I measurements, the values were below the LLD. Therefore, the reported mean and median measurements are for half of the LLD values. For the entire 13 years, only one plant reported an indicator value above the LLD. The gross beta and gamma spectral analyses showed no difference between the control and indicator stations. These airborne pathway measurements indicated that there was no increased radioactivity attributable to plant operations.

Terrestrial pathways that include milk, vegetation, grass, and food products, all gave calculated mean indicator measurements that were below that of the controls, Similarly for all waterborne pathways (drinking water, ground water, surface water, fish and invertebrates, and sediments), measurements of the media indicated that there was no adverse radiological impact to the surrounding environment attributed to plant operations for the study period. Individually, several plants have had measurements that resulted in indicator values being significantly higher than those from control stations.

The summary data strongly suggest that for the great majority (>99.9%) of the analyses, the control and indicator concentrations were indistinguishable. Potential plant produced radionuclides were detected in both indicator and control locations. In the past 13 years, the concentration of radionuclides in the environment has stayed nearly level. Prior to this time, radionuclide concentrations showed significant downward trends, mainly due to the decay of "fallout" radionuclides, evidenced in the historical data provided by the plants. The overall trend of the REMP data for these plants is "de minimis" levels of anthropogenic radioactivity with occasional samples showing radioactivity above the LLD. These data also indicate no correlation between total dose and distance from the plants.

The analytical results from the REMP study demonstrate that the routine operation of all facilities had no significant or measurable radiological impact to the environment for the study time period. Also, these environmental surveillance programs continued to demonstrate that the doses to members of the public as a result of nuclear plant operations remained significantly below the federally required dose limits specified in 10 CFR 20 and 40 CFR 190.

5. Tritium recapture study

Tritium is an isotope of hydrogen that decays with a half-life of 12.3 y to helium (³He), while emitting a low energy beta particle (18.6 keV). In nuclear reactors, tritium is formed in two different ways: by ternary fission of uranium, and by activation (Luykx & Fraser, 1986). Produced tritium that ultimately ends up in the nuclear reactor coolant can then be released into the environment through waste discharge in either gaseous or liquid forms (Kim & Han, 1999). Because tritium is generated in very large volumes, it is the predominant gaseous and liquid effluent released by these reactors.

Although tritium is not considered to be a particularly toxic radionuclide (Hill and Johnson 1993) and releases by commercial nuclear power plants have traditionally been well below regulatory limits, control and monitoring is important because of sensitive public concerns regarding radioactivity releases (Andersen 1995; Liu et al 2003). This is especially vital at the present time because recent, unplanned tritium releases have occurred at several nuclear power plants in the U.S. These unplanned releases may result in additional dose both to occupational workers and members of the public. This has led to increased scrutiny by plant owners and government agencies. In 2007, Harris also reported that the largest number of non-routine environmental sampling results was from tritium in surface water.

This section describes research conducted at the Cook Nuclear Plant (U.S.) to investigate the behaviour of tritium released in airborne effluents. In addition to studying the behaviour and movement of tritium in the environment, the study has been conducted to help develop a sampling and analysis protocol for other plants to adopt. Methods and results are primarily taken from Hinchcliffe, 2010.

5.1 Background

At the Cook Nuclear Plant, a site consisting of two PWRs, an event in 2007 set off an investigation into tritium release on the site. The plant is located in the state of Michigan along the eastern shore of Lake Michigan. In May of that year sampling began on the site as a result of a leak containing tritium found outside the auxiliary building. This sampling resulted in elevated tritium levels being discovered in the north storm drain outfall, which drains into Lake Michigan. This outfall had previously been sampled on a semi-annual basis with no previous indication of elevated tritium levels (Harris et al., 2008). The investigation concluded that tritium originating in the spent fuel pool was leaving the monitored vents on top of the containment buildings of both units and contributing to the elevated levels. Findings led to increased sampling of precipitation, air-conditioning condensate, surface and well water, and frost formed in refrigerators/freezers (Harris et al. 2008). The importance of precipitation was noted in the form of washout in which tritium can be scavenged by falling raindrops and tritium vapour can exchange with ordinary water in the atmosphere (Chamberlain & Eggleton, 1964; Tokuyama & Oonishi, 1997; Harris et al., 2008). In this way, precipitation becomes contaminated with tritium and sampling can be used to calculate a washout coefficient. This coefficient allows for estimation of tritium washout due to the precipitation which can be used to estimate the extent of tritium recapture on site property (Harris et al., 2008).

After an initial study in 2007 it was decided that additional sampling should be performed to determine the variations in recapture on the site as the weather changed through the year, and that snow should be sampled during the winter. Snow was sampled to determine if there was a difference in washout between rain and snow and to determine if recapture of tritium occurred differently with snow than it did with rain.

5.2 Methodology

The investigation of tritium washout was performed by taking samples at pre-determined site locations and analysing the collected samples for tritium. For rain collection, gauges

with a surface area of 7.85 x 10⁻⁴ m² were placed 0.25 m above the ground surface. Thirty two (32) rain gauges were placed in each land-based 22.5° sector of the plant site. Additional gauges were located in the predominant seasonal wind directions; SW in the summer and N and NE in the winter. The rain samples from this investigation were collected between 22 October 2007 and 21 October 2008 during 13 rain events. Snow was collected on 28 March 2008, 11 February 2009 through 16 February 2009, and 5 January 2010 through 10 March 2010.

Samples were taken both inside and outside the plant Protected Area, as shown in Fig. 6 and 7. The sample locations varied in distance from the containment stacks, ranging from 5 m (location 12) to 1.5 km (location 32). For control purposes, location 16 was placed 24 km from the release point. Snow was collected immediately after snow fall events, sampled from the surface of existing snow piles, and sampled with cores to evaluate depth differences from existing piles. Surface snow samples were obtained using 20 mL collection cups. Core samples yielded four to six, 10 cm sections, depending on pile depth. Ground level concentrations of tritium were also determined for calculation of a new washout coefficient. Sample analysis was done using a liquid scintillation counter (Model 2910TR, PerkinElmer). The samples were prepared by mixing 5 mL to 10 mL of the sample with 10 mL of scintillation cocktail in 20 mL vials and counting for 30 minutes. The tritium counting efficiency was 60%, and the lower limit of detection (LLD) was 30 Bq (819 pCi).

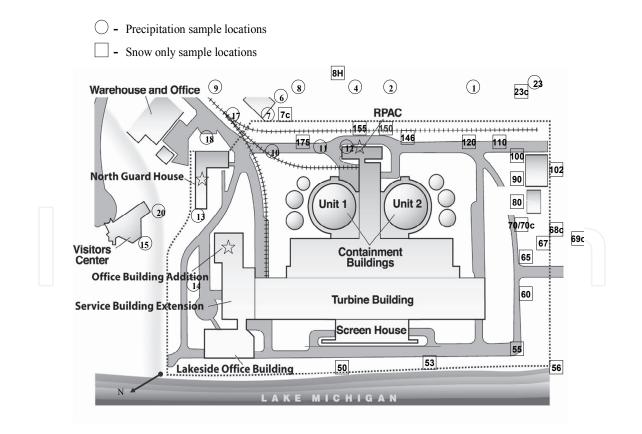
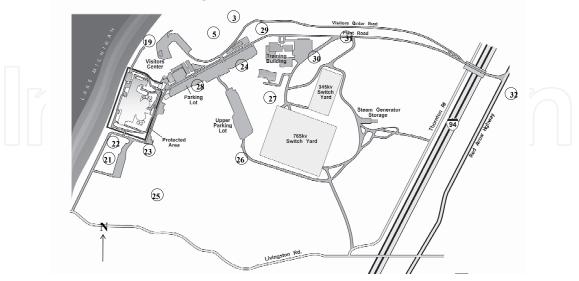


Fig. 6. Rain and snow sample locations located outside the Cook Nuclear Plant protected area



○ - Rain and Snow sample locations

Fig. 7. Sample locations for rain and snow within and immediately outside the Cook Nuclear Plant protected area

5.3 Results

5.3.1 Tritium concentrations

In total, 23 of the 32 rain locations had a positive result for tritium at some point in the study. With the snow, 44 of 56 sample locations resulted in a tritium concentration above the LLD. The highest rain concentration was 514 Bq L⁻¹ (13,900 pCi L⁻¹), and occurred at location 18, 130 m NE of the Unit 1 containment vent. The highest snow concentration was 1,014 Bq L⁻¹ (27,400 pCi L⁻¹), and occurred at snow sample location 110, 140 m SSE of the Unit 1 containment vent. The highest concentrations of tritium in the snow occurred in the old snow that had accumulated in piles on site. The positive results were found to match well with the meteorological data, with positive samples collected in the predominant wind direction.

There were a total of five core samples taken and all indicated the presence of tritium in at least one core piece. Four of the five core samples showed an increasing concentration of tritium closer to the surface of the snow pile. The only core sample that did not show this pattern was core sample V which was taken from the visitor's parking lot, 200 m NNE from the Unit 1 containment vent. All core pieces in this sample were below LLD, with the exception of core piece "2" (second piece from the bottom of the core).

5.3.2 Washout

A rain washout coefficient was calculated from the measured tritium concentrations found in the samples, the atmospheric tritium level, and the meteorological data gathered at the Cook Nuclear Plant. The washout coefficient is calculated by:

$$\Lambda = \frac{\omega}{\chi_0 \times H_{\text{eff}}}$$
(3)

where: Λ = washout coefficient (s⁻¹); ω = tritium deposition rate (Bq m⁻² s⁻¹); χ_0 = atmospheric tritium concentration at ground-level (Bq m⁻³); and H_{eff} = effective height (m). The effective height is calculated using the dispersion equation (Chamberlain & Eggleton, 1964):

$$H_{eff} = \frac{\sum_{s} \frac{Q}{\sqrt{2\pi}\sigma_{ys}U_{S}}}{X_{0,cal}}$$
(4)

where: Q = tritium emission rate (Bq m⁻² s⁻¹); σ_{ys} = standard deviation of distribution of concentration in the y direction (m); U_s= mean wind speed (m s⁻¹); $X_{0,cal}$ = mean concentration for the ground-level atmospheric concentration (m). The subscript S refers to the atmospheric stability. Using the local meteorological conditions, the deposition rate is given as:

$$\omega = \frac{C \times I_R}{3600} \tag{5}$$

where: C = tritium concentration in rainwater (Bq L⁻¹); and I_R = mean rainfall intensity (mm h⁻¹).

The emission rates were calculated using the continuous tritium release rates from the Cook Nuclear Plant Annual Radioactive Effluent Release Reports (ARERR) for the periods on which the precipitation events occurred. The precipitation intensities were calculated for each precipitation event, with an average value of 3.1 mm h⁻¹ for rain, and 1.1 mm h⁻¹ for the snow. For the rain samples the washout coefficient values ranged from 7.84 x 10-7 s⁻¹ to 1.13 x 10-4 s⁻¹ with an average washout coefficient of $(2.04 \pm 1.85) \times 10-5 \text{ s}^{-1}$. Washout coefficients were only calculated for the fresh snow. The reason for this as that as the snow sits, it accumulates tritium from the atmosphere, and thus skews the value of the washout coefficients for the fresh snow was analysed shortly after the snow event. The calculated washout coefficients for the fresh snow ranged from 2.21 x 10⁻⁶ s⁻¹ to 2.33 x 10⁻⁵ s⁻¹ with an average value of $(1.30 \pm 0.75) \times 10-5 \text{ s}^{-1}$. Upon comparing the rain and snow washout coefficient sample populations using a two sample test, it was determined that there was a significant difference between the washout coefficients in the snow and rain at the 0.05 decision rule level.

5.4 Discussion

Several other studies have reported values of washout coefficients in the literature. A theoretical washout coefficient based on the rate of exchange of tritiated water (HTO) vapour by rain, was calculated to be 10^{-4} s⁻¹ (Chamberlain & Eggleton, 1964). In Japan, Tokuyama & Oonishi (1997) used a mean rainfall intensity of 2 mm h⁻¹ and calculated a washout coefficient from releases at a nuclear plant to be $(7.3 \pm 4.1) \times 10^{-5}$ s⁻¹. The previous study at Cook Nuclear Plant calculated a washout coefficient of $(9.70 \pm 8.40) \times 10^{-5}$ s⁻¹, based on a rainfall intensity of 6.2 mm h⁻¹ (Harris et al., 2008). The rain washout coefficient in this study is smaller than those previously listed, but on the same order of magnitude as Tokuyama & Oonishi and Harris et al. The release rates used for this study were monthly averages rather than instantaneous rates used in several of the other studies. In addition, the calculated effective heights were calculated higher in this study than the other studies that listed the effective height used.

Annual fluctuations of weather have multiple effects on the process of tritium recapture at nuclear sites. The locations of greatest recapture vary between seasons and even within

particular seasons. Snow recapture at the Cook Nuclear Plant occurred in locations of the site that previously were not known for rain recapture, thus presenting new pathways of tritium movement needing to be studied. Snow also presents a unique problem, as it is not removed from the site at the same rate that rain would be removed, thus allowing for tritium to be absorbed into snow piles that accumulate around the site. This allows for snow piles to obtain tritium concentrations far above what simple washout can cause in precipitation alone. The piles also appear to accumulate a higher concentration of tritium on and near the surface of the pile where the snow is exposed to the atmosphere.

The results here suggest that individual sites will have to come up with their own sampling methods based on individual site characteristics such as annual fluctuations in wind and precipitation. Variables will change from site to site, and long term annual studies will need to be performed to fully comprehend the behaviour of tritium recapture. A single industry model would be unable to account for the many individual variables that would change between sites. Studies are on-going at Cook Nuclear Plant to better understand tritium behaviour.

6. Future research

Although the research presented here provided important insights into commercial nuclear power plant discharges, more studies are needed to truly understand effluent trends and nuclear power plant radioactivity. As long as commercial nuclear power plants continue to operate and release radioactivity into the environment, there will always be a need to monitor, track, and evaluate these releases. This is especially true as new power plants are constructed and existing plants make operational changes. Evolutions in standards, recommendations, and regulations may also warrant studies to address the potential impact. One expansive area of research that is developing comes from recommendations made by the ICRP in Publications 103 and 108 (2007, 2009). Specifically there is mention of radiological protection of the environment and the development of reference plants and animals. If nations adopt these recommendations, large scale studies at nuclear power plants may need to be undertaken.

In the U.S., a number of potential new areas of research involving nuclear power radiological releases are being investigated. First, revisions in 2009 of USNRC Regulatory Guides 1.21 and 4.1 have prompted plants to begin monitoring their ¹⁴C emissions. Only a few plants had done this prior to the release of these revisions and now several organizations are evaluating methods and procedures to best accomplish this. Other countries are also evaluating this scenario, if they are not already monitoring for ¹⁴C. In conjunction with the tritium washout study presented in Section 5, several plants are conducting their own studies on tritium releases and movement. The majority of plants in the U.S. are now monitoring groundwater and studying its movement and transport of radionuclides. Finally, with the construction and operation of new plants, studies on effluent fate and transport, dose assessment and environmental impact are sure to be conducted.

7. Conclusion

In conclusion, the research presented here will hopefully provide stakeholders with some additional insights on assessing nuclear power plant radiological releases and environmental monitoring programs. Evaluation of radionuclide impacts in terms of potential increased dose to people, in relation to natural background, is necessary to determine the true significance of any detection. Maintained effluent tracking and analysis will continue to be important for the future of the commercial nuclear power industry. Effluent trending may also reveal insight into the effect of increased reactor lifetime operation on radioactivity releases. Currently, many plants have been approved for, or are applying for, operating license extensions. Tracking data for siting of new nuclear power plants may also be used to determine environmental radionuclide build-up and long-term nuclear power health effects.

Although the research presented here showed that normal nuclear power operations have no significant effect on the environment, continued surveillance is important. The potential development of new pathways may require changes in monitoring techniques. Doses to members of the general public, and even occupational workers, may increase because of these pathways. Finally, continued industry analysis of radiological effluent releases and environmental monitoring are important for maintaining favourable public opinion about nuclear power. The more accurate, scientifically based information that citizens can be provided with, the more likely they are to make informed, non-emotional judgments about nuclear power.

8. Acknowledgment

This chapter was written from research results and material developed by the author over the last decade. I wish to thank the many friends, family, and colleagues, too numerous to mention by name, for their contributions. Special appreciation goes out to Dr. David Miller, Dr. Richard Brey, Dr. Thomas Gesell, Dr. George Sandison, Dr. George Miley, and the late Dr. Herman Cember. Their guidance and support have truly helped shape not only my career, but my life as well. I would also like to thank Dr. Kenneth Sejkora for sharing his knowledge in nuclear reactor aspects of environmental monitoring and effluents. I am also indebted to my students, especially William Hinchcliffe, Gavin Hawkley, and Peter Lee, for their assistance in many parts of the research presented in this chapter. Lastly, and most importantly, I would like to thank my wife, Dr. Maria Okuniewski. If it were not for her constant support, encouragement, and love, I would not be where I am today.

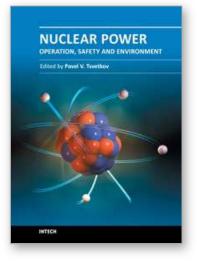
9. References

- Andersen R. (1995). Criteria for radiological releases, In: *Radiation Protection at Nuclear Reactors*, C. Maletskos, (Ed.), 21-44, MPP, ISBN 978-094-4838-55-6, Madison, USA
- Chamberlain, A. & Eggleton, A. (1964). Washout of tritiated water vapour by rain. *J Air Wat Poll*, Vol.8, pp. 135-149
- Eisenbud, M. & Gesell, T. (1997). *Environmental Radioactivity* (4th), Academic Press, ISBN 978-012-2351-54-9, San Diego, USA
- Electric Power Research Institute (2003). *Strategies for managing liquid effluents-options, actions, and results,* EPRI, Palo Alto, USA
- Glasstone, S. & Jordan, W. (1980). *Nuclear Power and its Environmental Effects*, American Nuclear Society, ISBN 978-089-4480-22-5, La Grange Park, USA
- Gilbert, R. (1994). *Statistical Methods for Environmental Pollution Monitoring*, Van Nostrand Reinhold, ISBN 978-0471288787, New York, USA

Radiological Releases and Environmental Monitoring at Commercial Nuclear Power Plants

- Harris, J. (2002). *Comparative Study of Commercial Nuclear Power Plant Radiological Effluents*, University of Illinois, Urbana, USA
- Harris, J. (2007). Public Health Analysis Resulting from Commercial Nuclear Power Plant Radiological Emissions, Purdue University, West Lafayette, USA
- Harris, J. & Miller, D. (2008). Radiological effluents released by U.S. commercial nuclear power plants from 1995-2005. *Health Phys*, Vol.96, pp. 734-743
- Harris, J.; Miller, D.& Foster, D (2008). Tritium behavior at a nuclear power reactor due to airborne releases. *Health Phys*, Vol.95, pp. 203-212
- Hill, R. & Johnson, J. (1993). Metabolism and dosimetry of tritium. *Health Phys* Vol.65, pp. 628-647
- Hinchcliffe, W. (2010). Investigation of Tritium Recapture at Cook Nuclear Power Plant from Airborne Effluent Releases, Idaho State University, Pocatello, USA
- Hollander, M. & Wolfe, D. (1999). Nonparametric Statistical Methods, Wiley, ISBN 978-047-1190-45-5, New York, USA
- IAEA (2010). Nuclear Power Reactors in the World (2010), IAEA, ISBN 978-92-0-105610-8, Vienna, Austria
- IAEA (2011). Nuclear Power Plant Information, In: *IAEA PRIS Database*, 10.03.2011, Available from: http://www.iaea.org/cgi-bin/db.page.pl/pris.charts.htm
- ICRP (1977). Recommendations of the ICRP ICRP Publication 26, Pergamon Press, ISBN 978-085-951-080-6, Oxford, England
- ICRP (1978). Radionuclide Release into the Environment: Assessment of Doses to Man ICRP Publication 29, Pergamon Press, ISBN 978-008-0323-35-0, Oxford, England
- ICRP (1991). 1990 Recommendations of the ICRP -ICRP Publication 60, Pergamon Press, ISBN 978-008-0411-44-6, Oxford, England
- ICRP (2007). 2007 Recommendations of the ICRP ICRP Publication 103, Pergamon Press, ISBN 978-070-2030-48-2, Oxford, England
- ICRP (2009). Environmental Protection: the Concept and use of Reference Animals and Plants -ICRP Publication 108, Pergamon Press, ISBN 978-044-4529-34-3, Oxford, England
- Jablon, S. et al. (1991). Cancer in populations living near nuclear facilities, a survey of mortality nationwide and incidence in two states. *J Am Med Assoc* Vol.265, pp. 1403-1408
- Kahn, B. (1980). Composition and measurement of radionuclides in liquid effluent from nuclear power stations, In: *Effluent and Environmental Radiation Surveillance*, J. Kelly, (Ed.), 63-74, American Society for Testing and Materials, Johnson, USA
- Kim, C. & Han, M. (1999). Dose assessment and behavior or tritium in environmental samples around Wolsong nuclear power plant. *Appl Radiat Isot*, Vol.50, pp. 783-791
- Liu, C.; Chao, J. & Lin, C. (2003). Tritium release from nuclear power plants in Taiwan. *Health Phys*, Vol.84, pp. 361-367
- Lopez-Abente, G. et al. (1999). Leukemia, lymphomas, and myeloma mortality in the vicinity of nuclear power plants and nuclear fuel facilities in Spain. *Cancer Epidemiology, Biomarkers & Prevention,* Vol8, pp.925-934
- Luykx, F. & Fraser, G. (1986). Tritium releases from nuclear power plants and nuclear fuel processing plants. *Radiat Prot Dosim* Vol16, pp. 31-36
- NCRP (1985). Carbon-14 in the environment NCRP Report No. 81, NCRP, ISBN, Bethesda, USA

- NCRP (1987). Public radiation exposure from nuclear power generation in the United States - NCRP Report No. 92, NCRP, ISBN , Bethesda, USA
- Nedveckaite, T. et al. (2000). Environmental releases of radioactivity and the incidence of thyroid disease at the Ignalina NPP. *Health Phys*, Vol.79, pp. 666-674
- Patrick, C. (1977). Trends in public health in the population near nuclear facilities: a critical assessment. *Nucl Saf,* Vol.18, pp. 647-662
- Quindos Poncela, L. et al. (2003). Natural radiation exposure in the vicinity of Spanish nuclear power stations. *Health Phys*, Vol.85, pp. 594-598
- Shleien, B.; Ruttenber, A. & Sage, M. (1991). Epidemiologic studies of cancer in populations near nuclear facilities. *Health Phys*, Vol61, pp. 699-713
- Tokuyama, H. & Oonishi, M. (1997). Precipitation washout of tritiated water vapor from a nuclear reactor. *J Environ Radioact*, Vol.34, pp. 59-68
- United Nations Scientific Committee on the Effects of Atomic Radiation (2000). Sources and Effects of Ionizing Radiation, UN, ISBN 978-921-1422-39-9, New York, USA
- United States Census Bureau (2010). *Statistical abstract of the United States:* 2010, GPO, Washington, D.C., USA
- USEPA (1977). Environmental radiation protection standards for nuclear power operations. 40 CFR 190, 42 FR 2860, January 13, 1977, GPO, Washington, D.C., USA
- USNRC (1975). Programs for monitoring radioactivity in the environs of nuclear power plants. Regulatory Guide 4.1, rev. 2, GPO, Washington, D.C., USA
- USNRC (1976a). Calculation of releases of radioactive materials in gaseous and liquid effluents from boiling water reactors, NUREG-0016, GPO, Washington, D.C., USA
- USNRC (1976b). Calculation of releases of radioactive materials in gaseous and liquid effluents from pressurized water reactors, NUREG-0017, GPO, Washington, D.C., USA
- USNRC (1978). Preparation of radiological effluent technical specifications for nuclear power plants, NUREG -0133, GPO, Washington, D.C., USA
- USNRC (1991). *Standards for protection against radiation*. 10 CFR 20, 56 FR 23361, May 21, 1991, GPO, Washington, D.C., USA
- USNRC (1995). Dose commitments due to radioactive releases from nuclear power plant sites in 1991, NUREG/CR-2850, GPO, Washington, D.C., USA
- USNRC (1996). Technical specifications on effluents from nuclear power reactors. 10 CFR 50.36a, 61 FR 39299, July 29, 1996, GPO, Washington, D.C., USA
- Vold, E. (1983). Ingestion pathway factor in dose assessment for annual airborne releases of radioactivity. *Health Phys*, Vol.47, pp. 429-441
- Walmsley, A. et al. (1991). The distribution of doses to members of the public around UK civil nuclear sites. *Radia Protect Dosimetry*, Vol.36, pp. 215-218
- Ziqiang, P. et al. (1996). Radiological environmental impact of the nuclear industry in China. *Health Phys*, Vol71, pp. 847-862



Nuclear Power - Operation, Safety and Environment Edited by Dr. Pavel Tsvetkov

ISBN 978-953-307-507-5 Hard cover, 368 pages Publisher InTech Published online 06, September, 2011 Published in print edition September, 2011

Today's nuclear reactors are safe and highly efficient energy systems that offer electricity and a multitude of co-generation energy products ranging from potable water to heat for industrial applications. At the same time, catastrophic earthquake and tsunami events in Japan resulted in the nuclear accident that forced us to rethink our approach to nuclear safety, design requirements and facilitated growing interests in advanced nuclear energy systems, next generation nuclear reactors, which are inherently capable to withstand natural disasters and avoid catastrophic consequences without any environmental impact. This book is one in a series of books on nuclear power published by InTech. Under the single-volume cover, we put together such topics as operation, safety, environment and radiation effects. The book is not offering a comprehensive coverage of the material in each area. Instead, selected themes are highlighted by authors of individual chapters representing contemporary interests worldwide. With all diversity of topics in 16 chapters, the integrated system analysis approach of nuclear power to our readers as one of the promising energy sources that has a unique potential to meet energy demands with minimized environmental impact, near-zero carbon footprint, and competitive economics via robust potential applications. The book targets everyone as its potential readership groups - students, researchers and practitioners - who are interested to learn about nuclear power.

How to reference

In order to correctly reference this scholarly work, feel free to copy and paste the following:

Jason T. Harris (2011). Radiological Releases and Environmental Monitoring at Commercial Nuclear Power Plants, Nuclear Power - Operation, Safety and Environment, Dr. Pavel Tsvetkov (Ed.), ISBN: 978-953-307-507-5, InTech, Available from: http://www.intechopen.com/books/nuclear-power-operation-safety-and-environment/radiological-releases-and-environmental-monitoring-at-commercial-nuclear-power-plants



InTech Europe

University Campus STeP Ri Slavka Krautzeka 83/A 51000 Rijeka, Croatia Phone: +385 (51) 770 447 Fax: +385 (51) 686 166 InTech China

Unit 405, Office Block, Hotel Equatorial Shanghai No.65, Yan An Road (West), Shanghai, 200040, China 中国上海市延安西路65号上海国际贵都大饭店办公楼405单元 Phone: +86-21-62489820 Fax: +86-21-62489821

www.intechopen.com

IntechOpen

IntechOpen

© 2011 The Author(s). Licensee IntechOpen. This chapter is distributed under the terms of the <u>Creative Commons Attribution-NonCommercial-ShareAlike-3.0 License</u>, which permits use, distribution and reproduction for non-commercial purposes, provided the original is properly cited and derivative works building on this content are distributed under the same license.



