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Radiological Impacts and Hydrogeological Challenges for Groundwater Systems during Nuclear Power Plant Operations: A Comprehensive Review

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ABSTRACT

The groundwater systems near nuclear power plants (NPPs) are critical pathways for possible radionuclide migration, both during normal operation and during accidental events. This review gathers the existing scientific literature on the radiological consequences, monitoring techniques and hydrogeological modeling difficulties of groundwater protection in the context of NPP operation. Important radionuclides of concern, such as cesium-137 (^{137}Cs), strontium-90 (^{90}Sr) and tritium ((^{3}H)are described and their analytical detection limits are set under regulations. subsurface transport modeling by 3D finite difference methods and assessment of groundwater vulnerability using DRASTIC model. In addition, the Water Quality Index of the Canadian Council of Ministers of the Environment (CCME WQI) is examined for use in assessing the hydro-geochemical stability in the vicinity of operating sites. The field-based evidence on tritium dynamics, assessment of public doses and ambient radiation baselines are presented in the case studies from Sanmen NPP, Rooppur NPP, Zaporizhzhia NPP and Krško NPP. Finally, this review addressed some of the engineered mitigation challenges, such as the application of a permeable reactive barrier using clay mineral adsorbents (saponite, sepiolite, bentonite) and provide suggestions for future integrated monitoring networks.

Keywords: *Groundwater quality, Nuclear power plants (NPPs), Radiological environmental monitoring, Tritium dynamic, DRASTIC vulnerability*

1 | Introduction

Nuclear power plants (NPPs) are a key component of global low-carbon energy production, and the long operation of NPPs requires strict environmental protection, especially the protection of freshwater bodies in the vicinity of NPPs [1, 2]. Generation 3 Pressurized Water Reactors (PWRs) are routinely designed in pairs and sited in coastal areas or river basins to guarantee an uninterrupted and immense source of secondary-side cooling water [3] as in the case of the generic APR1400 design. Hydrological pathways between plant facilities and local groundwater bodies are identified based on local geology and on-site engineering. In general, the nuclear and turbine islands are supported by competent bedrock (up to 30 m deep) or compacted/treated soil structures to provide seismic stability and to reduce settlement [3]. Large mat foundations and complicated tunnels for water intake and discharge are designed to separate plant activities from the regional aquifers [3].

Groundwater is susceptible to chronic or accidental releases of radioactivity, no matter how sophisticated the passive safety systems are (e.g., NuScale's submerged reactor pools and passive containment cooling, which help reduce the impact of design-basis Loss of Coolant Accidents [4]). Groundwater contamination can occur through leakage from large cooling reservoirs, failure of temporary radioactive waste storage facilities or leaks in underground cooling water lines [5, 6]. To assess and control these environmental risks, regulatory agencies require extensive

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engineering surveys before and during operation.

Set up hydrogeological background conditions and forecast contaminant flow paths and travel times [7]. This manuscript summarizes the hydrogeological problems, monitoring techniques and modeling tools which are used for the protection of groundwater systems during the life cycle of NPPs.

2 | Key Radionuclides and Dose Assessment

During the operation of an NPP, comprehensive Radiological Environmental Monitoring Plans (REMPs) are needed to characterize the environmental radioactivity in various media, such as air, soil, biota and water [2]. Groundwater monitoring is particularly critical since any radioactivity released may directly affect food chains or domestic drinking water. The radionuclides that are likely to occur in groundwater are classified according to the NPP life cycle: operational phase ('O') and dismantling and decommissioning phase ('D') [2]. The most important radionuclides of concern in routine situations are tritium (^{3}H), cobalt-60 (^{60}Co), strontium-90 (^{90}Sr), cesium-134 (^{134}Cs), cesium-137 (^{137}Cs) and iodine-131 (^{131}I). By contrast, decommissioning activities generate extra activation and fission products, including nickel-63 (^{63}Ni), carbon-14 (^{14}C) and transuranic radionuclides, such as americium-241 (^{241}Am) and plutonium-239 (^{239}Pu) [2].

The radiological risk to public health from contaminated groundwater is estimated by calculating the committed annual effective dose from the intake of contaminated water. The annual ingestion dose E(50) (mSv/year) is determined from the following mathematical formulation according to the guidelines set by the ICRP [2]:

$$E(50) = \sum_j e_j(50) \cdot \mathrm{I}_{j,ing} \quad (1)$$

where $e_j(50)$ is the committed effective dose coefficient for ingestion of radionuclide j (mSv/Bq) [2] and $I_{j,ing}$ is the annual activity intake of radionuclide j (Bq/year), expressed as [2]:

$$I_{j,ing} = A_j \cdot C_j \quad (2)$$

In equation (2), A_j is the annual consumption of water (L/year) and C_j is the activity concentration of radionuclide j in water (Bq/L) [2]. Groundwater monitoring programs must have highly sensitive derived detection limits ($y^{\#}$), ideally 10% of the groundwater guidance Activity Concentration Levels (ACL) for drinking water [2] to ensure regulatory compliance and public safety. As per ISO 11929 standard and Guide to the Expression of Uncertainty in Measurement (GUM), the decision threshold (y^*) and detection limit ($y^{\#}$) are parameters dependent on the background and are calculated by propagation of standard uncertainties [2]:

$$y^* = k_{1-\alpha} \cdot \tilde{u}(0) \quad (3)$$

$$y^{\#} = y^* + k_{1-\beta} \cdot \tilde{u}(y^{\#}) \quad (4)$$

The quantiles $k_{1-\alpha}$ and $k_{1-\beta}$ are usually set at 1.65 for $\alpha = \beta = 0.05$, and $\tilde{u}(0)$ is the combined standard uncertainty of the measurand at the value of the measurand equal to zero, and $\tilde{u}(y^{\#})$ is the uncertainty at the detection limit [2].

Table 1 | Radionuclides of Concern in Groundwater throughout NPP Lifecycle [2, 6].

Radionuclide	Symbol	NPP Stage	Half-life	Derived DL ($y^{\#}$, Bq/L)
Tritium	^{3}H	O, D	12.3 years	7.61×10^{1}
Cobalt-60	^{60}Co	O, D	5.27 years	4.03×10^{-1}
Strontium-90	^{90}Sr	O, D	28.8 years	4.89×10^{-2}
Cesium-137	^{137}Cs	O	30.1 years	1.05×10^{-1}
Nickel-63	^{63}Ni	D	100.1 years	9.13×10^{0}
Carbon-14	^{14}C	D	5,730 years	2.36×10^{0}

3 | Subsurface Flow and Contaminant Modeling

Numerical simulation is used to solve the governing equations of groundwater flow [3, 6] to predict migration of potential radionuclide plumes in groundwater. The transient flow of groundwater in a three dimensional, heterogeneous and anisotropic porous medium is represented by the following partial differential equation [3, 6]:

$$\frac{\partial}{\partial x}\left(K_x \frac{\partial h}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial h}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z \frac{\partial h}{\partial z}\right) = S_s \frac{\partial h}{\partial t} + W \quad (5)$$

where K_x, K_y, and K_z are the hydraulic conductivities along the principal coordinate axes (m/day), h is the hydraulic head (m), S_s is the specific storage (1/m), t is time (day), and W is the volumetric flux per unit volume representing sources/sinks such as recharge, well pumpage, or seepage (1/day) [3].

For steady-state, homogeneous conditions in a bounded aquifer, Equation (5) reduces to two dimensions to Laplace's equation, which is solved numerically by a central finite difference approximation. This results in the usual five-point finite difference formula [3]:

$$h_{i-1,j} + h_{i+1,j} + h_{i,j-1} + h_{i,j+1} - 4h_i, j = 0 \quad (6)$$

$$h_{i,j}^{m+1} = \frac{h_{i-1,j}^{m} + h_{i+1,j}^{m} + h_{i,j-1}^{m}}{3} \quad (7)$$

$$h_{i,j}^{m+1} = \frac{h_{i-1,j}^{m} + h_{i,j-1}^{m}}{2} \quad (8)$$

For unconfined aquifers, where the water table is dynamic and restricts flow, the Jacobi iteration needs to be adapted to allow for cells that intersect the water table. Since the hydraulic con-

ductivity is assumed to be zero above the water table, the unconfined boundary cells might have only 2 or 3 active neighbors rather than 5 [3].

Equation (7) is for a cell that has three adjacent active cells, and Equation (8) is for a boundary cell that has two adjacent active cells [3]. Convergence is obtained if the maximum absolute difference between successive iterations is less than a pre-defined tolerance, which is usually set to 5×10^{-4} m [3].

Equations (7) and (8) are based on linear head variations with standard node-averaging approximations. For unconfined aquifers in field applications, large water table fluctuations result in large non-linear changes in transmissivity (T = K h), and Picard or Newton-Raphson iterations may be necessary to ensure numerical stability when groundwater drawdown or recharge is rapid.

4 | Groundwater Vulnerability Assessment: The DRASTIC Model

Groundwater vulnerability mapping is a strategic tool to evaluate the susceptibility of groundwater to potential contamination. The US EPA DRASTIC model is commonly used for mapping intrinsic groundwater vulnerability in the vicinity of critical facilities such as NPPs [8]. The model combines seven hydrogeological parameters by a weighted scoring system [8]:

$$DRASTIC\ Index = D_r D_w + R_r R_w + A_r A_w + S_r S_w + T_r T_w + I_r I_w + C_r C_w \quad (9)$$

The seven parameters are represented by the letters: Depth to water table (D), net Recharge (R), Aquifer media (A), Soil media (S), Topography/Slope (T), Impact of the vadose zone (I), and hydraulic Conductivity (C). The subscript r is for the rating (1 to 10, depending on the relative susceptibility) and w is for the weighting factor (1 to 5, depending on the importance of the parameter) [8].

The usefulness of the DRASTIC model was illustrated in the area of the Zaporizhzhia NPP, where quaternary maps were prepared using GIS and historical borehole data were used for the delineation of intrinsic vulnerability [8]. Highly vulnerable zones (DRASTIC indices > 140) were mapped along the active stream channels and quaternary river terrace deposits of the Dnipro River where unconfined, highly conductive sandy aquifers are directly exposed [8]. This spatial analysis will help to optimize the placement of groundwater monitoring wells and prioritize localized remedial action should a radiological release occur.

5 | Hydrogeochemical Baselines and Water Quality Indices

Groundwater quality assessment during the operating life of an NPP must be able to differentiate between natural hydrogeochemical conditions and possible anomalies caused by the NPP [9, 10]. The Canadian Council of Ministers of the Environment Water Quality Index (CCME WQI) is a standard mathematical method for reducing complex water quality data to a single index value that is easily understood. The CCME WQI is based on three factors: Scope (F_1), Frequency (F_2), and Amplitude (F_3) which are measured as follows [9, 10]:

$$F_1 = \left(\frac{Number\ of\ failed\ parameters}{Total\ number\ of\ parameters} \right) \times 100 \quad (10)$$

$$F_2 = \left(\frac{Number\ of\ failed\ tests}{Total\ number\ of\ tests} \right) \times 100 \quad (11)$$

To compute the amplitude factor, the individual excursion of a failed test is determined. If the excursion is for a parameter that cannot be greater than a specified regulatory limit, then the excursion is determined as follows [9, 10]:

$$excursion_i = \left(\frac{Failed\ Test\ Value_i}{Objective_j} \right) - 1 \quad (12)$$

On the other hand, if a parameter (e.g. dissolved oxygen) cannot be below a desired level, the excursion is [9, 10]:

$$excursion_i = \left(\frac{Objective_j}{Failed\ Test\ Value_i} \right) - 1 \quad (13)$$

The normalized sum of excursions (nse) is computed for all the tests and F_3 is obtained by an asymptotic scaling function [9, 10]:

$$F_3 = \frac{nse}{0.01 \cdot nse + 0.01} \quad (14)$$

Lastly, 100 is subtracted from the three dimensional vector distance to obtain the CCME WQI score [9, 10]:

$$CCME\ WQI = 100 - \left(\frac{\sqrt{F_1^2 + F_2^2 + F_3^2}}{1.732} \right) \quad (15)$$

CCME WQI scores, in the baseline hydro-geochemical assessment carried out near the Rooppur NPP (RNPP) in Bangladesh, were found to be in the range of 34 to 48, which is classified as 'poor' or 'marginal' for drinking water [9, 10]. High natural concentrations of iron, manganese and total dissolved solids (TDS) [9, 10] were responsible for this low baseline rating. The results of Principal Component Analysis (PCA) with varimax rotation showed that natural silicate weathering and agricultural activities are the main sources of mineralization of groundwaters [10]. It is important to establish these high resolution baseline chemical profiles because they help to avoid false alarms for operational NPP releases due to natural ionic variations.

Baseline Monitoring Recommendation: Site operators should

implement a 3-step baseline monitoring protocol to avoid false positives due to natural mineral anomalies:

(a) sampling conducted over multiple seasons (at least two hydrological cycles),

(b) using multivariate statistical analysis (e.g., Principal Component Analysis with varimax rotation) to separate natural rock-water interaction signatures from man-made radionuclides sources and

(c) setting site-specific baseline variance thresholds before commercial operation.

6 | Field Case Studies and Baselines

6.1 | Drawdown of Cooling Pond at Chernobyl NPP

During the decommissioning of the Chernobyl NPP, the drawdown of the 22.9 km^2 cooling pond is a huge hydrogeological and ecological problem [5]. A reduction of the water level uncovered a large area of bottom sediments containing cesium-137 (^{137}Cs) and strontium-90 (^{90}Sr). Experimental monitoring of the dried sediment beds showed that the percentage of exchangeable forms of ^{90}Sr rose from 1% of the total inventory to 10% for a 4-year period [5]. This mobility was attributed to the dissolution of calcite and oxidation under aerobic conditions, and it represents a direct danger to regional aquifers through leaching and further transport in the groundwater towards the Pripyat River [5]. Quantitative modelling of these processes was essential to the design of on-site groundwater mitigation barriers [5]. Emergency cooling pond drawdown and historical releases led to contamination of shallow alluvial aquifers [11]. To help reduce this chemical mobilization and to prevent the leaching of ^{90}Sr to regional aquifers, permeable reactive barriers (PRBs) containing natural and synthetic zeolites, calcium phosphate/apatite amendments, and high-capacity clay minerals (e.g., bentonite) are recommended to capture mobile forms of strontium and cesium [11].

6.2 | Tritium Dynamics in the Vltava River Basin (Temelín NPP)

The Vltava River Basin (2002-2007) was extensively analyzed with regard to the dynamics of tritium to assess the correlation between river flow and the concentration of tritium discharged from the Temelín NPP [1]. The annual corrected tritium activity $A_{3H,\,cor,\,j}$ (Bq) is calculated as the difference between the total discharge and background activity, using the river flow rate as follows [1]:

$$A_{3_{H,cor,j}} = C_{3_{H,j}} \cdot Q_j \cdot t - C_{3_{H,B,j}} \cdot Q_j \cdot t \quad \textbf{(16)}$$

where $C_{3H,\,j}$ is the mean annual tritium concentration in year j (Bq/m^3), $C_{3H,\,B,\,j}$ is the background tritium concentration (Bq/m^3), Q_j is the mean annual river flow rate in year j (m^3/s), and t is the duration of a year in seconds [1]. This quantitative relationship helps to define long-term discharge parameters and to check surface-water to groundwater interaction regimes.

6.3 | Krško NPP Tritium Tracer Study

Tritium routinely discharged during operations is an excellent environmental tracer to assess aquifer vulnerability and groundwater travel times. Monitoring of tritium (3H) released to the Sava River from the Krško NPP was carried out in the Zagreb aquifer through an array of piezometers [11]. The Petruševec and Kosnica well fields had well-defined peaks of tritium activity during high-river stages (198.7 ± 20.9 TU and 168.6 ± 20.9 TU, respectively) [11]. These peaks arrived at different times (2.5 to 5.5 months after, depending on the well's distance from the river), and provided direct evidence of hydraulic connectivity and recharge of the unconfined gravel aquifer [11]. This tracer study demonstrates the value of NPP released isotopes for validation of hydrogeological transport models.

6.4 | Sanmen NPP Operational Baseline

A long-term multi-year study was conducted around the Sanmen NPP in China before and after the commissioning of the NPP (2014 to 2017 and 2018 to 2021, respectively) to assess the environmental radioactivity and public health indices [12, 13]. Routine analyses of raw water, factory water and tap water revealed that gross α radioactivity concentration in drinking water was 0.009 ± 0.003 Bq/L and gross β radioactivity concentration was 0.039 ± 0.010 Bq/L, which were within the natural background levels, and statistical analysis showed no monotonic temporal trend (Mann-Kendall test, $p > 0.05$) [12]. In addition, no statistically significant differences in the cumulative ambient doses (CAD) and the regional cancer incidence rates (including leukemia and thyroid cancer) were found before and after NPP operations (Mann-Whitney U test, $p > 0.05$) [12, 13]. These results validate the operational safety containment of the plant.

7 | Challenges and Mitigation Engineering

Modern plants have greatly diminished the likelihood of release events, but tritium migration is still a daunting engineering challenge. Tritium is chemically bound to water molecules as tritiated water (3H1HO or 3H2O) and thus has the same properties as normal water and cannot be filtered or chemically precipitated [11]. The isotopic adsorption and physical retention of tritiated water on clay mineral adsorbents have been recently assessed in geochemical research [14]. The static adsorption experiments show that the natural clay minerals such as montmorillonite, sepiolite, saponite, palygorskite and clinoptilolite have different properties in absorbing water in their interlayer structure [14].

Environmental engineers are working on permeable reactive barriers (PRBs) that use these saponite-zeolite and bentonite-zeolite composites to slow tritium migration in groundwater and provide a potentially engineered barrier to prevent migration of tritium into drinking water aquifers near nuclear facilities [14].

Further, remote-sensing parameters like Net Primary Productivity (NPP) and Normalized Difference Vegetation Index (NDVI) can be used to monitor the ecological effects in the region around nuclear sites. Planners can build multi-year baseline trends of forest vegetation productivity and soil moisture using satellite derived data (e.g. MODIS and Landsat) in models like the Carnegie-Ames-Stanford Approach (CASA). The regional environmental surveillance around nuclear facilities is based on integrated multi-source satellite remote sensing while the ecological modeling is useful in distinguishing localized environmental stressors from those that are more broadly climate-driven, thus providing a comprehensive assessment of the NPP's environmental footprint. Multi-spectral sensors (such as MODIS, Landsat) can be used to monitor the dynamics of vegetation health, soil moisture and land surface temperature in the vicinity of cooling reservoirs and thermal discharge areas on a long-term basis. Ecohydrological models (e.g., the Carnegie-Ames-Stanford Approach (CASA)) are used to calculate forest Net Primary Productivity (NPP) to monitor the vigor of the regional ecosystem, evapotranspiration rates, and seasonal variations in recharge around nuclear facilities. Ecohydrological productivity indicators can be used in conjunction with subsurface modelling to improve early detection of changes in shallow water table and localized environmental stress [15].

8 | Conclusions

Protecting groundwater in the operational phase of nuclear power plants requires a multi-disciplinary approach. It is important to establish high-resolution hydrogeochemical baselines, such as with the CCME WQI, to ensure that any hydrogeochemical anomalies are not mistakenly blamed on operation leaks. Additionally, numerical finite difference flow modeling and GIS-based DRASTIC vulnerability mapping offer valuable predictive tools to define subsurface pathways, to optimize groundwater monitoring networks, and to design reactive barriers. Future monitoring programmes need to be integrated with real-time ambient dose monitoring to provide the necessary confidence in the long-term sustainability of local freshwater resources, and with comprehensive radiochemical analysis of tritium and other mobile fission products.

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It does not contain any experimental studies involving human participants or animal subjects performed by any of the authors.

Conflicts of Interest

The authors declare no conflicts of interest.

Authors Contribution

The first author and second author have equal contribution in the research work. All authors read and approved the final manuscript.

Data Availability Statement

The data that support the findings of this study are available on request from the corresponding author.