FACTORS AFFECTING SUBSURFACE RADIONUCLIDE MIGRATION

COMPLEX NETWORK THEORETICAL APPROACH TO INVESTIGATE THE INTERDEPENDENCE BETWEEN FACTORS AFFECTING SUBSURFACE RADIONUCLIDE MIGRATION

By

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Lay Abstract

Waste products from uranium mining and milling operations are called uranium mine tailings, which are radioactive. Generally, uranium mine tailings are disposed of and isolated in dam-like structures referred to as uranium mine tailings dams (UMTD). One of the most common causes of UMTD failure is the extreme weather condition. When a UMTD fails, a part of tailings, consisting of radionuclides uranium, thorium, and radium, infiltrate into the subsurface through the vadose zone. Radionuclide behavior and transport in the subsurface is influenced by several environmental factors. The objective of the present study is to understand the factors affecting radionuclide migration by *i*) conducting a scoping review on radionuclide migration in the subsurface to describe the factors studied in the literature, and *ii*) understanding and analyzing any relation among the factors and deriving the most dominant factors based on their relation. This study can be used further to develop accurate and reliable radionuclide fate and transport models with minimal uncertainty.

Abstract

Mining of uranium ore and its extraction using the milling process generates solid and liquid waste, commonly termed uranium mine tailings. Uranium mine tailings is radioactive, as it consists of residual uranium, thorium, and radium, which amounts to 85% of the original ore's radioactivity. Due to the extensively long half-lives of uranium (4.5×10^9 years), thorium (75,400 years), and radium (1,620 years) and their harmful radioactive, it is imperative to isolate uranium mine tailings from the environment for a longer period. Containment of uranium mine tailings in dam-like structures, called uranium mine tailings dam (UMTD), is the most followed disposal and storage method. Like a conventional water retention dam, UMTDs are also susceptible to failure, mainly due to adverse weather conditions. Once the UMTD fails, a fraction of the radioactive tailings infiltrates and migrate through the vadose zone contaminating the groundwater sources underlying it. Radionuclide behavior and migration in the subsurface are affected by several environmental factors. To minimize the uncertainty and improve current radionuclide fate and transport models, it is vital to study these factors and any interdependence existing between them. This study aims to understand these environmental factors by *i*) enlisting the factors affecting subsurface radionuclide migration through scoping review of articles and reports, and *ii*) analyzing the interdependence existing between the factors using the complex network theory (CNT) approach and identifying the dominant factors among them. Factors such as chemical and biological characteristics of soil stratigraphy, groundwater, and radioactive tailings plume, meteorological, and hydrogeological are found to influence radionuclide behavior and transport mechanisms in the vadose zone. CNT approach described soil microorganisms, fraction of organic carbon, infiltration rate of the soil, transmissivity, clay fraction in the soil, particulates in groundwater, and infiltrating rainwater as dominant factors in the NoF based on their centrality

measures and sensitivity analysis of the network of factors (NoF). Any uncertainty associated with these factors will affect and propagate through the model. Hence, sufficient resources should be directed in the future to characterize these factors and minimize their uncertainty, which will lead to developing reliable fate and transport models for radionuclides.

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Firstly, I want to express my sincere gratitude to my supervisor Dr. Sarah Dickson-Anderson for her extraordinary support and faith in me throughout my journey at McMaster. Her constant encouragement has always helped me to keep up my morale during the toughest times. Working with her was a great and enlightening experience for me. I am also grateful to my co-supervisor Dr. Peijun Guo and my committee members Dr. Wael El-Dakhakhni, and Dr. Shinya Nagasaki for their constructive feedback and guidance. It was an excellent experience working on a complex network theory paper with Dr. Mohamed Ezzeldin and Dr. Ahmed Yosri and I would like to thank them for their support and guidance. Special thanks to Monica Han for helping me during the lab sessions. I would like to thank Dr. Schuster-Wallace and my peers and colleagues: Katie White, Mohamed Khafagy, Zoha Anjum, and Kayla Lucier for their feedback during research group sessions.

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Declaration of Academic Achievement

This dissertation was prepared according to the guidelines for preparing a sandwich thesis presented by the School of Graduate Studies at McMaster University. Chapter 2 includes a scoping review study that is being prepared to be published as a journal article. Chapter 3 includes a research paper that was already published. Contributions of the author to this dissertation are outlined below:

Chapter 2: Narayanan, B. L., & Dickson-Anderson, S. **Current state of knowledge on the radionuclide transport in the vadose zone: a scoping review.** In-writing.

This paper was initiated by Brinda Lakshmi Narayanan and Dr. Sarah Dickson-Anderson. All aspects of conceptualization and methodology were discussed with Dr. Sarah Dickson-Anderson. Database search, literature collection, and screening process were carried out by Brinda Lakshmi Narayanan. The original draft was prepared by Brinda Lakshmi Narayanan, which was further reviewed and edited by Dr. Sarah Dickson-Anderson. This part is included as a literature section in the dissertation to identify possible research gaps in the literature and list the factors affecting radionuclide migration in the vadose zone.

Chapter 3: Narayanan, B. L., Yosri, A., Ezzeldin, M., El-Dakhakhni, W., & Dickson-Anderson, S. 2021. **A complex network theoretic approach for interdependence investigation: An application to radionuclide behavior in the subsurface**. *Computers & Geosciences*, *157*, 104913.

This paper was initiated and formulated by Brinda Lakshmi Narayanan as a part of a course CIV ENG 704. Key aspects of conceptualization were discussed with Dr. Sarah Dickson-Anderson, Dr. Wael El-Dakhakhni, and Dr. Mohamed Ezzeldin at all stages. Formal analysis was performed by

Brinda Lakshmi Narayanan with the help of Dr. Mohamed Ezzeldin. Sensitivity analysis and software coding was performed by Brinda Lakshmi Narayanan. The original manuscript was prepared by Brinda Lakshmi Narayanan. The manuscript was reviewed and edited by Dr. Sarah Dickson-Anderson, Dr. Wael El-Dakhakhni, Dr. Mohamed Ezzeldin, and Dr. Ahmed Yosri. Dr. Ahmed Yosri and Dr. Sarah Dickson-Anderson aided Brinda Lakshmi Narayanan in addressing editorial comments and modifying the manuscript based on the reviewer's comments. This work should be included in the dissertation as it describes the application of the complex network theory approach to identify dominant factors affecting radionuclide fate and transport models and the interdependence existing between them.

1. Introduction

Uranium radioisotopes are utilized for both military and civilian purposes. Power generation is one of the major civilian applications for uranium isotopes. Uranium is extracted from uranium ore minerals mined from the earth. Canada is one of the top uranium producers in the world and was the largest until 2009 (World Nuclear Association, 2021). Mining operations in Canada started in 1955 in the Elliot Lake region in northern Ontario. However, the discovery of high-grade uranium in Saskatchewan (Campbell et al., 2015), together with an increase in the supply and demand ratio for uranium, resulted in the initiation of closure operations for the Quirke and Panel mines in 1990 and Stanleigh mine in 1996, all located in the Elliot Lake region. Currently, all operating uranium mines in Canada are in Saskatchewan (Canadian Nuclear Safety Commission, 2015).

After mining, uranium is extracted from its ore through a milling process, which generates a solid waste (consisting of crushed rock) and a liquid waste that is commonly referred to as uranium mine tailings (containing processing chemicals and radioactive elements, or radionuclides) (Kossoff et al., 2014). During the milling process, 15% of the radioactivity in the ore will be extracted and produced into a yellow cake, while the rest will be retained in the uranium mine tailings. After the short-lived radionuclides have decayed, 85% of the ore's initial radioactivity will remain in the tailings due to the presence of long-lived radionuclides such as residual uranium-238, thorium-230, and radium-226 with half-lives of $4.5x10^9$, 75,400, and 1,620 years, respectively (Cotta et al., 2020; Hart, 2015). Subsequently, it is imperative to isolate tailings from the environment to contain the radioactivity for hundreds, to hundreds of thousands, of years. Methods for containing radioactive uranium mine tailings include disposal in impoundment structures, backfilling into underground open-pit mines, and deep marine disposal. Among these disposal methods, impoundment structures such as dams, commonly referred to as uranium mine tailings dams (UMTD), is the most widely applied (Office for the London Convention and Protocol and Ocean Affairs, 2013).

Ore mining and milling operations for all metals have led to the construction of over 3500 tailings dams around the world (Lyu et al., 2019). Like any other human-made structure, tailings dams are susceptible to damage and failure. Twenty-one UMTD failures have been reported globally since 1954 (World Information Service on Energy, 2022), with an average of three tailings dam failures per year since 2010. Heavy rainfall has been listed as a significant cause of tailings dam failures (Lyu et al., 2019). This can be attributed to rapidly evolving climatic conditions, the distance of the tailings dam from the nearest sea, and the location of the tailings dam in equatorial regions (Azam and Li, 2010). Heavy rainfall may induce seepage, overtopping (Lyu et al., 2019), or slope instability and erosion (Chambers and Highman, 2011), causing UMTDs to fail. Specifically, as the elevation of the tailings increases over time, the hydrostatic and hydrodynamic pressures on the dam also increase (Gui and He, 2021). This results in increased seepage and dam erosion, and ultimately facilitates uranium mine tailings infiltration through the vadose zone, and potentially into the underlying groundwater (Nair et al., 2013). Transport of radionuclides is less consistent in the vadose zone when compared to the saturated zone due to the presence of multiple phases in the vadose zone (i.e., air and water) (Ozutsumi et al., 2020). As radionuclide transport primarily occurs through water, the vadose zone can act as a natural barrier for migrating radionuclides protecting groundwater resources from contamination (Testoni et al., 2017). Modeling radionuclide transport through the vadose zone is comparatively more complex and requires high computational resources. This can be attributed to the difficulty in characterizing hydraulic information in the vadose zone, as well as the fact that its properties are constantly altering due to recharge and drainage events (Huo et al., 2013). Hence it is crucial to have a thorough understanding of radionuclide behavior in the vadose zone.

Journal articles and reports published over the past decade (i.e., from 2010 to the present) investigating radionuclide migration in the vadose zone indicate different factors influencing their fate and transport. The chemical characteristics of the soil (e.g., Rumiyin et al., 2016), groundwater (e.g., Weaver et al., 2020), and radioactive plume (e.g., Cotta et al., 2020) play vital roles in radionuclide behavior and migration. Microorganisms and organic matter present in the soil (Maloubier et al., 2020) and groundwater (Zhao et al., 2011) influence the valence state of radionuclides, thereby affecting their migration behavior. Extreme rainfall is one of the causes of UMTD failure. Infiltrating rainfall also accelerates the migration and distribution of radionuclides in the vadose zone (Mohanty et al., 2014). Hydrogeologic parameters of the site such as minerals (Maher et al., 2013), clay fraction in the soil (Cheng and Saiers, 2015), degree of saturation (Jakimavičiūtė-Maselienė et al., 2016), infiltration rate (Nair et al., 2013), presence of fractures (Cadini et al., 2013), preferential flow pathways (Ebel and Nimmo, 2013), and hydraulic conductivity (Pannecoucke et al., 2019) also play a crucial role in radionuclide migration in the subsurface. The above-stated factors co-exist by dependency in nature. A thorough understanding of each factor’s influence on radionuclide migration can only be obtained by understanding their interdependency. So far, there has been very limited research to identify any interdependence present among these factors.

The objective of this study is to *i*) conduct a scoping review to assess the current state of knowledge regarding radionuclide fate and transport in the vadose zone, and *ii*) analyze the interdependence between the factors influencing radionuclide transport in the vadose zone to determine the criticality of the interdependence and which factors are dominant.

This thesis contains three chapters in addition to this introduction (Chapter 1). Chapter 2 presents the current state of knowledge regarding the factors influencing radionuclide fate and transport in the vadose zone and identifies research gaps and needs. Chapter 3 uses complex network theory (CNT) to investigate the interdependence among the factors influencing radionuclide behavior in the vadose zone. Chapter 4 presents the conclusions, discusses the limitations of this work, and provides recommendations for future work.

2. Literature review

2.1 Introduction

The failure of UMTDs results in the release of substantial amounts of radioactive mine tailings into the environment. The consequences of UMTD failure under extreme conditions and the resulting impact on nearby communities need to be evaluated carefully to implement proper management strategies and prepare for preventive and restoration measures for both operating and decommissioned UMTDs (Canadian Environmental Assessment Agency, 1996). Records regarding the extent and persistence of groundwater quality degradation underlying areas flooded as a result of UMTD failures have been reported by Kossoff et al. (2014), which are more severe in the absence of remedial actions. The radioactive contaminant plume from the tailings initially migrates through the vadose zone in the subsurface (Zhang et al., 2021) as it migrates to the saturated zone. A scoping review was conducted as an initial step towards understanding radionuclide transport in the vadose zone. This chapter aims to assess the current state of knowledge and identify key concepts and research gaps regarding radionuclide transport in the unsaturated zone and is based on the research question "*What is the current state of knowledge regarding radionuclide behavior and transport in the vadose zone?*".

2.2 Methodology

The review was based on the framework proposed by Arksey and O'Malley (2005) and incorporates modifications suggested by others (e.g., Daudt et al., 2013; Davis et al., 2009; Levac et al., 2010). The six basic stages of the review include (Fig. 1 (Pham et al., 2014)): 1) formulating a research question; 2) identifying relevant studies; 3) screening studies relevant to the research question; 4) characterizing data from literature; 5) summarizing results; and 6) consultation exercise (Arksey and O'Malley, 2005).

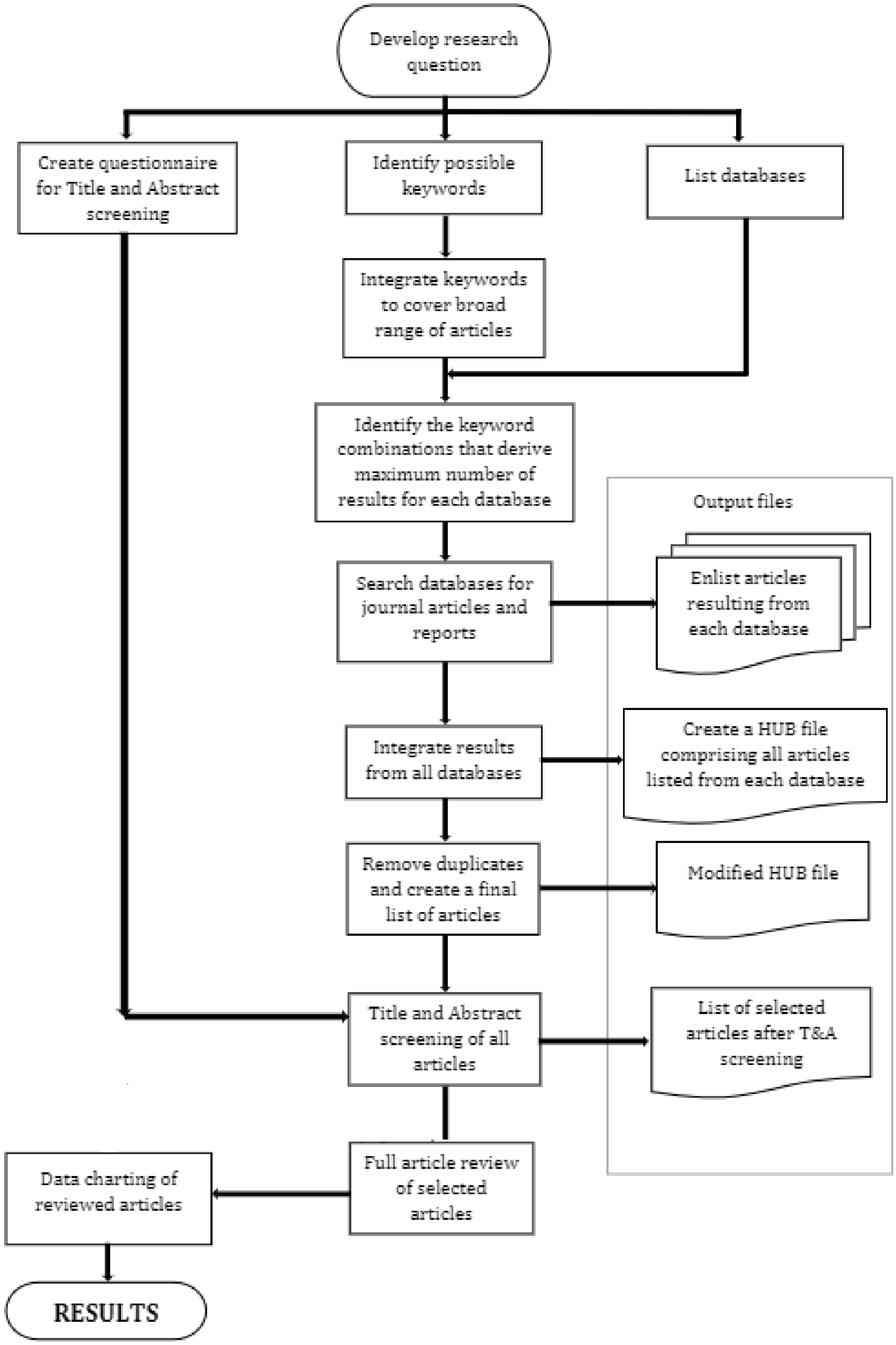


Fig. 2-1. Flowchart of stages followed in scoping review

The databases employed, and the keywords used within each database, are presented in Table 1. Journal articles and reports published in English between January 2010 and August 2021 (inclusive) were included in the review.

Table 2-1. Databases and keyword syntaxes used for literature search

	Database									
Keyword syntax	International Nuclear Information System	Scholars Portal	Engineering Village	Scopus	Web of Science	JSTOR Sustainability	GreenFile	ProQuest	ASCE's Civil Engineering	National Academies Press
“radionuclides” OR “radioisotopes” AND “unsaturated zone”	✓	✓							✓	✓
"radionuclides" OR "radioisotopes" AND "unsaturated zone" OR "vadose zone"			✓	✓	✓	✓	✓	✓		
"radionuclides" AND "unsaturated zone" AND "transport" OR "retardation"	✓								✓	
"radionuclides" OR "radioisotopes" OR “radioactive elements” AND "unsaturated zone" OR "vadose zone" AND "transport" OR "migration" OR "retardation"	✓	✓	✓	✓	✓	✓	✓	✓		
"radionuclides" OR "radioisotopes" OR "uranium" OR "thorium" OR "radium" OR "strontium" OR "plutonium" OR “cesium” AND "unsaturated zone" OR "vadose zone" AND "transport" OR "migration" OR "retardation"	✓	✓	✓	✓		✓	✓	✓		
"radionuclides" OR "radioisotopes" OR "uranium" OR "thorium" OR "radium" OR "strontium" OR "plutonium" OR "cesium" AND "unsaturated zone" OR "vadose zone" AND "transport" OR "migration" OR "retardation" OR "mobilization" OR “infiltration” OR "Seepage"	✓			✓	✓					
"radionuclides" OR "radioisotopes" OR "uranium" OR "thorium" OR "radium" OR "strontium" OR "plutonium" OR "cesium" AND "unsaturated zone" OR "vadose zone" OR "unsaturated porous medium" OR "unsaturated fractured medium" AND "transport" OR "migration" OR "retardation" OR "mobilization" OR "infiltration" OR "seepage" OR "movement"		✓	✓		✓		✓	✓		

The initial search returned 1652 articles and reports, which were subject to an initial screening based on title and abstract. The screening questionnaire was designed to select articles and reports focusing on radionuclide transport in the vadose zone (see Appendix T1). Articles and reports covering both the unsaturated and saturated zones, as well as those that did not contain enough information to be able to assess inclusion criteria, were included at this stage. Following this stage, 243 articles remained (i.e., 1,409 articles and reports were excluded in this stage). The remaining 243 articles were subject to a full-text review. Articles for which a full-text version was not accessible, that did not include both radionuclides and the unsaturated zone in their study, or that addressed radionuclide transport in the gaseous phase alone, were excluded at this stage. A total of 129 articles and four reports remained following this stage (i.e., 110 articles and reports were excluded). Relevant data were extracted from the remaining articles, charted using Excel, and subsequently sorted and interpreted. The information charted (See Appendix T2) included the year of publication, type of article/report, methodology, relevant subsurface zone, type of medium, and external factors and mechanisms. Discussion of the charted data is presented in the next section.

2.3 Results and Discussion

Within the study period, the largest number of studies was published in 2010 (n[a]=21) and the smallest number in 2016 and 2017 (n=6), respectively. Of the 129 articles included, 115 were primary research, and 14 were review articles. A keyword analysis was conducted to aid in identifying fundamental information from the articles included. Where keywords were not available, five to six keywords were assigned based on the title and abstract review. The keyword analysis identified the major sites at which subsurface radionuclide migration was studied. Most of the literature was based on the Hanford, Savannah River, and Goiania waste repository sites, all of which are radioactive waste management and storage facilities. Radionuclides leaching into the subsurface from waste storage and disposal sites is a widely discussed topic in the literature.

[a] n includes the number of articles and reports

Nuclear weapon test sites (e.g., Nevada test site) and the sites of nuclear power plant accidents (i.e., Chernobyl and Fukushima) have also been used to study radionuclide behavior in the environment. Other studies focussed on radionuclide release from proposed deep geological repositories (DGRs) (e.g., Yucca Mountain, Sierra Pena Blanca, Mexico).

Fig. 2 classifies the journal articles according to the field of study and shows the largest number of articles published in journals related to hydrogeology, geochemistry, radionuclide contamination, and environmental science. Modeling-based studies were published in journals focused on computational mathematics.

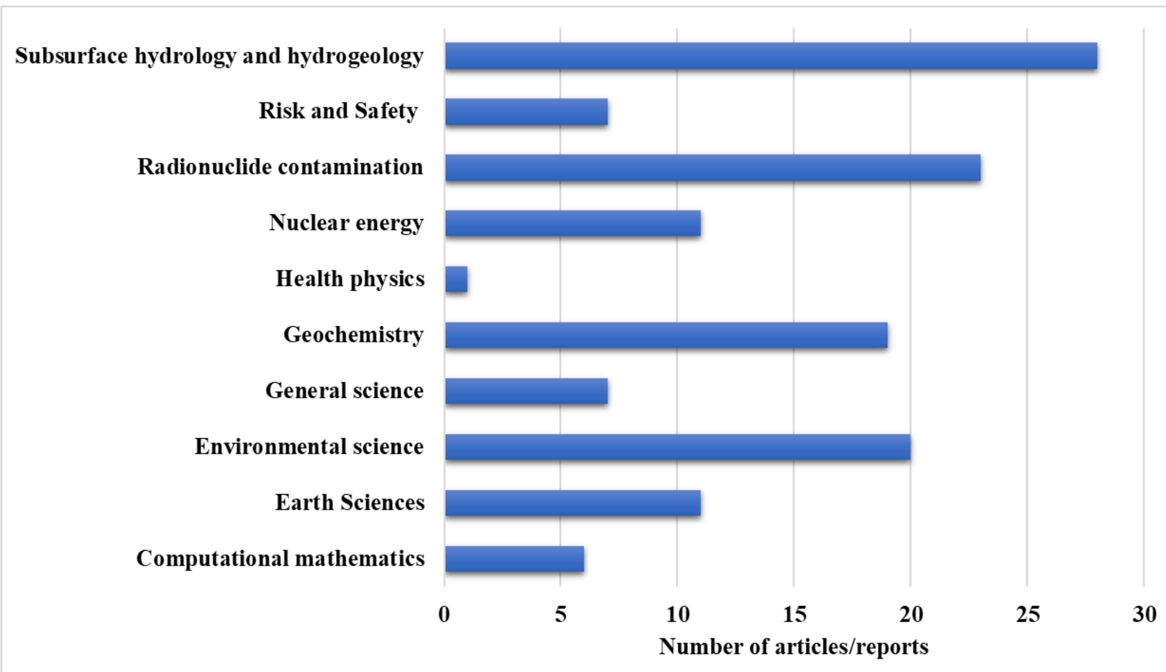


Fig. 2-2. Field of journal publications

The research articles returned were classified according to methodology, subsurface zone, and stratigraphy as shown in Fig.3. The studies predominantly apply either experimental (45) or modeling (46) approaches to understand radionuclide transport in the unsaturated zone. Only a few studies (i.e., 23 out of 117) utilized both experimental and modeling methods in a single paper. Meta-analyses were applied in three articles by Ramirez-Guinart et al. (2020a; 2020b; 2020c) to analyze sorption distribution coefficient data (K_d) available for uranium, cesium, and americium to reduce the variability present in the K_d values. Improving the understanding of radionuclide transport through both the vadose and saturated zones together has become an increasing focus over the past decade. These studies typically apply the same stratigraphy for both subsurface zones. Only one article (Dam et al., 2015), studied a mixed stratigraphy, with soil sediments in the vadose zone and fractured rock in the saturated zone. Few studies included a buffer medium (bentonite clay - an engineering barrier) as the vadose zone in their study (Chang et al., 2021; Samper et al., 2011; Zhang and Zhang, 2010).

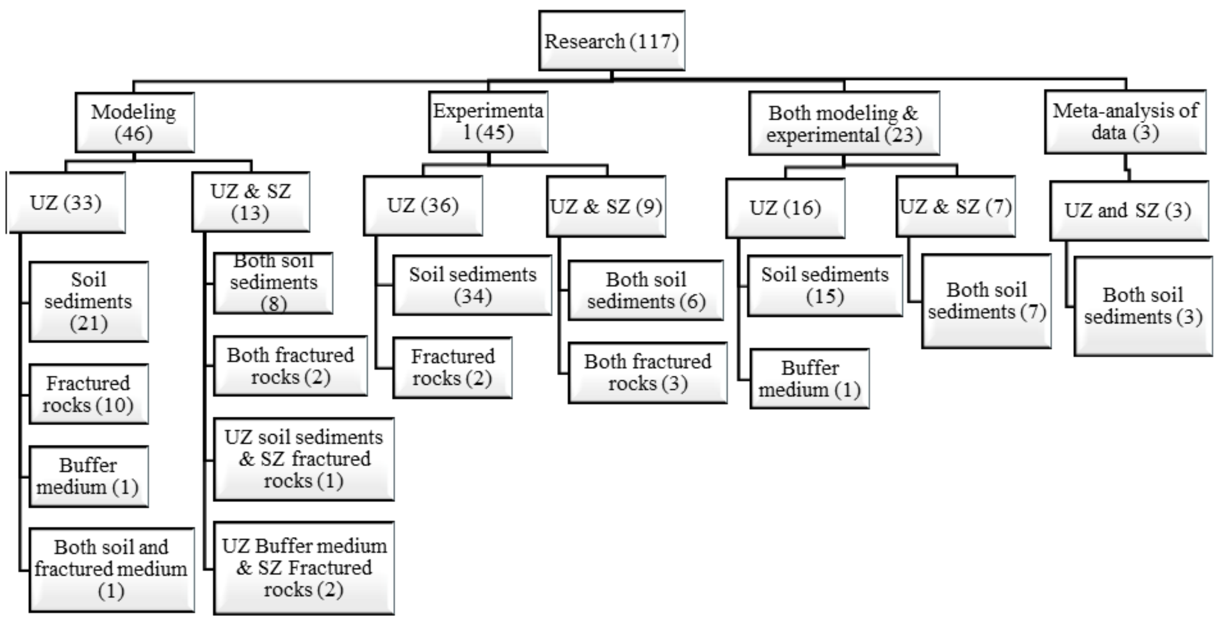


Fig. 2-3. Research articles classified based on methodology, zone, and stratigraphy
Note: Number of research articles under each category is presented inside the parentheses.
UZ – unsaturated/vadose zone, SZ – saturated zone.

The relative frequency with which each radionuclide is studied, and the methodology used to study each are presented in Fig. 4. Some papers collectively include most radionuclides under the term ‘radionuclides’ in their study. Uranium is predominantly studied in all types of research studies (i.e., experimental, modeling, both experimental and modeling, meta-analyses), especially in the context of release from waste storage sites (Vazquez-Ortega et al., 2021; Paradis et al., 2020;

Perdrial et al., 2018; Dam et al., 2015). The subsurface fate and transport of some radionuclides such as cesium, plutonium, and strontium have been investigated using the modeling and experimental techniques. These long-lived radionuclides are highlighted in the literature as high-level radioactive waste leakage from several nuclear waste repositories, including Hanford's underground storage facilities (Cao et al., 2017). Cesium, plutonium, and strontium are also linked with studies regarding the Chernobyl event (Bugai et al., 2012). Cesium, plutonium, radioiodine, strontium, and uranium are the most studied radionuclides using experimental techniques; however, experimental research studies typically focus on a single radionuclide at a time. Modeling techniques were predominantly used to study radionuclides, including tritium. Cesium, strontium, and uranium (Berns et al., 2018; Coutelot et al., 2018) had the largest number of studies that used a combination of modeling and experimental techniques.

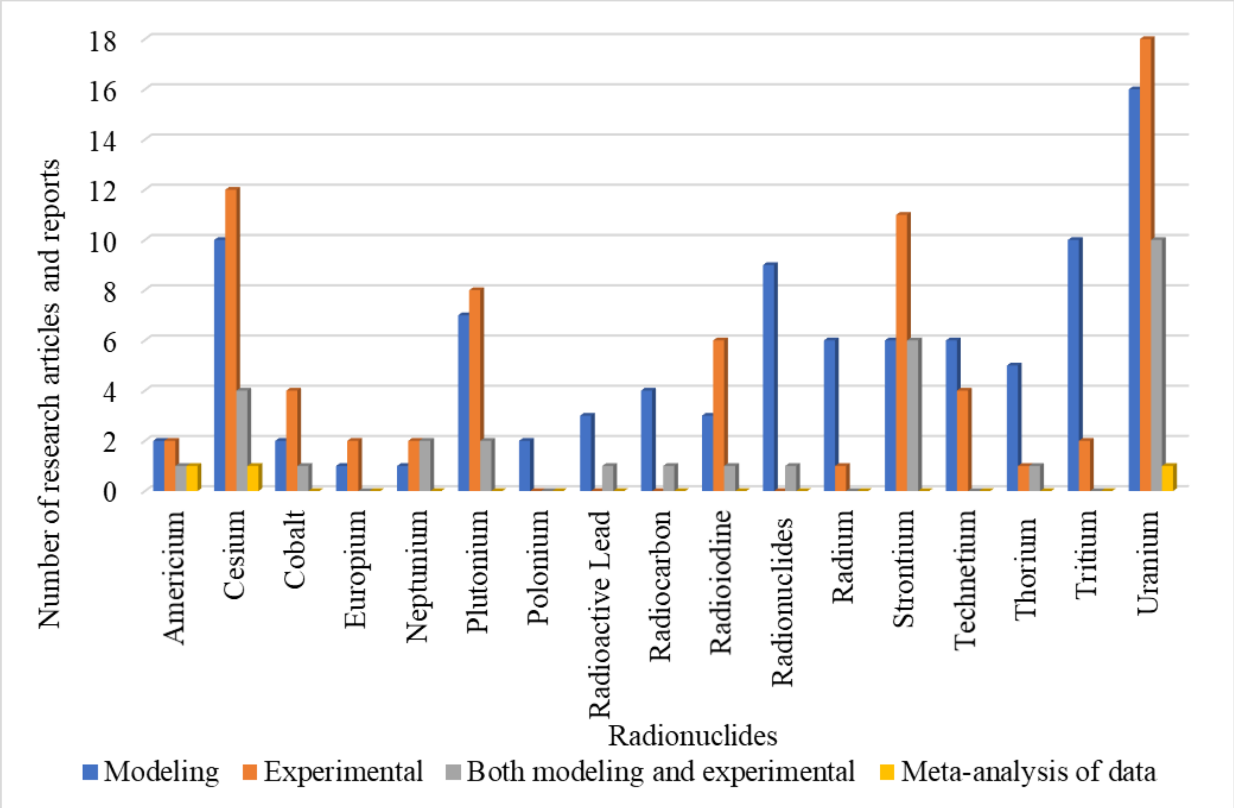


Fig. 2-4. Radionuclides studied in research articles and reports

Figure 5 presents information on research studies following different experimental approaches for different radionuclides. Research following experimental methods significantly applied laboratory-based experiments (40) compared to field experiments (4). Maloubier et al. (2020) was the only research article that used both field and laboratory experiments to study the effect of natural organic matter on plutonium migration under vadose conditions. Adsorption/desorption, a commonly discussed mechanism in the extracted literature, was mainly studied using lab-based experiments, especially using a combination of flow-through column and batch experiments (for example, Kim et al., 2019; Szecsody et al., 2014; Um et al., 2010). Field experiments were mostly carried out using a field-scale lysimeter (Maloubier et al., 2020; Erdmann et al., 2018; Peruski et al., 2018; Liu et al., 2013). Paradis et al. (2020) used single- and multi-well injection-extraction field tests to understand uranium mobility in the alluvial aquifer during recharge events.

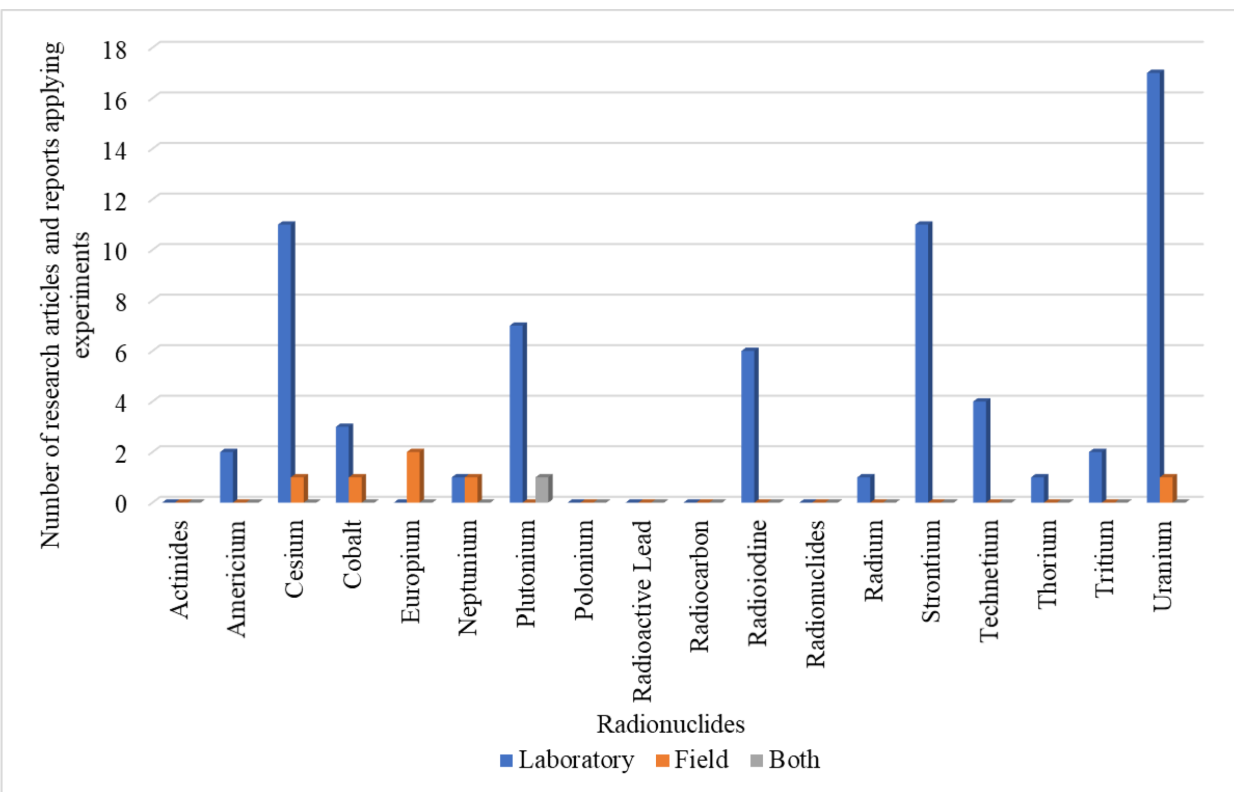


Fig. 2-5. Research studies applying different experimental methodologies for radionuclides

As indicated in fig. 6, the highest number of review articles and reports presented a review on the fate and transport of all radionuclides in general (for example, Medved'and Černý, 2019; Rechard et al., 2014a). Some review studies also studied individual radionuclides such as uranium (Freedman et al., 2017; Golovich et al., 2011), plutonium (Romanchuk et al., 2020), and radioiodine (Moore et al., 2020; Neeway et al., 2019). The migration behavior of radionuclides such as cesium, neptunium, strontium, and thorium, was also reviewed collectively in a few articles and reports (Bugai et al., 2020; Maher et al., 2013). Review articles and reports focussed mainly on modeling techniques followed (Freedman et al., 2017; Rechard et al., 2014a & 2014b; Smith et al., 2014; Payne et al., 2013) and transport behaviors of radionuclides such as speciation (Romanchuk et al., 2020; Maher et al., 2013; Golovich et al., 2011), sorption (Bugai et al., 2020; Rigali et al., 2016; Kersting, 2013; Crawford, 2010), precipitation (Moore et al., 2020; Neeway et al., 2019), reduction (Felmy et al., 2011), and advection and diffusion (Medved'and Černý, 2019) (See Figure 4).

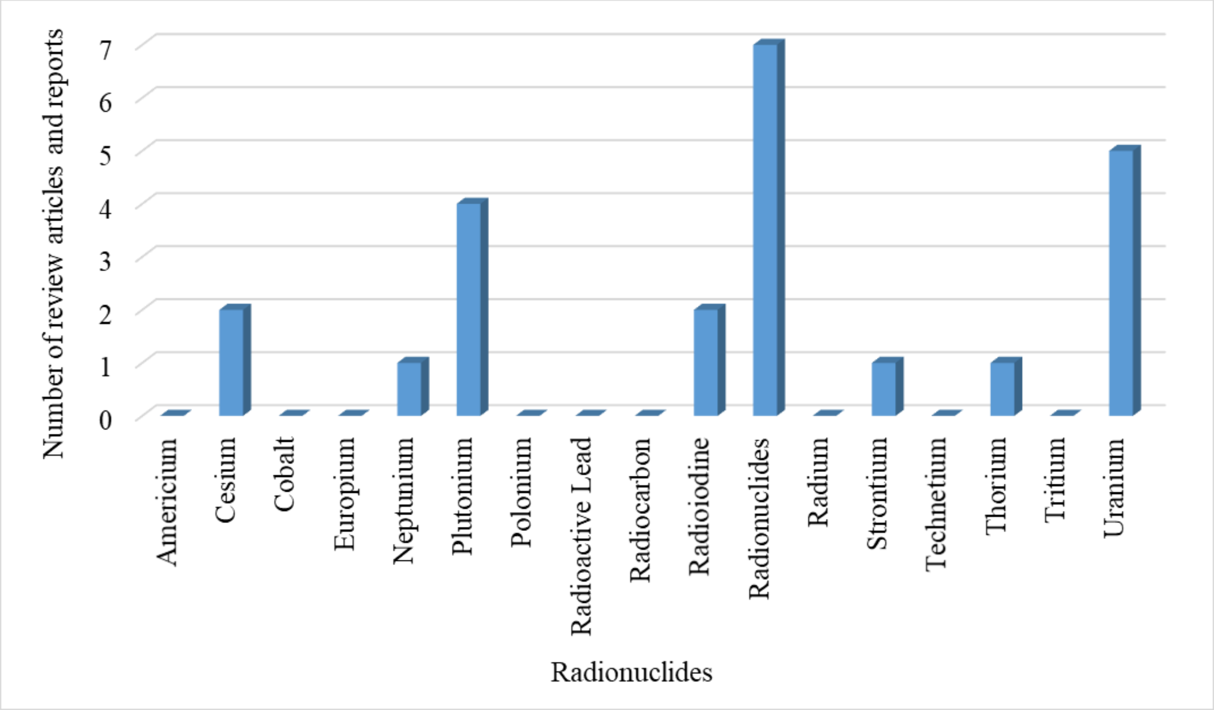


Fig. 2-6. Radionuclides studied in review articles and reports

When radionuclides enter the subsurface, they undergo several transport processes as indicated in Fig. 7. Adsorption/desorption is the most widely studied radionuclide transport mechanism in the literature, with a primary focus on adsorption. Adsorption is the physical attachment or chemical bonding of radionuclides to the surface of other compounds present in the subsurface (Rakesh et al., 2017). The sorption process can be further described by surface complexation and ion exchange reactions. Some studies apply surface complexation models to investigate the interactions between ionic species and the soil-water interface under varying geochemical conditions to determine their impact on radionuclide migration (Coutelot et al., 2018). Few studies focus on the sorption of radionuclides due to ion exchange, which is the exchange of ions of like charge between a solution and a solid (Rakesh et al., 2017). Advection and dispersion were found to be the next most studied radionuclide transport mechanisms. Advective transport is hydraulically driven radionuclide migration and is a widely discussed mechanism in modeling-based research (Ha et al., 2020; Medved and Cerny, 2019). Advective transport is predominant in a water-saturated zone which explains the retardation of strontium in vadose zone conditions (Huo et al., 2013). Water content in the vadose zone is amplified by a high infiltration rate, resulting in solutes migrating more quickly and deeply into the subsurface through the vadose zone during intermittent rainfall events (Mohanadhas and Govindarajan, 2018). Dispersion of radionuclides local-scale molecular diffusion and field-scale macro-dispersion (Suresh Kumar, 2015) is caused by velocity variations and alternate flow paths (Kim et al., 2019). Mineral transformation and altering speciation of radionuclides lead to precipitation of radionuclides, depending on the waste solution and sediment chemistry (Perdrial et al., 2018). Highly acidic radioactive waste solutions cause sediment weathering as they seep into the vadose zone and the presence of phosphate ions results in the precipitation of uranium in the form of meta-ankoleite, a uranyl phosphate mineral

(Wang et al., 2017). Diffusion is observed in low permeability media subjected to thermal gravitation (Setiawan and Ekaningrum, 2019). Matrix diffusion is also a crucial mechanism in fractured rock formations, wherein porous rock matrix adjacent to fracture walls retards radionuclide transport in the fractures (Kim et al., 2019). The matrix porosity and radionuclide diffusion coefficients impact the rate of diffusion into the matrix, promoting the retention of radionuclides. (Cadini et al., 2013; Robinson et al 2012b). Radionuclides such as uranium, neptunium, and plutonium are multi-valent and their mobility is primarily dependent on their current oxidation state in the subsurface (Maher et al., 2013). Oxidation and reduction reactions in the vadose zone can greatly affect the valence state of the radionuclides and further influence their mobility (Peruski et al., 2018).

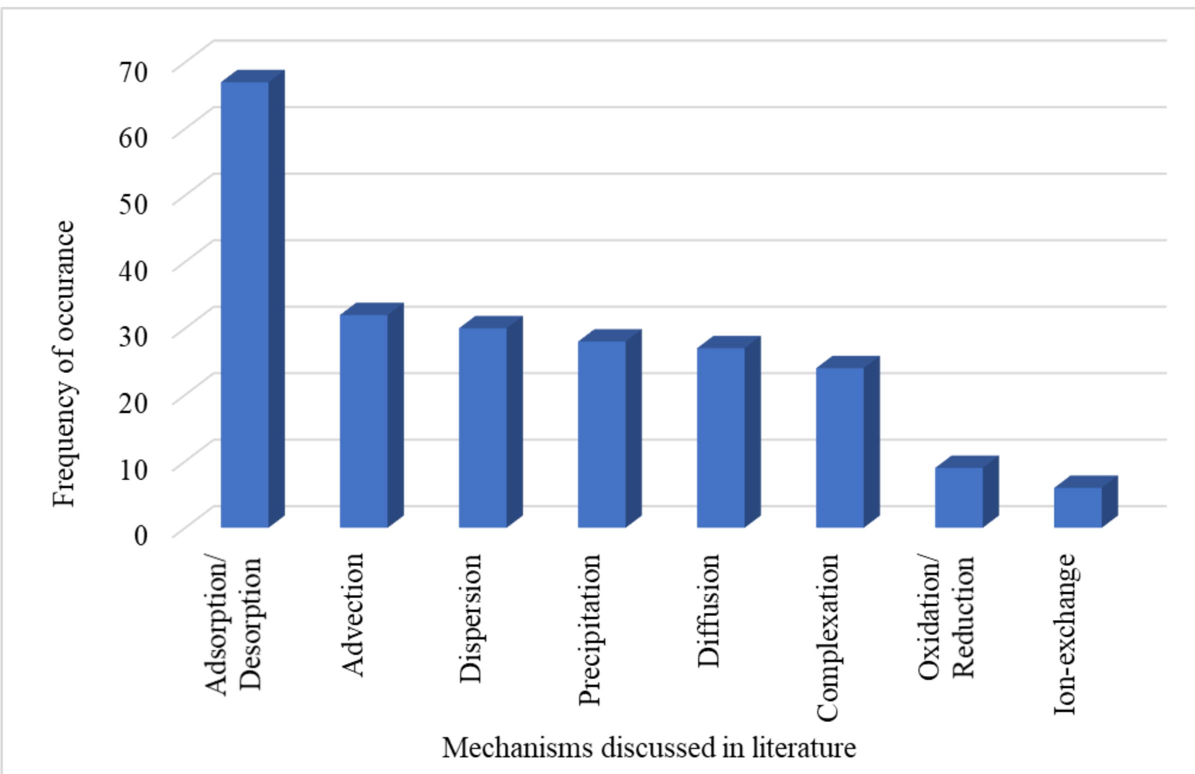


Fig. 2-7. Mechanisms of radionuclide fate and behavior studied in the literature

2.3.1. Factors affecting radionuclide mechanisms

Radionuclide behavior and transport mechanisms in the vadose zone are greatly influenced by factors such as chemical (Xu et al., 2015; Kovacheva et al., 2014; Zhang and Zhang, 2010; Cheng and Saiers, 2010) and biological (Bagwell et al., 2020; Berns et al., 2018) characteristics of the stratigraphic layers, groundwater, and radioactive plume, as well as the meteorological (Peruski et al., 2018) and hydrogeological (Cotta et al., 2020; Ebel and Nimmo, 2013; Grogan et al., 2010) conditions.

2.3.1.1. Chemical characteristics of soil, groundwater, and radioactive plume

Chemical perturbations in the pH and ionic strength of both the groundwater and radioactive contaminant plume can influence the transport of radionuclides differently. For example, high pH values enhance strontium adsorption to quartz sand, and higher ionic strength leads to strontium desorption from soil resulting in its remobilization, irrespective of pH levels (Weaver et al., 2020). Desorption can be attributed to the diminishing of the repulsive energy barrier between likely (negatively) charged plutonium colloid and soil sediments due to high ionic strength. However, while Zuo et al. (2010) observed a similar effect of pH on plutonium, Xie et al. (2013) observed retention of colloidal plutonium even under high ionic strength conditions. The redox potential of groundwater and contaminant plumes flowing through the subsurface affects the valence states of radionuclides, thereby influencing the radionuclide's speciation behavior. Redox speciation of radionuclides influences its migration by governing the sorption and solubility properties (Crawford, 2010). For example, the oxidised form of uranium, U(VI), is soluble while its reduced form, U(IV), is less soluble and comparatively immobile because of sorption to reduced iron- or sulphur-surface species and anaerobic microorganisms in the soil (Coutelot et al., 2018). Similarly, neptunium is more mobile in its oxidised form Np(V) and mostly found in a dissolved state whereas its reduced form, Np(IV), has high sorption affinity and is likely to be retarded by sorption and complexation mechanisms (Peruski et al., 2018).

The cation exchange capacity (CEC) of the clay fraction present in the soil increases the sorption of radionuclides (Rumynin et al., 2016). For example, similar to radionuclides like strontium (Sr^{2+}) and cesium (Cs^{+}), cations (i.e., $NH_4^{+}, K^{+}, Ca^{2+}, Mg^{2+}$) also have a strong adsorption affinity to surfaces of clay particles. Results indicated that among all cations, ion exchange between NH_4^{+} and Sr^{2+} adsorbed to the surfaces of illite, can result in desorption of strontium ions leading to its migration into the subsurface (Baker et al., 2010). In some cases, the presence of organic matter can interfere with the CEC of illite, especially for Cs^{+} ions. Organic matter can hinder the sorption of Cs^{+} to clay mineral by blocking the available sorption sites due to the formation of organo-clay complexes (Berns et al., 2018). Temperature is another crucial factor that impacts processes such as sorption and speciation of radionuclides (Crawford, 2010). Extreme temperature variations (i.e., frozen (-18°C) to dry (33°C)) can cause instability in soil aggregates due to contraction and expansion of particles. Extreme temperature fluctuations in fluvisol soil led to the increased surface area which resulted in high sorption of cesium. Also, sorption of organic matter (i.e., humic substances) on clay particles decreased with temperature fluctuations resulting in more sorption sites available for cesium (Kovacheva et al., 2014). A rise in temperatures from 20°C to 40°C caused a rapid increase in the plutonium sorption capacity of shale rocks (Zuo et al., 2010).

Radionuclides are subject to decay reactions, such that the concentration of progenies will increase for some time as decay progresses. For example, uranium decay results in decreasing uranium concentration over time, while the concentration of its progenies such as thorium, radium, and lead increase over the initial 5000 years (Cotta et al., 2020). Each of these radionuclides will undergo different transport mechanisms, with thorium, radium, and lead subject to more retardation (due to sorption) than uranium, which will migrate faster (Naveira-Cotta et al., 2013).

Radionuclides present in the subsurface can be observed in different sizes (i.e., dissolved (< ~10^{-3} μm), colloidal (~10-3 < x < ~1 μm), particulate (> ~1 μm)) (Kersting, 2013). Radionuclide (i.e., cesium, radioiodine, and strontium) released from waste weathered sediments located in Hanford is elevated by radionuclides in colloidal, and particulate size fractions (Perdrial et al., 2015). Hydrolysis of radionuclides in porewaters can form ‘intrinsic’ colloids, or they can adsorb onto organic or inorganic colloids which are present ubiquitously in the vadose zone and form ‘pseudo’ colloids (Peruski et al., 2018). The transport of colloid-associated radionuclides in fractures is governed by the rate at which the colloids were filtered by fractures and the reaction rate of sorption of radionuclides onto the filtered colloids. As the filtration and sorption rates increase there is significant retardation of colloidal radionuclides in fractures (Reiche et al., 2016). Different radionuclides have varying degrees of affinity for organic or inorganic colloids. Either way, colloid-facilitated contaminant transport enhances the mobility of radionuclides that have low solubility (Cheng and Saiers, 2010). Solute and colloid-facilitated radionuclide migration are discussed commonly in the literature, while studies on particulate-release of radionuclide migration are scarce. High sorption affinity of strontium to zeolites and feldspathoids present in the soil led to the formation of particulate strontium. Elution of particulate strontium was predominantly observed during transient flow conditions (Perdrial et al., 2015).

2.3.1.2. Biological components in soil, groundwater, and radioactive plume

Organic content in the subsurface is a widely discussed factor in the literature. Organic content is present ubiquitously in the subsurface either as a part of the porous media or dissolved in groundwater. Natural organic matter (NOM) present in the soil sediments causes plutonium sorption and retardation by forming ternary complexes between the sediment surface, NOM, and plutonium. In addition to sorption, NOM prevents plutonium migration by stabilizing the reduced form of plutonium (i.e., Pu(IV)) in the soil and inhibits its oxidation to Pu(V) which is significantly mobile in the subsurface environment (Maloubier et al., 2020). Dissolved organic matter (DOM) in groundwater also affects the sorption of radionuclides such as neptunium, americium, plutonium, and uranium, with the influence of DOM varying under different acidic and alkaline conditions (Zhao et al., 2011).

Microorganisms also influence radionuclide behavior. Katsenovich et al. (2013) observed that *Arthrobacter*, found in contaminated Hanford sediments, accumulate uranium on their surfaces by ion exchange and precipitation mechanisms. Microorganisms are also responsible for the reductive immobilization of uranium (Bagwell et al., 2020).

2.3.1.3. Meteorological

The vadose zone is considered to be a natural protective barrier to aquifers due to its ability to promote radionuclide retention (Zhang et al., 2021). Meteorological events such as rainfall can trigger infiltration of rainwater into the subsurface. Percolating rainwater alters the flow conditions in the vadose zone causing remobilization of immobile radionuclides and alters the chemistry of the radioactive plume and groundwater by dilution (Libera et al., 2019). A few studies have investigated the impact of infiltration on physio-chemical changes in the subsurface and the resulting impacts on the migration of tritium (Libera et al., 2019), neptunium (Peruski et al., 2018), cesium, strontium (Mohanty et al., 2014), and europium (Liu et al., 2013).

2.3.1.4. Hydrogeological

Minerals present in the soil affect radionuclide fate and transport. Radionuclides can adsorb onto mineral surfaces comprising iron oxyhydroxides, manganese oxyhydroxide, zeolites, clay minerals, and phosphates (Maher et al., 2013). Although quartz is not an effective radionuclide adsorbent (Maher et al., 2013), quartz coated with iron and manganese hydroxide is found to adsorb radioactive cobalt through surface complexation reactions (Rumynin et al., 2016). Sorption distribution values for strontium, technetium and tritium were observed to be higher in fractured rocks with fillings/coatings of zeolite when compared to fractured rocks without fillings and coatings (Kim et al., 2019). In addition to its low conductivity, phosphate-bearing clay mineral, apatite can adsorb uranium through ion-exchange and surface precipitation mechanisms, thereby indicating its suitability to be incorporated into engineered barriers (Rigali et al., 2016). Among all minerals, the impacts of clay minerals are most discussed in the literature. Clay contributes larger specific surface areas for the sorption of radionuclides compared to other soil types (Ramirez-Guinart et al., 2020a). Each type of clay mineral exhibits a different adsorption efficiency towards radionuclides. While Cheng and Saiers (2015) observed significant cesium adsorption on illite and comparatively negligible on kaolinite, Coutelot et al. (2018) found higher and irreversible adsorption of uranium to kaolinite, especially at pH $\approx$ 5. The texture of soil sediments and rock grains also influence subsurface radionuclide migration as they indicate the specific surface area and the pore volume, which are crucial for sorption and precipitation mechanisms (Ramirez-Guinart et al., 2020b). Extremely slow vertical downward migration of uranium is observed in clayey (Pontedeiro et al., 2010) and silty formations (Mohanadhas and Govindarajan, 2018) when compared to sandy soils. This is due to the lower permeability and higher sorption capacity of clay relative to sand. The impact of texture and grain size was also noted by Zuo et al. (2010), who observed that the sorption distribution coefficient value of plutonium increased significantly with decreasing grain size in shale rock, especially when the grain size is less than 0.2 mm.

2.4 Research gaps

The multitude of factors discussed in this chapter indicates the complexity of radionuclide behavior and transport in the subsurface. This is made even more challenging because many of these factors are interlinked. For example, infiltrating rainfall is a meteorological factor affecting the migration of radionuclides and additionally altering the chemistry of contaminant plumes by dilution (Libera et al., 2019). Another example shows that the influence of DOM on the sorption of radionuclides is affected by varying acidic and alkaline conditions in the subsurface (Zhao et al., 2011). This raises the following concerns: 1) Are there more linkages existing between these factors? 2) Do they have a collective impact on radionuclide migration? 3) What factors have a dominant effect on radionuclide fate and transport? It is imperative to obtain answers to these questions, to develop accurate representative models of subsurface radionuclide migration in the event of UMTD failures. To understand a chaotic interdependence among numerous factors, simple yet clear visualization, and analysis technique should be applied. This research gap is addressed using the complex network theory approach in Chapter 3.

3. A complex network theoretic approach for interdependence investigation: an application to radionuclide behavior in the subsurface

Highlights

- CNT conceptual model for factors influencing fate and transport in the subsurface
- CNT-guided investigation of the importance of and interrelationships among factors
- CNT-based determination of dominant factors propagating model uncertainty
- CNT-guided resource allocation strategy to reduce uncertainty in model

Abstract

The failure of uranium mine tailings dams results in the infiltration and spreading of tailings in the subsurface. The fate and transport of radionuclides in the subsurface depends on several confounding, complex interdependent factors that describe the elements of the integrated system (i.e., meteorological; hydrological; hydrogeological; and, soil, groundwater, and mine tailings chemistry). The factors describing the integrated system have typically been investigated independently; however, their interdependence and resulting collective influence on the subsurface migration of radionuclides are yet to be explored. The current study develops a complex network theoretical approach to analyze these interdependencies. In this respect, a network of factors (NoF) was developed, and its characteristics (e.g., diameter, density, characteristic pathlength, average clustering coefficient, and factor centrality measures) were evaluated to determine the importance of considering these interdependencies when developing radionuclide fate and transport models. A sensitivity analysis was subsequently performed on the NoF to characterize the propagation of uncertainty associated with the factors in the NoF through a fate and transport model. The sensitivity analysis indicated that microorganisms present in the soil and mine tailings, fraction of organic carbon in the soil matrix, infiltration, and transmissivity must be well characterized (i.e.,

to minimize their uncertainty) when developing an integrated subsurface radionuclide fate and transport model, as uncertainty in these parameters will be amplified in the model output. The NoF developed in this study can be used to allocate data collection resources strategically to minimize uncertainty in fate and transport models. This improves the reliability of fate and transport models and ultimately leads to better management and remediation strategies to mitigate impacts from UMTD failures.

Keywords: Uranium mine tailings dam, Radionuclide transport, Centrality measures, interdependencies, Network of Factors, Network sensitivity analysis

3.1 Introduction

Canada is the second-largest uranium producer in the world, contributing approximately 13% of global uranium production (World Nuclear Association, 2020). Uranium is extracted from its ore through a milling process that involves a series of physical and chemical subprocesses (Kossoff et al., 2014). Waste produced from the uranium mining and milling process, termed uranium mine tailings, comprises unextracted uranium left in the ore debris as well as the by-products of uranium-238 decay. Such by-products may include uranium-234, thorium-230, and radium-226, which can retain their radioactivity for thousands of years due to their long half-lives. Therefore, uranium mine tailings are generally classified as low-level radioactive waste (National Research Council, 2012), and are most often contained in surface impoundment structures called uranium mine tailings dams (UMTDs) (International Atomic Energy Agency, 2004). The failure of a UMTD may lead to a catastrophic socioenvironmental crisis due to the release of radionuclides into the environment, which may infiltrate into the subsurface. Radionuclide fate and transport in groundwater systems have been investigated following UMTD failures in Langer Heinrich Uranium Mill, Namibia (Abiye and Shaduka, 2017), Mary Kathleen Uranium Mill, Australia (Lottermoser and Ashley, 2005), and Church Rock Uranium Mill, USA (Millard et al., 1983). These studies showed that the subsurface fate and transport of each type of radionuclide present in the tailings are affected by several common processes.

Understanding the subsurface fate and transport of radionuclides released from UMTDs requires integrating the processes influencing them at the ground surface, across the vadose zone, and within the saturated zone in a single model. However, such integrated models require a significant amount of data encompassing each of the factors that characterize the relevant processes in the system (i.e., each region of the model: ground surface, vadose zone, aquifer). It is imperative to consider the effects of the complex interdependencies between these factors as they coexist in the system, as factors with the most influence will propagate uncertainty throughout the corresponding fate and transport model. As such, an understanding of these interdependencies can help determine which factors need to be characterized most accurately (i.e., how to allocate resources for factor characterization) to minimize uncertainty in model output.

The influence of the factors affecting the subsurface migration of radionuclides have typically been studied assuming that they are independent, with only a few studies investigating the interdependencies between these factors (e.g., Eichholz et al., 1982; Anderson et al., 2007; IAEA, 2013; Wu et al., 2017; Boreham, 2017), albeit only in a pair-wise fashion. One method of analyzing a system of interdependent factors is to use a *complex network theory* (CNT) (Sivakumar, 2015) approach. This approach has been used to identify the dominant components of the hydrologic cycle, where the physical processes (e.g., precipitation, surface runoff, infiltration) were represented by nodes, and their interdependencies were represented by weighted, directed links. The interdependencies were identified based on CNT metrics, including example clustering coefficient, degree distribution, and network centrality measures (Sivakumar, 2015). Martinez et al. (2019) utilized CNT to identify the environment-activity interactions to limit subjectivity and reduce uncertainty in Environmental Impact Assessment (EIA) processes. Agarwal et al. (2020) applied CNT to optimize the design of hydrometric networks by identifying the most critical nodes. These studies, along with others, demonstrate the utility of CNT to analyze the systems of interconnected components/nodes and subsequently identify the most influential nodes.

The focus of this paper is the application of CNT to facilitate the visualization and investigation of the interlinkages between (previously identified) factors influencing radionuclide fate and transport in an integrated subsurface system model. The understanding of linkages between these factors will facilitate the identification of the most dominant factors, as well as the interdependencies between them. This understanding can help to inform efficient resource allocation programs to collect specific data required to construct integrated subsurface models while minimizing uncertainty. This is critical, as radionuclide fate and transport models are an important tool for groundwater planning, management, and remediation strategies—all of which are needed to protect the health of those who depend on groundwater for their potable water supply.

3.2 Methods

3.2.1. Factors Influencing Radionuclide Transport in Subsurface Environments

Factors influencing radionuclide fate and transport were identified based on knowledge of these processes (e.g., Fetter, 2001; Subramanya, 2008), and supplemented based on a review of the literature. Relevant peer-reviewed journal articles published in English between 1980 and 2020 were identified through a search of all databases within Engineering Village and augmented via reference mining and with grey literature identified through Google Scholar. Combinations and derivatives of the following keywords were searched: *radionuclides, uranium, radium thorium, subsurface, unsaturated zone, saturated zone, fracture, porous medium, transport, behavior, environmental factors, hydrogeology, climate, mine tailings, groundwater, geochemical,* and *decay*. Articles that identified and explored physical, chemical, and biological factors influencing the behavior, fate, and transport of radionuclides in the subsurface across different zones were assessed for inclusion. This was achieved by reviewing the title and abstract, and subsequently the full article when relevant. While more than 1,000 articles were returned, only the first 1,000 articles were reviewed. Duplicates were removed from the search results.

Based on this search, twenty-seven factors influencing radionuclide fate and transport in the subsurface were included in this study and placed into six categories: meteorological, hydrological, hydrogeological, soil chemistry, groundwater chemistry, and mine tailings chemistry (Table 1). The factors identified, and their interdependencies, can enhance or hinder the mobility of radionuclides and influence their fate and transport either directly or indirectly (i.e., through other parameters). For example, the presence of clay coatings on fracture walls may facilitate or retard the transport of radionuclides. As clay particles are deposited onto fracture surfaces, the aperture decreases, and therefore the transmissivity (i.e., factor T) decreases, hindering radionuclide mobility. Transmissivity can also impact the hydrodynamic shear stress, which can mobilize clay particles thereby facilitating radionuclide mobility when adsorbed to clay.

Table 3-1. Factors influencing subsurface radionuclide migration

Category	Factors	References
Meteorological	Precipitation* (i)	Mohanty et al., 2015; Olsen et al., 1986
	Surface air temperature** (T_R)	Yu and Li, 2012; Ghiorse and Wilson, 1988
Hydrological	Surface runoff** (R)	Subramanya, 2008; Fetter, 2001
	Infiltration rate of the soil* (F)	Chen et al., 2005; de Jonge et al., 2004
Hydrogeological	Transmissivity* (T)	Zhang et al., 2015; Anderson et al., 2007
	Soil matrix porosity* (η)	Lopez-Galindo et al., 2008; Robinson et al., 1998
	Saturation* (S)	Testoni et al., 2017; Chen et al., 2005
	Clay fraction in the soil* (C_p)	Durrant et al., 2018; Brennan et al., 2014
Soil chemistry	Soil pH** (pH_g)	Lauber et al., 2009
	Soil microorganisms* (X_g)	Anderson et al., 2007; Simonoff et al., 2007
	Cation exchange capacity* (C_c)	Brennen et al., 2014; Bekhit et al., 2006
	Fraction of organic carbon* (f_{oc})	Lin et al., 2018
Groundwater chemistry	pH* (pH_w)	Li et al., 2016; de Jonge et al., 2004
	Temperature* (T_w)	Beyer et al., 2016; Liu et al., 2003
	Particulate concentration* (p_w)	Weisbrod et al., 2002; Eichholz et al., 1982
	Redox potential* (ε_w)	Gavrilescu et al 2009
	Ionic strength* (I_w)	Xie et al., 2013; de Jonge et al., 2004
	Alkalinity* (A_w)	Du et al., 2017; Jerden and Sinha, 2003
	Dissolved organic carbon* (DOC)	Zhao et al., 2011
Mine tailings chemistry	pH* (pH_m)	Li et al., 2016; Bekhit et al., 2006
	Temperature* (T_m)	Beyer et al., 2016; Liu et al., 2003
	Particulate concentration* (p_m)	Weisbrod et al., 2002; Eichholz et al., 1982

	Redox potential* (ε_m)	Krupka and Serne, 2002
	Ionic strength* (I_m)	Xie et al., 2013
	Alkalinity* (A_m)	Du et al., 2017; Jerden and Sinha, 2003
	Age of tailings* (t)	Hart, 2004; Gavrilescu et al., 2009
	Tailings microorganisms* (X_m)	Lusa et al., 2019

*Factors that directly influence the subsurface radionuclide migration.
**Factors that indirectly influence radionuclide migration through interaction with other factors.

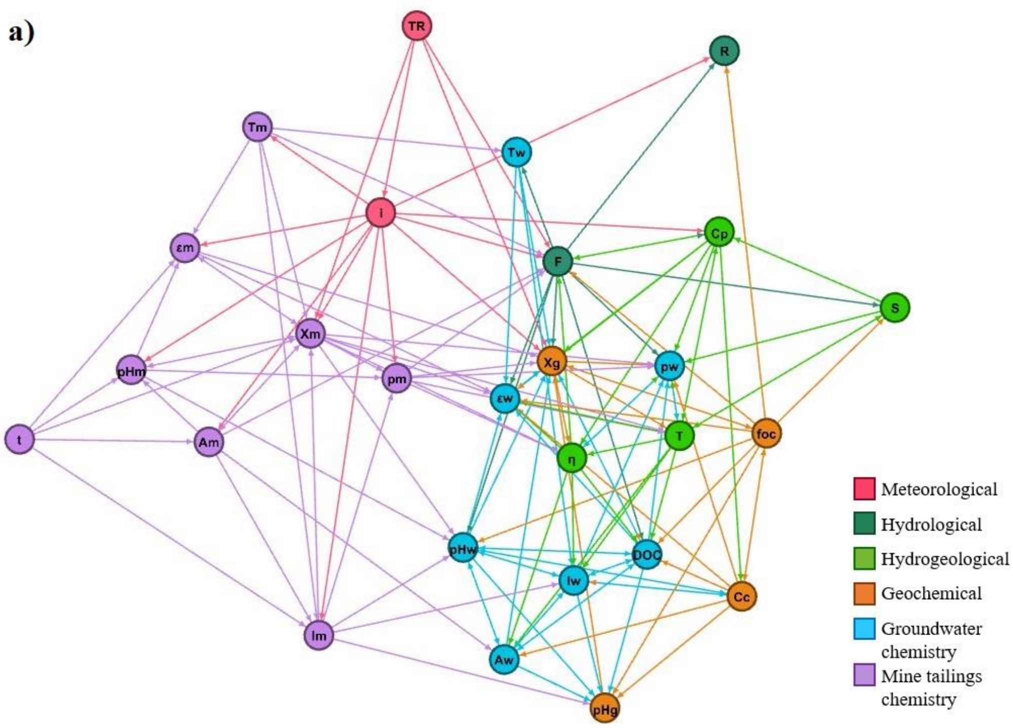
3.2.2. Network development

Within the CNT, interdependencies can be described mathematically through an adjacency matrix ($\boldsymbol{A}$). When two factors are independent, a value of zero is assigned to the corresponding entry of the adjacency matrix. On the other hand, interdependence is indicated by a value of one. Such interdependence can be either unidirectional or bi-directional, where the former is represented by a directed link and the latter is conceptualized as an undirected link. A general form of $\boldsymbol{A}$ is represented in Eq. 1 (Barabási, 2016):

$$\boldsymbol{A} = \begin{bmatrix} A_{1-1} & A_{1-2} & A_{1-3} \ldots\ldots A_{1-27} \\ A_{2-1} & A_{2-2} & A_{2-3} \ldots\ldots A_{2-27} \\ A_{3-1} & A_{3-2} & A_{3-3} \ldots\ldots A_{3-27} \\ & & \vdots \\ & & \vdots \\ & & \vdots \\ A_{27-1} & A_{27-2} & A_{27-3} \ldots\ldots A_{27-27} \end{bmatrix} \quad (1)$$

where $A_{i\text{-}j}$ represents the influence of factor i on factor j. This adjacency matrix $\boldsymbol{A}$ can be represented graphically in the form of a directed or undirected network by a network of factors (NoF). The adjacency matrix developed for this study, representing the interdependencies between the factors influencing radionuclide fate and transport in the subsurface, is presented in Appendix A and the corresponding directed NoF is shown in Fig. 1.

This NoF is represented as $G = \{N, L\}$, where N is the number of nodes and L is the number of links. The nodes represent the factors that influence the subsurface radionuclide migration, while the links represent the interdependence between these factors. Linkages between the factors reflect the existence of mathematical, physical, or logical relationships between the factors and can be either unidirectional or bidirectional. These linkages are thus used to represent interdependencies among factors. In this work, the linkages were determined based on previous (experimental, field, and analytical) related studies reported in the literature (e.g., Wang et al., 2014; Zhang et al., 2015; Bekhit et al., 2009) as well as the authors' knowledge in the field. The NoF developed in the present study is an unweighted-directed network with N=27 and L=144, where a link directed from factor 1 to factor 2 indicates the influence of factor 1 on factor 2, or factor 2's dependency on factor 1.



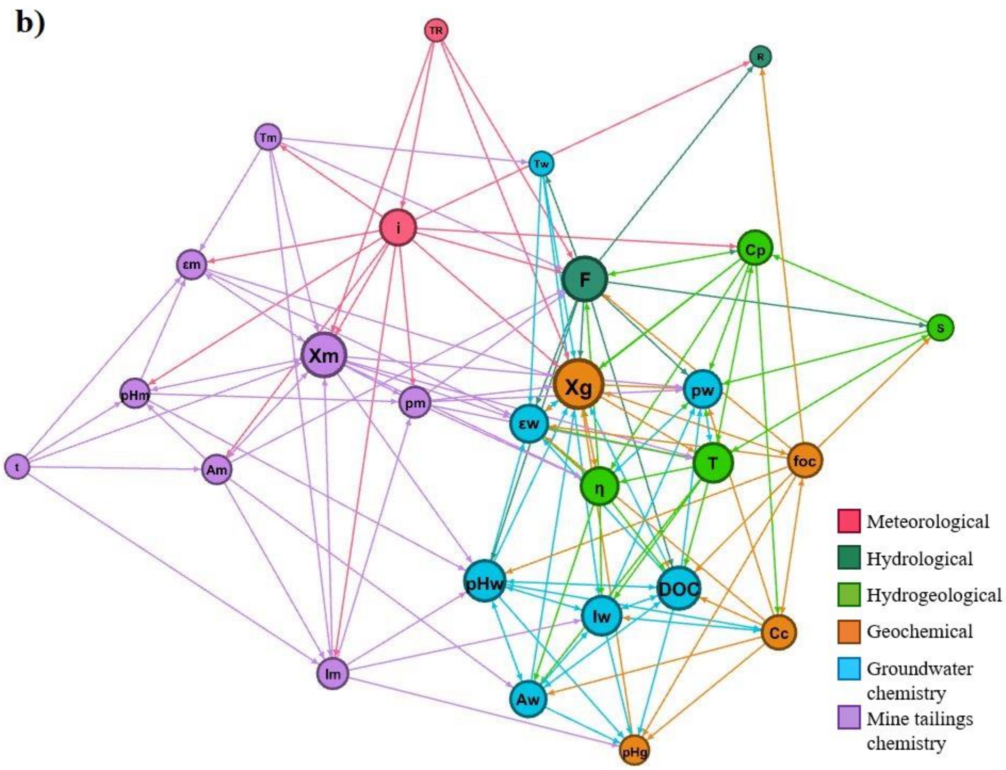


Fig. 3-1. The Network of Factors (NoF): a) all factors are of equal sizes irrespective of their interdependencies with other factors; and b) size nodes representing factors increase with the number of interdependencies.

It should be emphasized that the NoF developed in this study is based on the fact that UMTD failures result in the migration of radionuclide from the surface, through the vadose zone, to the aquifer. The factors influencing transport within this entire system have been included in a single NoF in which the same node is used to represent similar factors in different zones, as the interlinkages do not differ across zones. For example, the saturation factor (*S*) impacts the particulate concentration (p_m) in the groundwater, as well as the clay fraction (C_p) of the soil; hence, *S* is represented by a single node in the NoF. In addition, while most factors will have different values in different geological formations (e.g., granite (fractured) and sedimentary rock (porous)), the links between factors in the NoF are based on processes rather than values and are therefore not affected across different formations.

It should also be emphasized that once radionuclides are released from UMTDs into the environment, the fate and transport of all radionuclides in the mixture is controlled by the same processes (i.e., infiltration, advection, dispersion, adsorption/desorption, diffusion, decay, dissolution), which are characterized by the same factors (e.g., transmissivity, porosity, saturation, clay fraction in the soil, soil microorganisms, cation exchange capacity, fraction of organic carbon, chemistry of groundwater and mine tailings, age of tailings). While the magnitude of these factors may differ between radionuclides, a change in a factor will propagate to other linked factors regardless of the radionuclide. For example, uranium(VI), thorium(IV) and radium(II) have different sorption affinities for clay minerals. Uranium(VI) ions have a greater adsorption affinity for kaolinite and montmorillonite (Bachmaf and Merkel, 2011), thorium(IV) has a greater adsorption affinity for geothite (Melson, 2011) and bentonite (Khalili et al., 2012), and radium(II) ions has a greater adsorption affinity for bentonite, illite (Alhajji et al., 2016) and kaolinite (Reinoso-Maset and Ly, 2016). As such, the NoF employed in this study can be applied to a collective set of radionuclides released from UMTDs irrespective of the nature of each radionuclide.

3.2.3. Network Characteristics

The NoF is evaluated based on several metrics, including the network density (*D*), average degree ($<k>$), characteristic path length (*d*), network diameter (d_{max}), and clustering coefficient (C_{coe}). The metric *D* indicates the degree of closeness between the factors, and is estimated as the ratio between the actual number of links in the network and the maximum theoretical number of links that could be formed in that network (Gao et al., 2017):

$$D = \frac{L}{N(N-1)} \tag{2}$$

When the value of *D* is closer to 1.0 (i.e., a dense network), the factors are highly interdependent and connected intricately. In this situation it is crucial to identify the dominant factors based on their interdependence (Barabási, 2016).

The metric $<k>$ indicates the average direct influence between nodes in the network (Barabási, 2016). As the NoF is directed, the average degree can be expressed by average in-degree ($<k_{in}>$) and average out-degree ($<k_{out}>$). $<k_{in}>$ indicates the average number of links pointing towards the nodes, whereas $<k_{out}>$ represents the average number of links pointing away from the nodes. In directed networks, the $<k_{in}>$ and $<k_{out}>$ values should be equal, and are calculated as (Barabási, 2016):

$$< k_{in} > = < k_{out} > = \frac{L}{N} \tag{3}$$

The values of $<k_{in}>$ and $<k_{out}>$ can be calculated for each category in the NoF to provide an understanding of the relative impact each category has on radionuclide migration. Equation 3 can thus be modified to calculate $<k_{in}>$ and $<k_{out}>$ for each category as follows (Gao et al., 2017):

$$< k_{in} > = \frac{L_{cat\_in}}{N_{cat}} \tag{4}$$

$$< k_{out} > = \frac{L_{cat\_out}}{N_{cat}} \tag{5}$$

where L_{cat_in} and L_{cat_out} are the total number of inward- and outward-links of the factors in a category and N_{cat} is the number of factors in that category.

The metric *d* reflects the average number of shortest paths, which exist directly and indirectly, between all node (i.e., factor) pairs in the network (Gao et al., 2017). This metric indicates the average number of interdependencies required to form a chain of factors that have a collective impact on the subsurface radionuclide migration. This metric also indicates the degree of closeness between the factors, and is calculated as:

$$d = \frac{1}{N(N-1)} \sum_{i,j=1,N; i \neq j} d_{ij} \tag{6}$$

where d_{ij} is the shortest path between nodes i and j (Santiago et al., 2016), and is 1.0 when the factors i and j are directly linked.

The metric d_{max} refers to the maximum shortest pathlength between any pair of nodes in the network (Gao et al., 2017). In the NoF, d_{max} indicates the number of interdependencies required to connect the furthest two factors. The maximum shortest pathlength thus contains the maximum number of factors that can have a possible collective impact on radionuclide fate and transport.

The metric C_{coe} evaluates the extent to which a node's neighbours interact with each other (Barabási, 2016). Therefore, in the NoF, high C_{coe} indicates that a factor and its neighbours are highly interconnected and form a cluster. Such local clusters represent closed sets of interdependent factors that have a collective impact on the subsurface radionuclide migration. C_{coe} of node i in a directed network is represented by the ratio of actual directed triangles of node i to all possible number of triangles that can be formed by node i (Fagiolo, 2007):

$$C_{coe}(i) = \frac{(A + A^T)^3_{ii}}{2[k_i\,(k_i - 1) - 2k_i^{\leftrightarrow}]} \tag{7}$$

where $\boldsymbol{A^T}$ is the transpose of $\boldsymbol{A}$, k_i is the node degree (k_{in} + k_{out}), and $k_i^{\leftrightarrow}$ refers to the bidirectional links of the node i.

It is noteworthy that the values of C_{coe} and d can be used to determine whether the NoF is closer to a: 1) small-world network (i.e., Watts-Strogatz model), which implies that any two randomly chosen nodes in the network are connected; 2) random network, with nodes that connect in a chaotic manner; or 3) regular network, characterized by essentially an equal number of links per node, resulting in high C_{coe} values, significantly deviating from a small-world network behavior (Barabási, 2016). Given specific N and $<k>$ values, the corresponding d and C_{coe} can be calculated as follows (Gao et al., 2017):

$$d = \begin{cases} \dfrac{\ln N}{\ln <k>} & \text{for random networks} \\ \dfrac{N \times (N + <k> - 2)}{2 \times <k> \times (N-1)} & \text{for regular networks} \end{cases} \tag{8}$$

$$C_{coe} = \begin{cases} \dfrac{<k>}{N} & \text{for random networks} \\ \dfrac{(3 \times <k>) - 6}{(4 \times <k>) - 4} & \text{for regular networks} \end{cases} \tag{9}$$

The closer conformance of the NoF to the Watts-Strogatz model highlights the intricate interdependence between the different factors, and subsequently the need to study the latter.

3.2.4. Centrality Measures

Centrality is a key evaluation metric in CNT because it reveals the relative influence of individual nodes on the overall network structure (Freeman, 1979). Five centrality measures are used in the current study, related to the in-degree (ID_C), out-degree (OD_C), closeness (C_C), betweenness (B_C), and eigenvector (E_C), and are estimated relative to the maximum centrality value based on all nodes in the network. It is noteworthy that nodes with similar centralities may or may not have equal importance to the underlying network (Liu et al., 2016); therefore, multiple centrality measures are considered in the current study to provide a more in-depth analysis.

The in- and out degree centralities (i.e., ID_C and OD_C) help to identify the nature of connection. For example, factors with high ID_C values are susceptible to impacts from a large number of factors, and their states are mostly dependent on that of others. In contrast, high OD_C

values reflect that the corresponding factors can impact a large number of factors, thus having more impact on the system (i.e., radionuclide migration in the context of the present study). For a directed network, ID_C and OD_C of node *i* are calculated as:

$$ID_C(i) = \frac{k_{in}(i)}{N-1} \tag{10}$$

$$OD_C(i) = \frac{k_{out}(i)}{N-1} \tag{11}$$

where $k_{in}(i)$ and $k_{out}(i)$ are the in-degree and out-degree of node *i*, respectively (Nomikos et al., 2013).

The C_C of a node is a measure of its closeness to the rest of the nodes in the network (Gao et al., 2017). In the NoF, factors with high C_C values are closely connected, directly or indirectly, to a large number of other factors forming intricately connected sets that collectively affect radionuclide fate and transport. Note that, factors of high C_C strongly interact with their interrelated factors due to the shorter pathlength existing between them. C_C of node *i* is estimated based on the shortest pathlengths between it and all other nodes (Freeman, 1979):

$$C_C(i) = \frac{N-1}{\sum_{j \in N, j \neq i} d_{ij}} \tag{12}$$

The B_C is based on the frequency with which a node is present in the shortest paths between other node pairs (Freeman, 1977). In the NoF, factors with high B_C values connect large number of factor pairs that are not directly connected. Therefore, they act as a communication bridge within the network. Such factors can significantly affect radionuclide behavior as they act as the primary connectors in a set of independent factors that have a collective impact. The B_C of node *i* is calculated as:

$$B_C(i) = \left(\frac{1}{(N-1)(N-2)}\right) \sum_{j \neq i \neq k} \frac{\sigma_{jk}(i)}{\sigma_{jk}} \ \forall\, j,k \in N \tag{13}$$

where $\sigma_{jk}(i)$ is the number of shortest paths between nodes *j* and *k* that pass through the node *i*, and σ_{jk} is the total number of shortest paths between nodes *j* and *k* (Santiago et al., 2016).

The E_C of a node reflects the degree of interconnectivity of its neighbouring nodes (Ruhnau, 2000). Accordingly, factors with high E_C values have predominant indirect connections to factors other than their close neighbours and can therefore impact the radionuclide behavior significantly. E_C of node *i* is calculated as:

$$E_C(i) = \frac{1}{\lambda_{max}(A)} \sum_{j}^{N} a_{ji} v_j \qquad (14)$$

where v_j represents the eigenvector corresponding to the maximum eigenvalue $\lambda_{max}(A)$ of the matrix $\boldsymbol{A}$ (Santiago et al., 2016).

3.2.5. Sensitivity Analysis

In general, the robustness of complex networks can be measured by local or global *node removals* (Albert et al., 2000). Within the context of the current study, robustness assessment aids in identifying the factors that propagate uncertainty through the NoF through evaluating the variability of the network metrics (i.e., *d*, C_{coe}) due to factor removal, either randomly or in a predetermined (i.e., targeted) order.

A random node removal encompasses the consecutive removal of factors irrespective of any measure or property related to the factors, whereas a targeted node removal includes the subsequent removal of factors based on a certain property (Li et al., 2013). For random node removals, 300 realizations were applied for each scenario and the average was used as the representative response.

Under a targeted node removal scenario, factors were removed based on their centralities (i.e., ID_C, OD_C, C_C, B_C, and E_C) in a descending order. It is noteworthy that more than one factor may have the same centrality value, and thus the order in which these factors are removed in a targeted node removal can affect the removal of consecutive factors. This can lead to different trends of network characteristics. Therefore, each targeted node removal scenario was carried out through 100 realizations and the average was considered as the representative response. Variations in d and C_{coe} were subsequently plotted against the fraction of factors removed (f) for both random and targeted node removal to compare the impact of the different removal scenarios.

3.3 Results and Discussions

3.3.1. Network characteristics

The NoF is characterized as a dense network (D=0.21) with significant interdependencies between its factors in comparison to that presented by Gao et al. (2017), which was sparse in nature (D=0.06) with few and unique risk spreading paths. The large value of D indicates that the factors affecting radionuclide transport in the subsurface are intricately connected directly and/or indirectly through multiple links. This underscores the need to investigate the interdependencies between these factors to improve the understanding of radionuclide transport in the subsurface.

The $<k_{out}>$ (and $<k_{in}>$) of the NoF is 5.33 (Table 2), which implies that every factor in the network can influence, or be influenced by, an average of five other factors. This indicates a remarkably high degree of interdependencies among the factors, which must be integrated into an accurate understanding of radionuclide behavior in the subsurface. Average degree measures for each category in the NoF are also shown in Table 2.

Table 3-2. $<k_{in}>$ and $<k_{out}>$ of the NoF and its categories

Category	$<k_{in}>$	$<k_{out}>$
All	5.33	5.33
Meteorological	0.5	7.5
Hydrological	5.5	4.5
Hydrogeological	4.75	6.25
Soil chemistry	6.75	5.75
Groundwater chemistry	8.14	4.43
Mine tailing chemistry	3.63	5.13

The groundwater chemistry category has the highest $<k_{in}>$ value (8.14), attributable to the larger number of inward links associated with the redox potential (ε_w; k_{in}=12), particulate concentration (p_w; k_{in}=11), and dissolved organic carbon (DOC; k_{in}=9) factors. This large number of inward links supports the fact that these factors exhibit the potential to be significantly impacted by soil chemistry (i.e., soil microorganisms (X_g), cation exchange capacity (C_c)), hydrological (i.e., infiltration rate of the soil (F)), and hydrogeological (i.e., Transmissivity (T), porosity (η)) factors. The meteorological category has the lowest $<k_{in}>$ value (0.5), as its factors such as precipitation (i; k_{in}=1) and surface air temperature (T_R; k_{in}=0) are relatively insensitive to changes in factors associated with other categories. In contrast to its low $<k_{in}>$ value, the meteorological category has the highest $<k_{out}>$ value (7.5). This is attributed to the factor i (k_{out} = 11), which can influence a large number of other factors. The groundwater chemistry category has the smallest $<k_{out}>$ value (4.43), which is attributed to the factors p_w (k_{out} = 2) and ε_w (k_{out} = 1) as they have relatively little influence on other factors. This discussion highlights the need for CNT models to analyze such a complex system as the importance associated with each factor should be based on its degree.

The metric d has a value of 1.95 (i.e., the average shortest pathlength includes a set of three interdependent factors), which indicates that the NoF is relatively small with a short distance between the factors. The d_{max} of the NoF is 4.0, indicating that a maximum of five factors can form a set with a collective impact on subsurface radionuclide transport. This means that a change in a factor can impact one other factor directly and three other factors indirectly. For example, i impacts F directly, and C_p (clay fraction in the soil), X_g, p_w indirectly.

Saturation factor, S has the highest C_{coe} value (0.833) in the NoF (Fig. 2), implying that S along with its neighbours (i.e., p_w, C_p, f_{oc}, F, and T) form a local cluster (Fig. 3). The cluster surrounding *S* is consistent with the understanding of the system as follows: fraction of organic carbon (f_{oc}) impacts *S* through the amphipathic property of the organic carbon, while *S* impacts *T* and controls the contribution of C_p and f_{oc} in the vadose zone. All of these factors influence the transport of p_w. The cluster formation around *S* also indicates that this factor along with its neighbouring factors act as a relatively closed set that will collectively, rather than individually, impact radionuclide fate and transport in the subsurface.

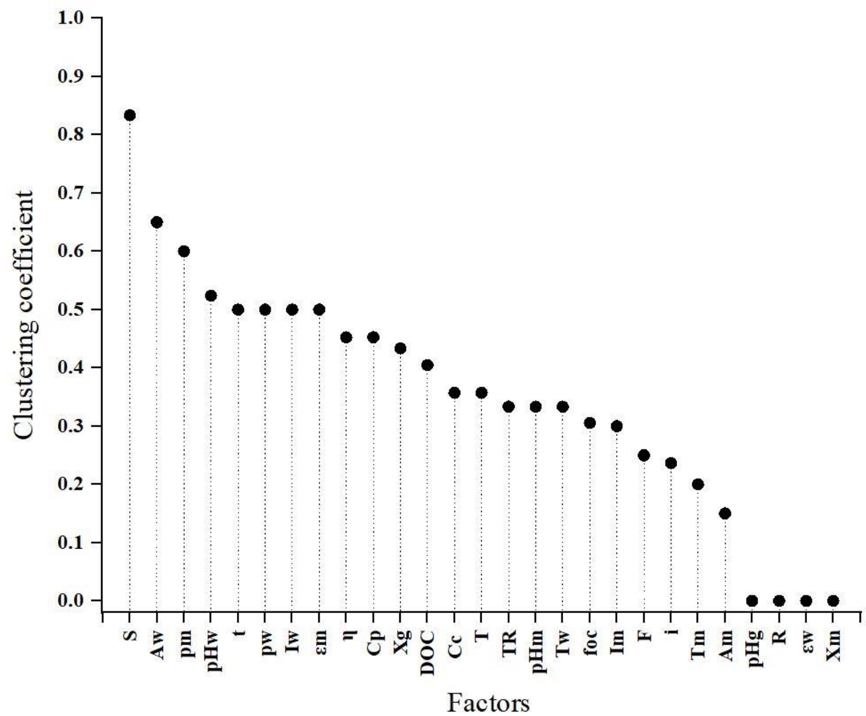


Fig. 3-2. Clustering coefficient (C_{coe}) of all factors in the NoF

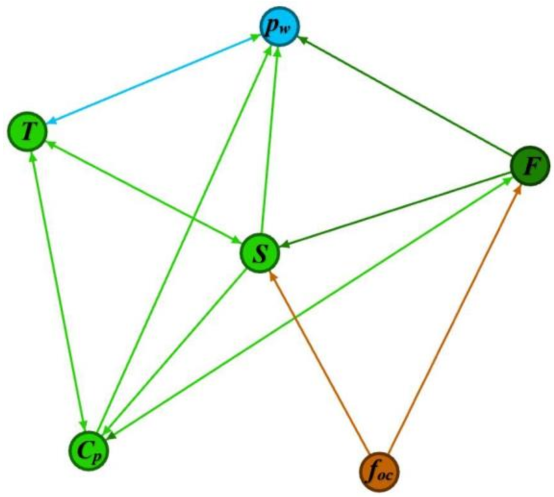


Fig. 3-3. The saturation (S) node and its interdependent factors
Note: The interdependencies between *S*'s neighbours and other factors in the network are not shown here in order to present a clear cluster formation surrounding *S*

C_{coe} and d values of the NoF were compared to those of regular and random networks having the same N and $<k>$ values. It is found that C_{coe} (0.360) and d (1.95) of the NoF are smaller than those of the regular network (0.577 and 2.96, respectively). C_{coe} of the NoF is larger than those of the random network (0.197) but d of the NoF is approximately equal to that of random network (1.96). Therefore, the NoF conforms more to the Watts-Strogatz model (i.e., has a small-world property), where nodes are densely connected. This further emphasizes the cruciality of considering the interdependencies between all factors impacting radionuclide fate and transport in the subsurface.

3.3.2. Centrality measures

The normalized ID_C and OD_C values for all factors in the NoF are presented in Fig. 4. Factors with higher ID_C values are more sensitive to being impacted by other factors, while those with relatively low ID_C values are considered independent of other factors. Factor X_g has the highest ID_C (0.538), which is expected as the mobility of microorganisms is controlled by the balance of attractive and repulsive forces between them and a collector surface. These forces are governed by the physicochemical properties of the water matrix (i.e., pH_w (pH), ε_w, T_w

(temperature), A_w (alkalinity)), the surface properties of the microorganisms and solid collectors, hydrodynamic forces (i.e., T), and interstices present on solid collectors (i.e., η) (Ghiorse and Wilson, 1988; Heath, 1983; Taylor et al., 2004). When present in the subsurface, soil microorganisms (i.e., X_g) may facilitate or hinder radionuclide migration (Simonoff et al., 2007). The survival and mobility of microorganisms are thus affected by nearly 50% (i.e., ID_C = 0.538) of the factors in the NoF. Hence, the role of X_g in radionuclide fate and transport must be carefully evaluated by considering its interdependencies with other factors. Factors p_w and ε_w, in the water matrix category, also have high ID_C values as they are sensitive to infiltration originating from UMTDs and the physical and chemical properties of the soil (Brindha and Elango, 2014). Factors with an ID_C value of zero (i.e., age of tailings (t) and T_R), are not dependent on other factors; their interdependencies are unidirectional as other factors may be influenced by them. For example, as tailings age (i.e., Factor t changes with time), their mineralogy changes and new minerals (e.g., smectite) that readily adsorb radionuclides are formed (Hart, 2004). This can affect the fate and transport of radionuclides.

Factors with high OD_C values influence many other factors in the NoF directly, while the opposite is true for factors with low OD_C values. Factor i has the highest OD_C (0.423) value in the NoF (Fig. 4), which is expected as i impacts the chemistry of the uranium mine tailings while contained in their open impoundment structures and can mobilize C_p and X_g from the soil matrix (through F and T) (Mohanty et al., 2015). Hence, it is essential to consider the effect of the interdependencies between i and other factors on the transport of radionuclides in the subsurface. Fig. 4 also indicates factors F, f_{oc}, and X_m (microorganisms in the mine tailings) have the same high OD_C values (0.346). These high OD_C values highlight the importance of including these factors (i.e., i, F, f_{oc}, and X_m) accurately in order to adequately represent radionuclide migration in the subsurface. Conversely, factors with low OD_C values have little influence over other factors in the NoF. In this work, the surface runoff factor, *R*, has an OD_C value of 0; as such, its magnitude and accuracy will have little impact on any fate and transport model for radionuclides in the subsurface.

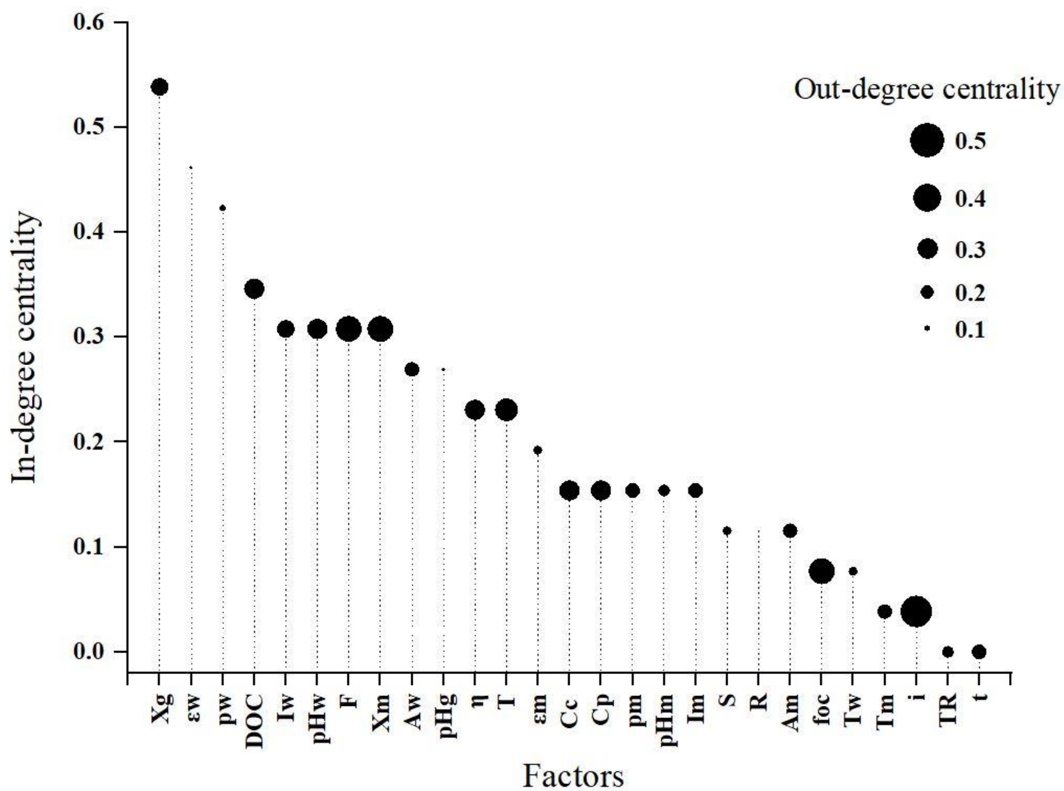


Fig. 3-4. Normalized degree centrality values of all factors in the NoF

Factor *F* has the highest C_C (0.696) value in the NoF (Fig. 5), indicating that it is closely linked to multiple other factors, either directly or indirectly. Therefore, changes in the value of *F* can rapidly influence such factors. This supports the hydrogeological understanding of the system, as *F* will transport radionuclides from the tailings through the vadose zone to the saturated zone, influence *S* and mobilize C_p in the vadose zone, and alter the groundwater chemistry (i.e., pH_w, T_w, p_w, ε_w, DOC, I_w (ionic strength)) in the saturated zone. Factor *R* has a negligible C_C value (0.0) as it is directly connected with only three other factors (i.e., *i*, *F*, f_{oc}). As such, it has little influence over other factors in the NoF since indirect connections between *R* and other factors can only be established through *i*, *F*, and f_{oc}. Most of the C_C values in this NoF range from 0.4 to 0.7, indicating that the NoF is dense with many factors connected through short paths. This also implies the importance of considering the collective, rather than the individual, impact of the factors on the migration of radionuclides in the subsurface.

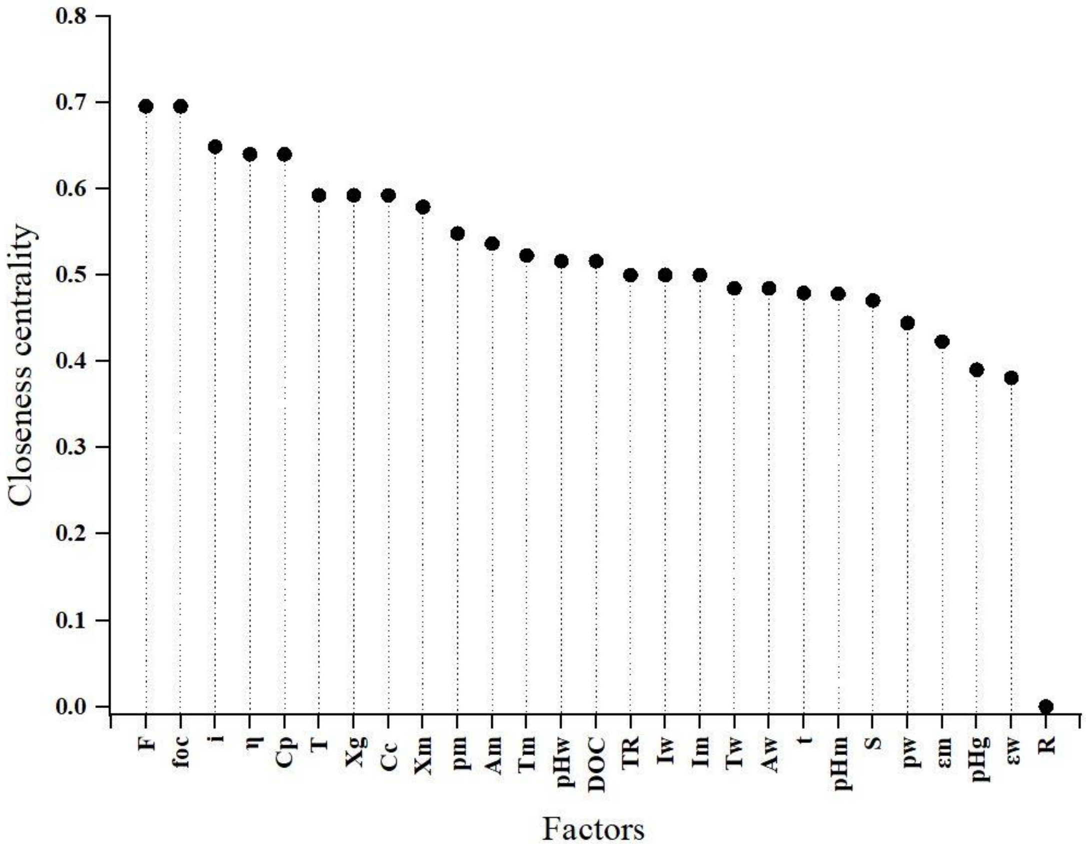


Fig. 3-5. Closeness centrality (C_C) values of all factors in the NoF

The B_C values of X_g (0.136) and *F* (0.096) are seven and five times higher than the average B_C of all other factors (0.018), respectively (Fig. 6). This indicates that these two factors act as primary connectors along most of the shortest paths in the NoF. This is consistent with the general understanding of the role of microorganisms in hindering and facilitating radionuclide transport in the subsurface. For example, Li et al. (2019) found that the bacterial strain MRS-1 *Bacillus cereus* tends to form complexes with uranium metal ions thereby facilitating uranium transport. Microorganisms have also been found to affect the fate of radionuclides by altering their chemical state. For example, Abdelouas et al. (2000) showed that sulphur-reducing bacteria are capable of reducing uranium from U(VI) to U(IV). The B_C value for F is large as it dictates the amount of infiltration, and thus the mass of uranium mine tailings, entering the subsurface. Most surface factors have B_C values of zero (i.e., R, T_R) indicating that these factors do not act as major connectors within the NoF. Although the mineralogy of uranium mine tailings evolves with time (i.e., new minerals are formed), the chemistry of the tailings (i.e., pH (pH_m), temperature (T_m), particulate concentration (p_m), ionic strength (I_m), redox potential (ε_m), alkalinity (A_m)) is not influenced by other factors in the NoF until the new materials are released to the environment (Hart, 2004). Therefore, the B_c value of the age of mine tailings (t) is also zero.

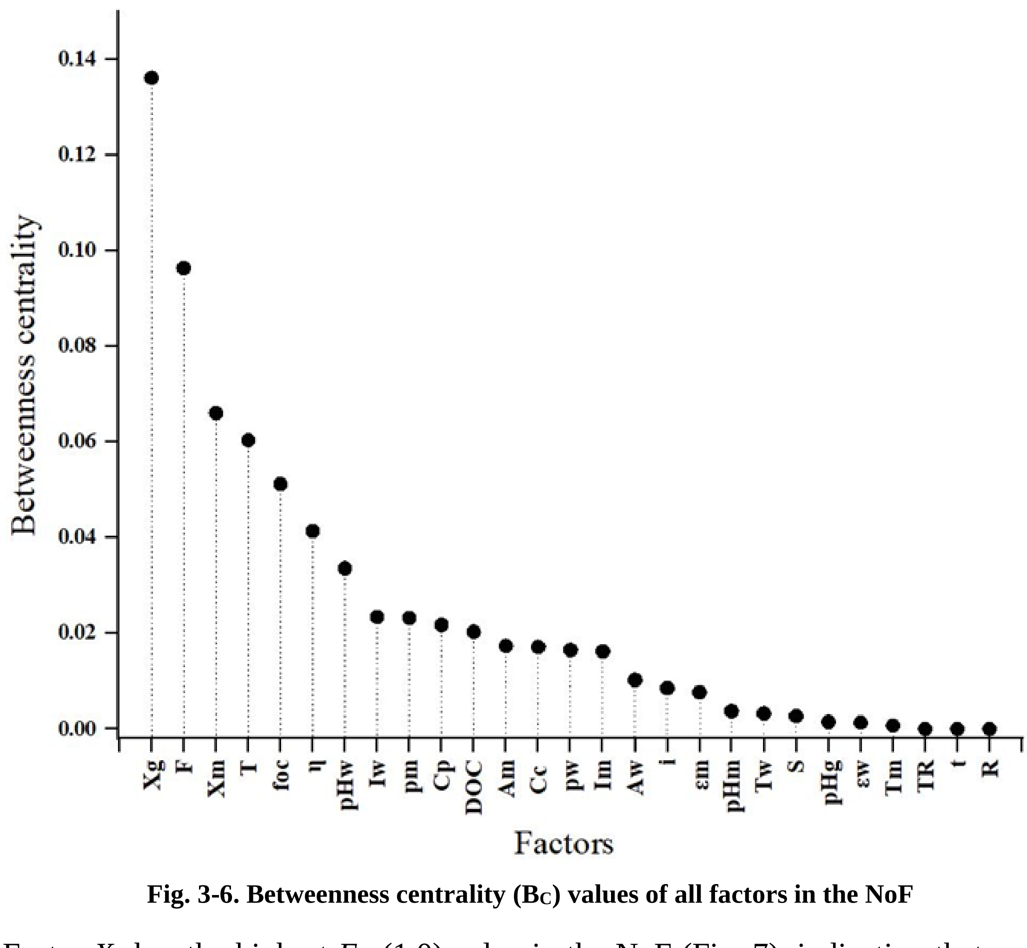


Fig. 3-6. Betweenness centrality (B_C) values of all factors in the NoF

Factor X_g has the highest E_C (1.0) value in the NoF (Fig. 7), indicating that most of X_g's direct neighbours have a high number of interdependencies in the NoF. As such, any impact on X_g

can significantly affect the radionuclide migration, since it is part of the set that contains a majority of the dominant factors in the NoF. Meteorological factors (i, T_R), T_m, and t have E_C values of zero. These factors are not impacted by others in the network.

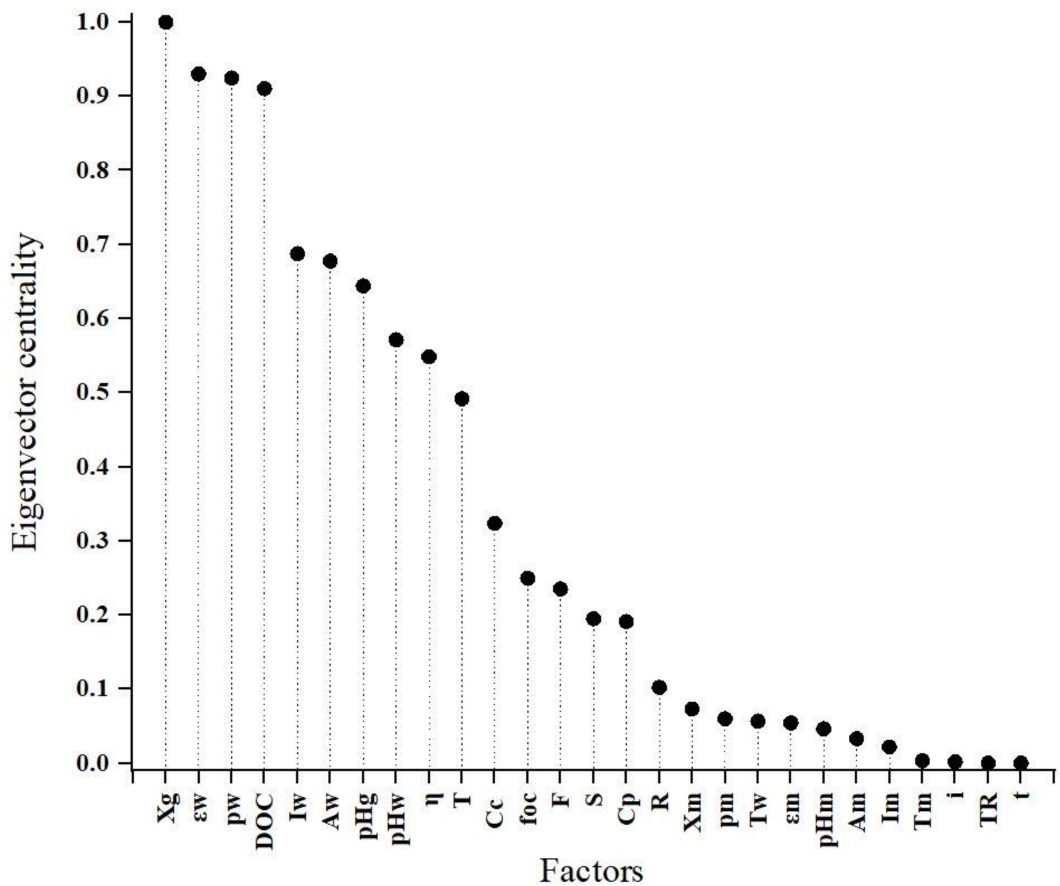
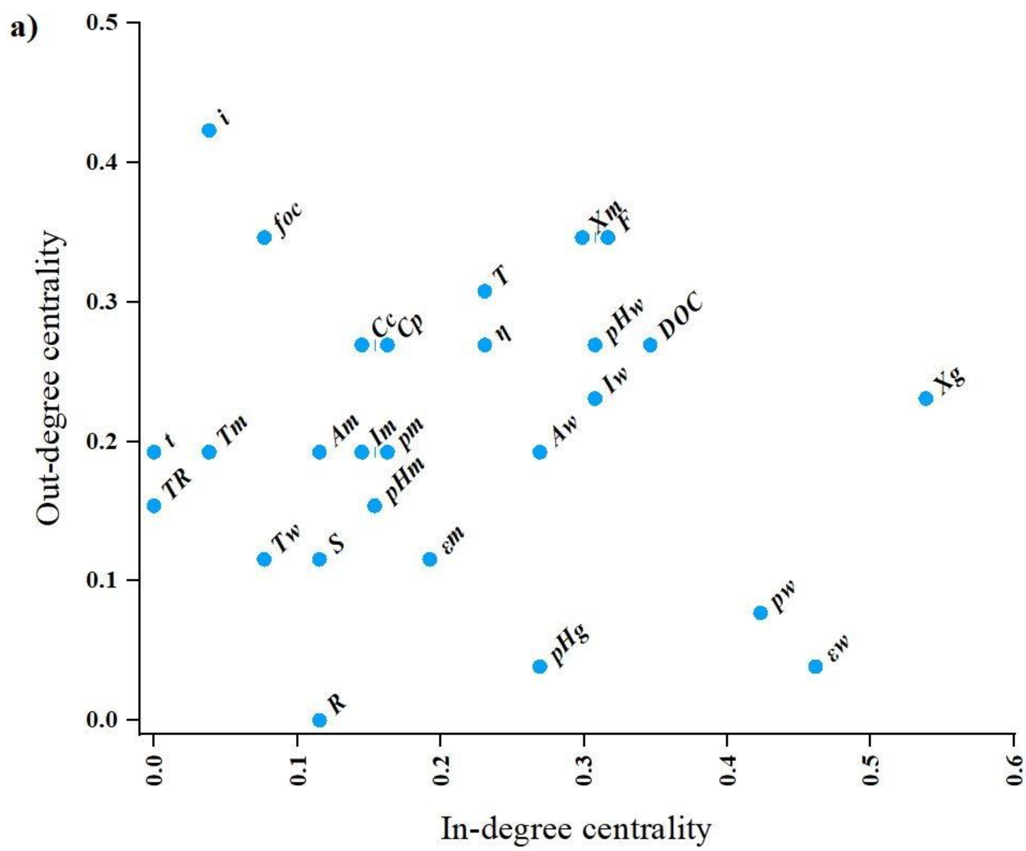


Fig. 3-7. Eigenvector centrality (E_C) values of all factors in the NoF

The centrality analysis results show that some factors exhibit high values for more than one measure of centrality. Several factors exhibit both high ID_C and OD_C values (i.e., F, DOC, X_m, I_w, pH_w), while other factors either influence other factors (i.e., high OD_C – i, f_{oc}, T, η, C_p, C_c) or are influenced by other factors (i.e., high ID_C – X_g, p_w, ε_w, A_w, pH_g (soil pH)) (Fig. 8(a)). Two pairs of centrality measures exhibit strong positive correlation: ID_C and E_C (r = 0.86) and OD_C and C_C (r = 0.82) as shown in Figs. 8(b) and 8(c), respectively. The positive correlation between ID_C and E_C indicates that factors with high ID_C (i.e., X_g, ε_w, p_w) can significantly influence or be significantly influenced by factors other than their close neighbours. The positive correlation between OD_C and C_C implies that factors with high OD_C (i.e., i, f_{oc}, T, η, C_p, C_c) will influence most other factors in the NoF, either directly or indirectly through short pathlengths. No other correlations were identified among the considered centrality measure pairs.



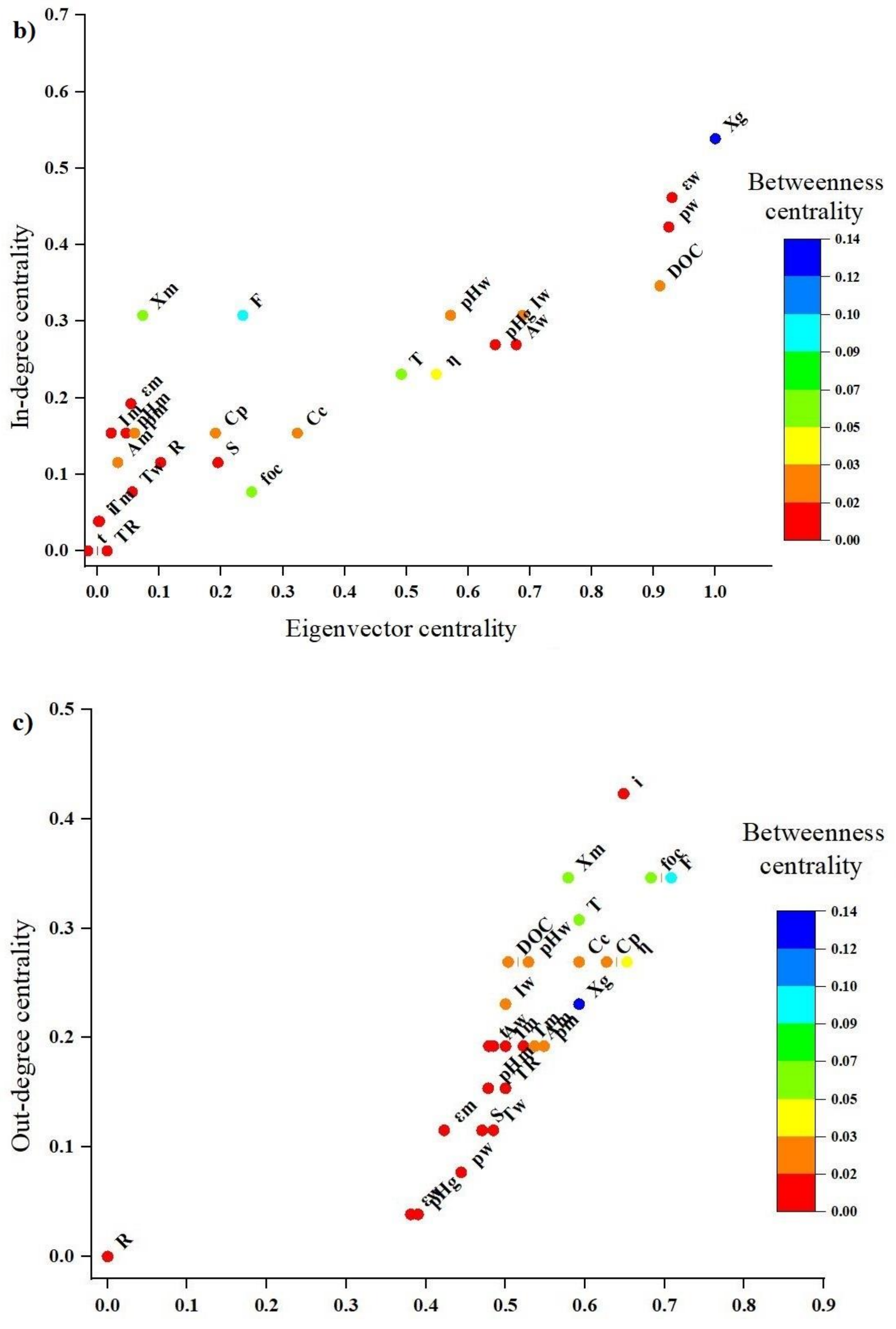
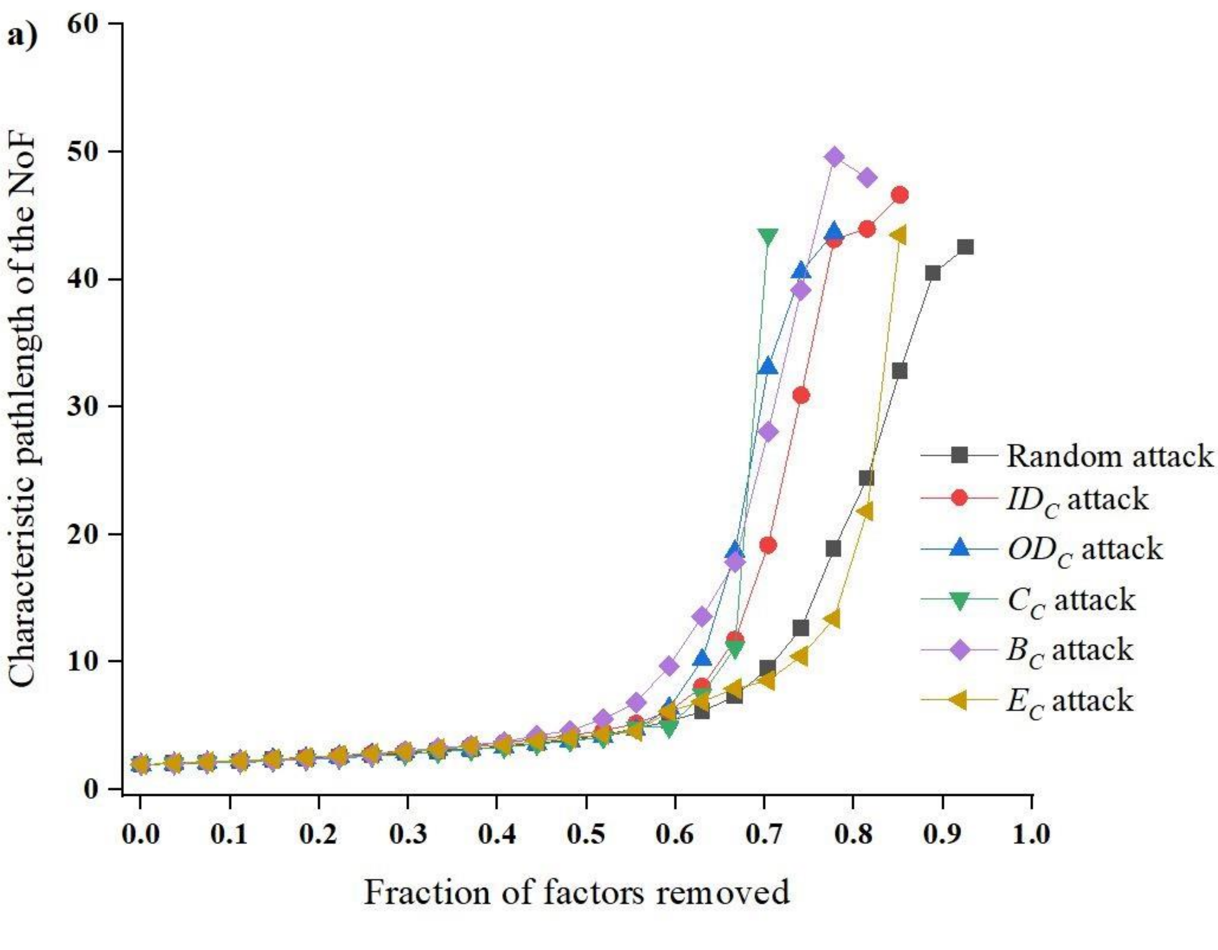


Fig. 3-8. Correlation among centrality measures for a) ID_C and OD_C; b) E_C, ID_C, and B_C; and c) C_C, OD_C and B_C

3.3.3. Sensitivity Analysis

The sensitivity analysis helps to individualize factors which can largely disrupt the metrics of the NoF, and thus neglecting their impact can significantly alter our understanding of radionuclide fate and transport mechanisms. Both random and targeted node removals were conducted to represent the propagation of undefined and defined levels of uncertainties in the network factors, respectively. The data from these node removals are included in Appendix B. For both random and targeted node removals, the factors remain well-connected even after removing 50% of the factors and their corresponding links from the NoF (Fig. 9(a)). Beyond that limit (i.e., $f > 0.5$), d increased exponentially for both node removal types (i.e., the NoF no longer exhibits a small-world property and $d > ln\ N$) with a higher rate of increase under targeted removal scenarios. This can be explained by the fact that any two factors in the NoF are connected through multiple pathways rather than a unique path. As node removal proceeds beyond $f = 0.5$, the removal of both groundwater and mine tailing water matrix properties (i.e., pH_w, pH_m, I_w, I_m, A_w, A_m) caused an accelerated rate of increase in d, particularly in a B_C-based node removal scenario. These properties are intricately connected with colloid-facilitated radionuclide transport phenomena as they strongly affect colloid retention and mobilization (Shiratori et al., 2007; Xie et al., 2013).

As the NoF contains interlocked local clusters (Fig. 5), a gradual decrease in C_{coe} is expected under random node removal scenarios (Fig. 9(b)). The C_{coe} trends under targeted node removals vary significantly when compared to those under random node removals (Fig. 9(b)). The rate of decrease in C_{coe} is higher for targeted node removals than random node removals, and it is the highest for the B_C-based node removal. However, C_{coe} drops to zero (or nearly zero) for all targeted node removals when $f \geq 0.8$. Factors with high B_C values (i.e., X_g, F, T, f_{oc}, and X_m) are significant factors within local clusters, as they act as connectors between them. Therefore, their removal can fragment local clusters rapidly. Factors X_g, f_{oc}, and X_m are colloid-related, thus influencing colloid-facilitated radionuclide transport. In addition, factors F and T impact the advective rate at which X_g and X_m are transported, thus influencing the fate and transport of radionuclides in the subsurface. The removal of either set of factors (i.e., X_g, f_{oc}, and X_m or F and T) disconnects the indirectly linked factors across local clusters, which decreases C_{coe}. Thus, it is critical to reduce uncertainty in X_g, f_{oc}, X_m, F, and T as much as possible, as uncertainty in these factors will amplify that of the model output (subsurface radionuclide fate and transport in the context of this study) more than uncertainty in other factors will.



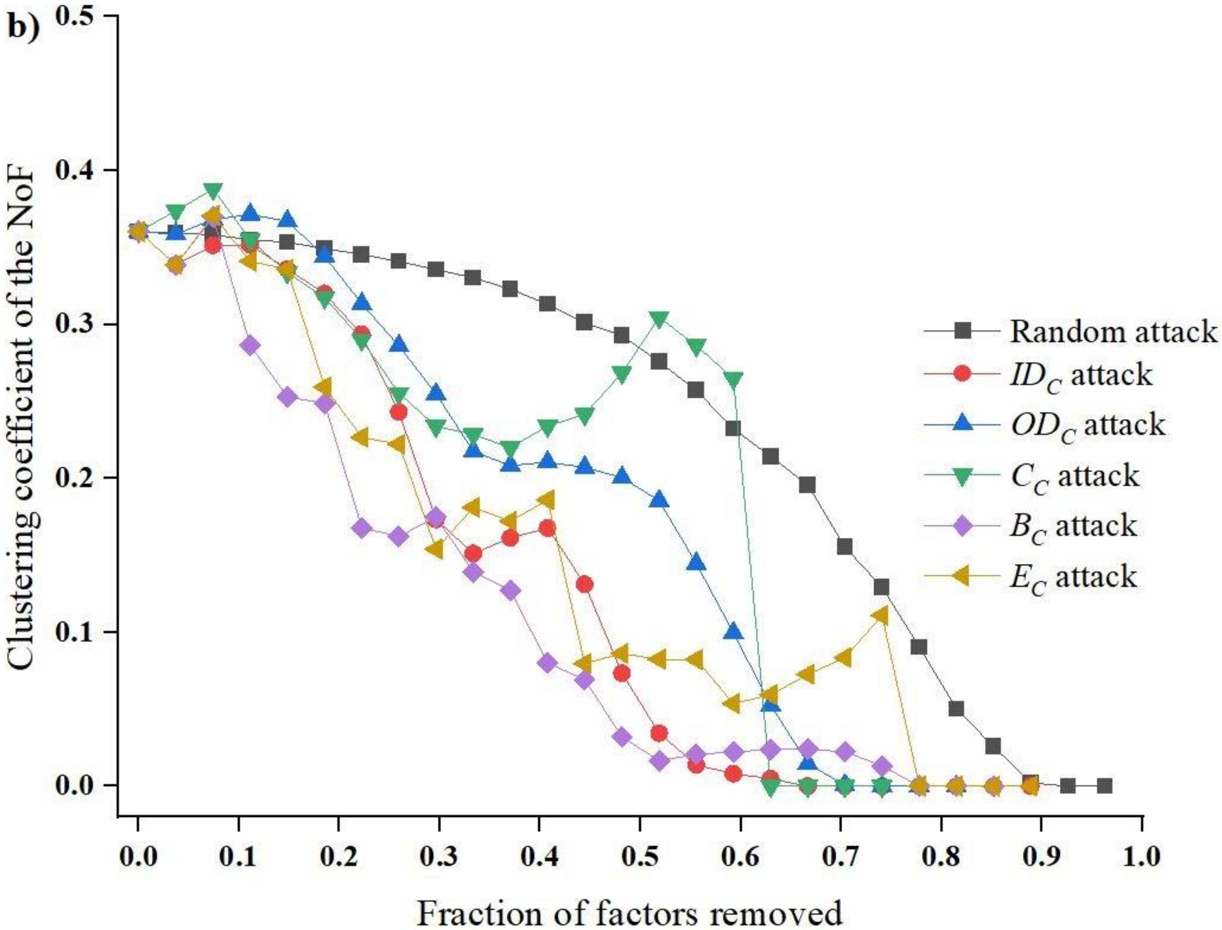


Fig. 3-9. Variation in network metrices such as: a) characteristic pathlength; and b) clustering coefficient of the NoF due to random and targeted node removal

3.4 Conclusions

Radionuclide migration in the subsurface is influenced by several interdependent factors that characterize the elements of the system (i.e., meteorological, hydrological, hydrogeological, chemistry of mine tailings, soil, and groundwater). A good understanding of the impacts of the interdependencies among these factors can aid in the development of reliable subsurface radionuclide fate and transport models. To that end, the current study adopts a CNT approach to investigate these impacts, and subsequently identify the primary factors influencing the fate and transport of radionuclides in the subsurface.

Of all categories, the groundwater chemistry category has the least influence on other categories and is highly influenced by other categories. The meteorological category has the most influence on other categories and is hardly influenced by other categories. A local cluster was formed between S, p_w, C_p, f_{oc}, F, and T, indicating that they act collectively, rather than individually, to impact radionuclide fate and transport in the subsurface. Factors X_g, p_w, and ε_w have the highest ID_C indicating that they are influenced by a number of other factors; as such, these factors will experience the most uncertainty as it propagates through the model. Conversely, i, F, f_{oc}, and X_m influence the most other factors in the NoF; uncertainty in these factors will be propagated through the model. F is closely linked (i.e., short pathlengths) to multiple other factors, and both X_g and F act as primary connectors among most of the shortest paths in the network. As such, uncertainties associated with factors influencing X_g and F will rapidly propagate (cascade) through the model. Additionally, these uncertainties will be amplified as they are added to those associated with X_g and F, and collectively propagate quickly to downstream factors along the path. Factors i, f_{oc}, T, η, C_p, C_c will influence most other factors in the NoF, either directly or indirectly through short pathlengths. Therefore, uncertainty in these factors will be propagated through the model. The sensitivity analysis indicated that the factors X_g, f_{oc}, X_m, F, and T should be carefully characterized to minimize their uncertainty when developing an integrated subsurface radionuclide fate and transport model, as uncertainty in these parameters will be amplified in the model output.

The CNT-based approach applied in this study can be used to investigate and conceptualise the interlinkages between all factors influencing radionuclide fate and transport in the subsurface, which is crucial for *i*) identifying the factors with the most influence on radionuclide fate and transport; and *ii*) strategically allocating data collection resources in order to minimizing uncertainty in corresponding fate and transport models. Together, these lead to more reliable fate and transport models, and ultimately better management and remediation strategies to mitigate impacts from UMTD failures. It should be highlighted that the NoF presented in this paper encompasses unweighted, directed links. Future work could include weighed links, which would require knowledge of the explicit relationships between the factors. These relationships could either be based on mathematical expressions or empirical observations, the latter of which would require a substantial amount of data to develop.

Acknowledgments

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Declaration of Competing Interest

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

Data Availability

The values presented in the adjacency matrix (Table A.1 in appendix A) are based on the information obtained from the references in Table. 1.

Computer code availability

Gephi v0.9.2 was used to develop the NoF and calculate centrality measures and the network metrices. Standard functionality of NetLogo v6.0.4 was utilized to write code in Scala for performing sensitivity analysis and to generate the plots for this manuscript. The code is freely available at https://github.com/BNarayan16/NoF/tree/BNarayan16-NoF-sensitivityanalysis

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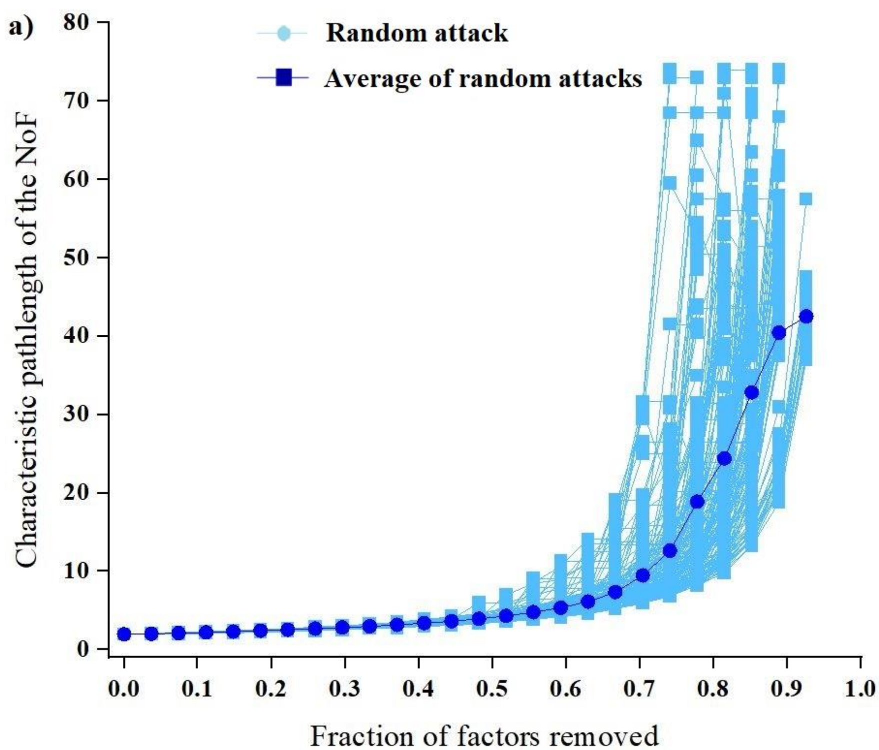
Appendix A

Table A. 1. Adjacency matrix values of each individual pairs of factors in the NoF

	Influencing factors																										
Dependeing factors	i	T_R	R	F	T	η	S	C_p	pH_g	X_g	C_c	f_{oc}	pH_w	T_w	p_w	ε_w	I_w	A_w	DOC	pH_m	T_m	p_m	ε_m	I_m	A_m	t	X_m
i	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
T_R	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
R	1	0	0	1	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
F	1	1	0	0	0	1	0	1	0	0	0	1	0	0	0	0	0	0	0	0	1	1	0	0	1	0	0
T	0	0	0	0	0	0	1	1	0	1	0	0	0	0	1	0	0	0	0	0	0	1	0	0	0	0	1
η	0	0	0	0	1	0	0	1	0	1	0	0	0	0	1	0	0	0	0	0	0	1	0	0	0	0	1
S	0	0	0	1	1	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
C_p	1	0	0	1	1	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
pH_g	0	0	0	0	0	0	0	0	0	0	1	1	1	0	0	0	1	1	1	0	0	0	0	1	0	0	0
X_g	1	1	0	1	0	1	0	1	1	0	0	1	1	1	0	1	0	1	1	0	0	1	1	0	0	0	0
C_c	0	0	0	0	0	0	0	1	0	0	0	1	1	0	0	0	1	0	0	0	0	0	0	0	0	0	0
f_{oc}	0	0	0	0	0	0	0	0	0	1	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
pH_w	0	0	0	1	0	0	0	0	0	0	0	1	0	0	0	0	1	1	1	1	0	0	0	1	0	0	1
T_w	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0
p_w	0	0	0	1	1	1	1	1	0	1	1	0	0	0	0	0	1	0	1	0	0	1	0	0	0	0	1
ε_w	0	0	0	1	1	1	0	1	0	1	1	1	1	1	0	0	0	0	1	0	0	0	1	0	0	0	1
I_w	0	0	0	0	1	1	0	0	0	0	1	0	1	1	0	0	0	1	1	0	0	0	0	1	0	0	0
A_w	0	0	0	0	1	1	0	0	0	0	1	0	1	0	0	0	1	0	1	0	0	0	0	0	1	0	0
DOC	0	0	0	1	1	1	0	0	0	1	1	1	1	0	0	0	1	1	0	0	0	0	0	0	0	0	0
pH_m	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	1	1
T_m	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
p_m	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	1	0	0	1
ε_m	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	1	0	0	0	0	1	1
I_m	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	1	1	0
A_m	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	1
t	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
X_m	1	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	1	0	1	1	1	1	0

Appendix B

In this appendix section, graphs in Fig. B.1 depicts the variation in the network characteristics during all 200 random node removals and the average of all the random node removals at each fraction of factor removed is given and Fig. B.2 and Fig. B.3 indicates the same during 100 trials of each centrality-based node removal and their average.



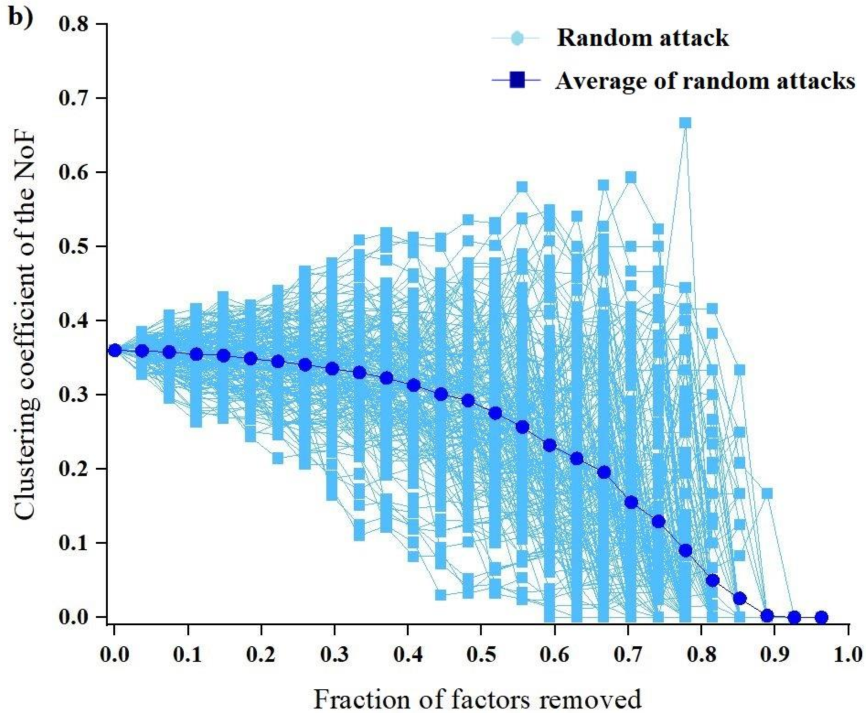
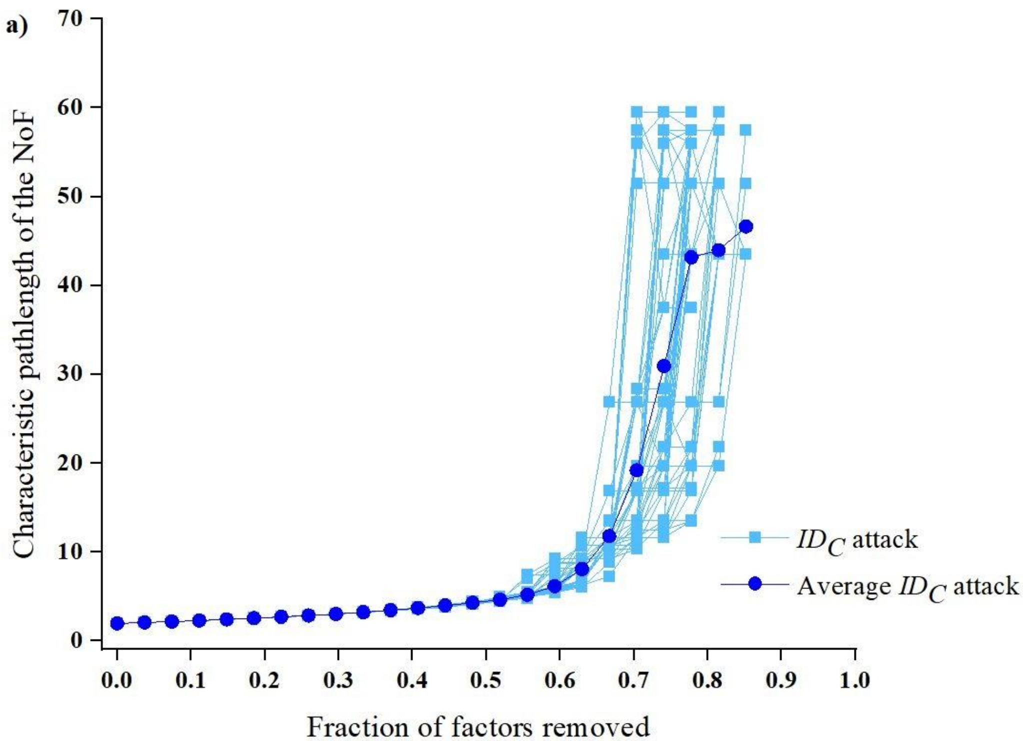
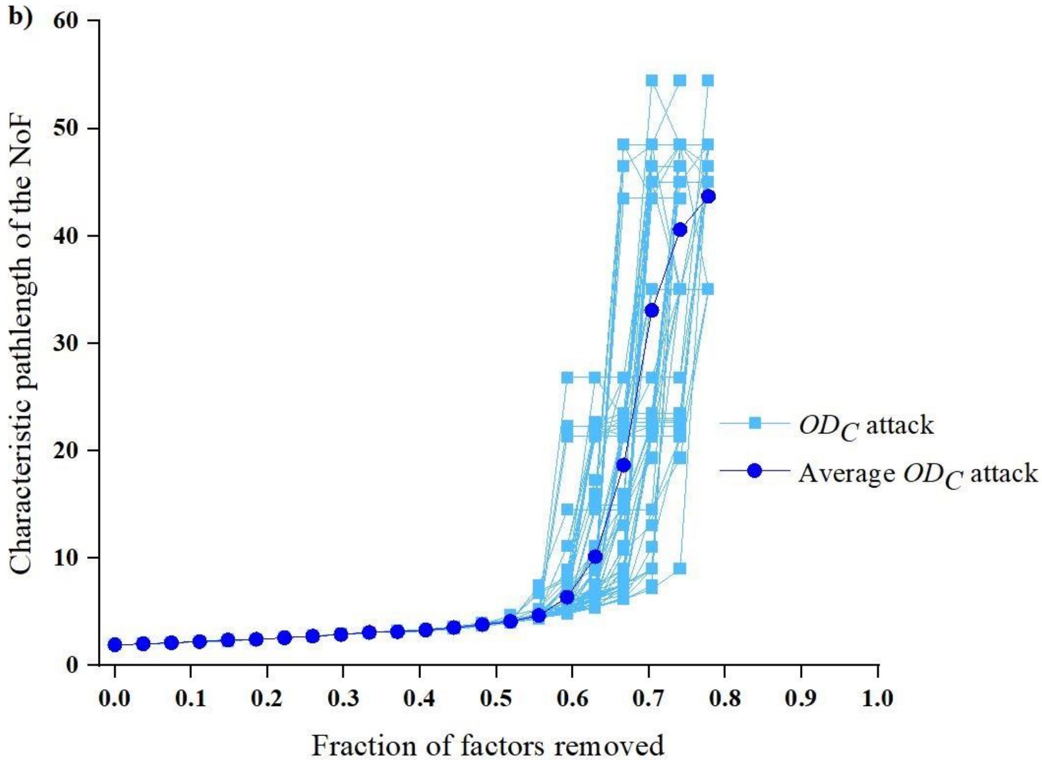
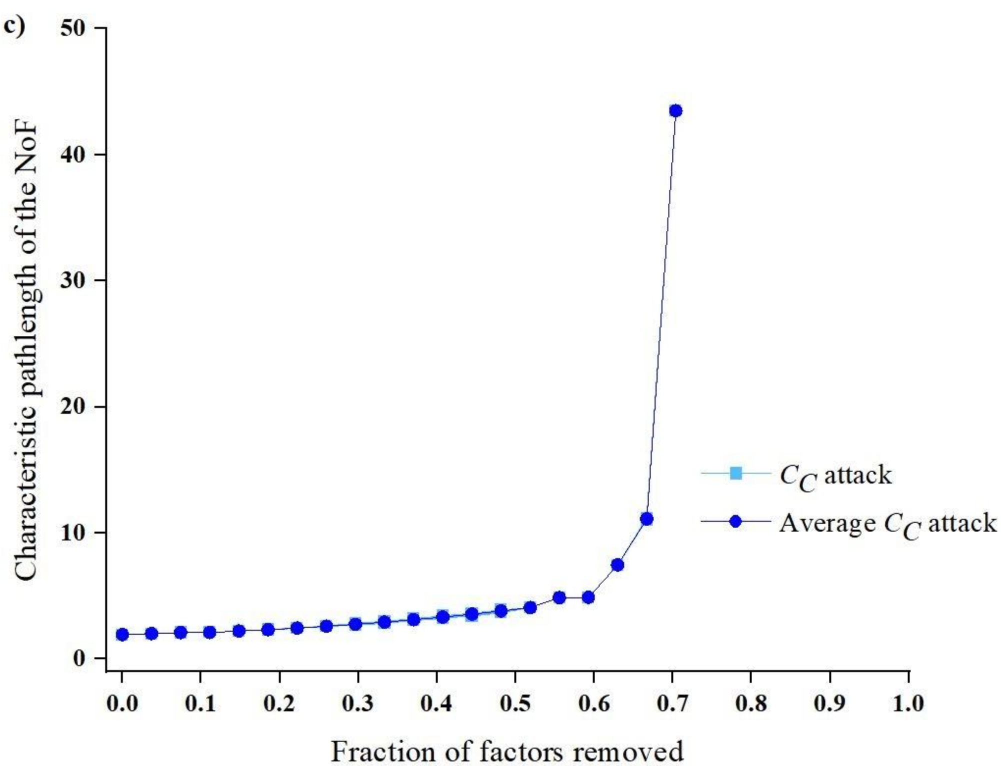
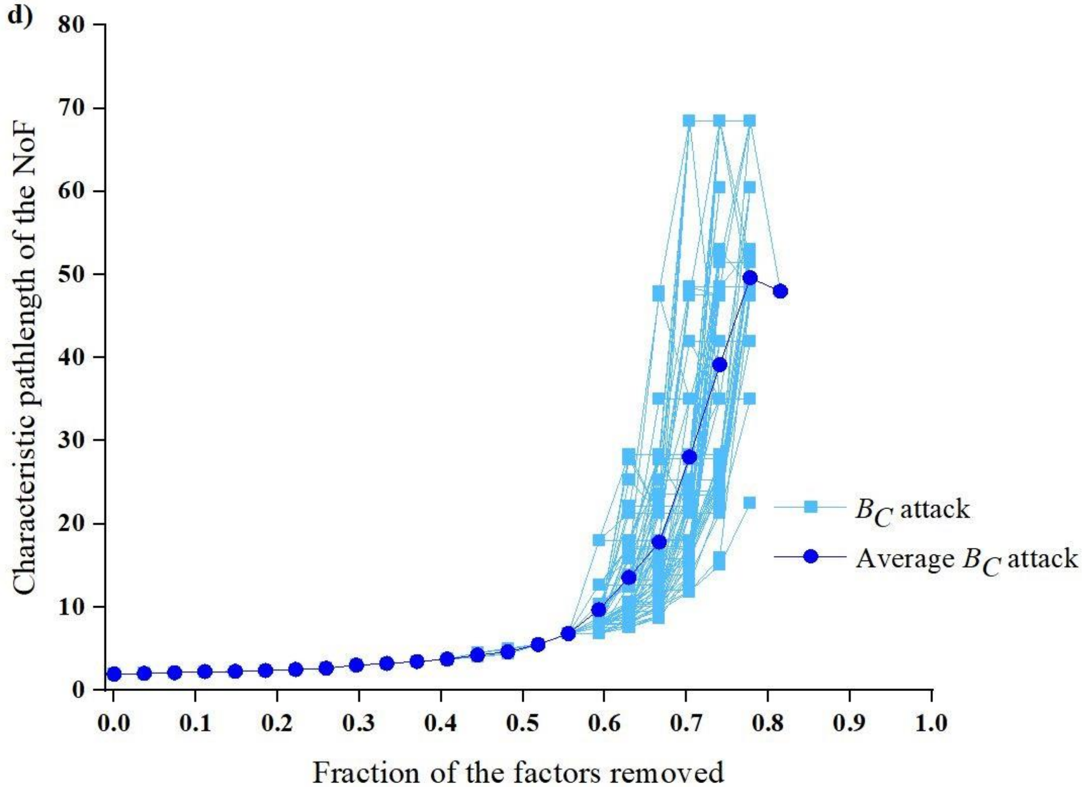


Fig. B. 1. Variation in the network characteristics such as a) characteristic pathlength; and b) clustering coefficient during 200 random node removal trials and their average during the removal of each factor




b)
Characteristic pathlength of the NoF
Fraction of factors removed
OD_C attack
Average OD_C attack


c)
Characteristic pathlength of the NoF
Fraction of factors removed
C_C attack
Average C_C attack



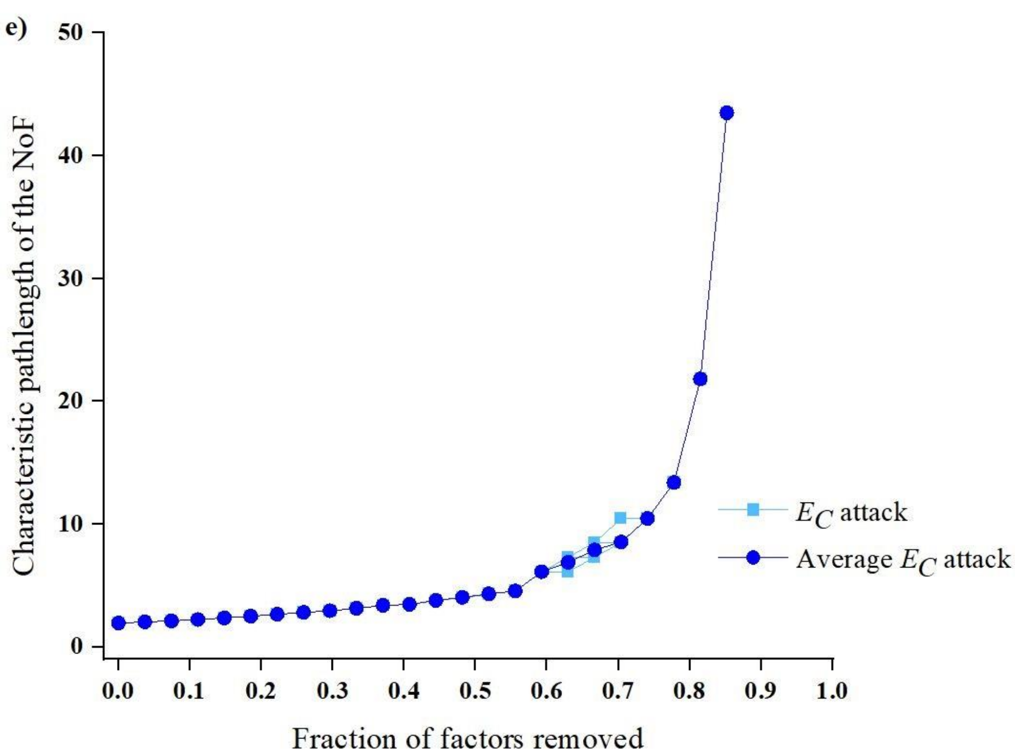
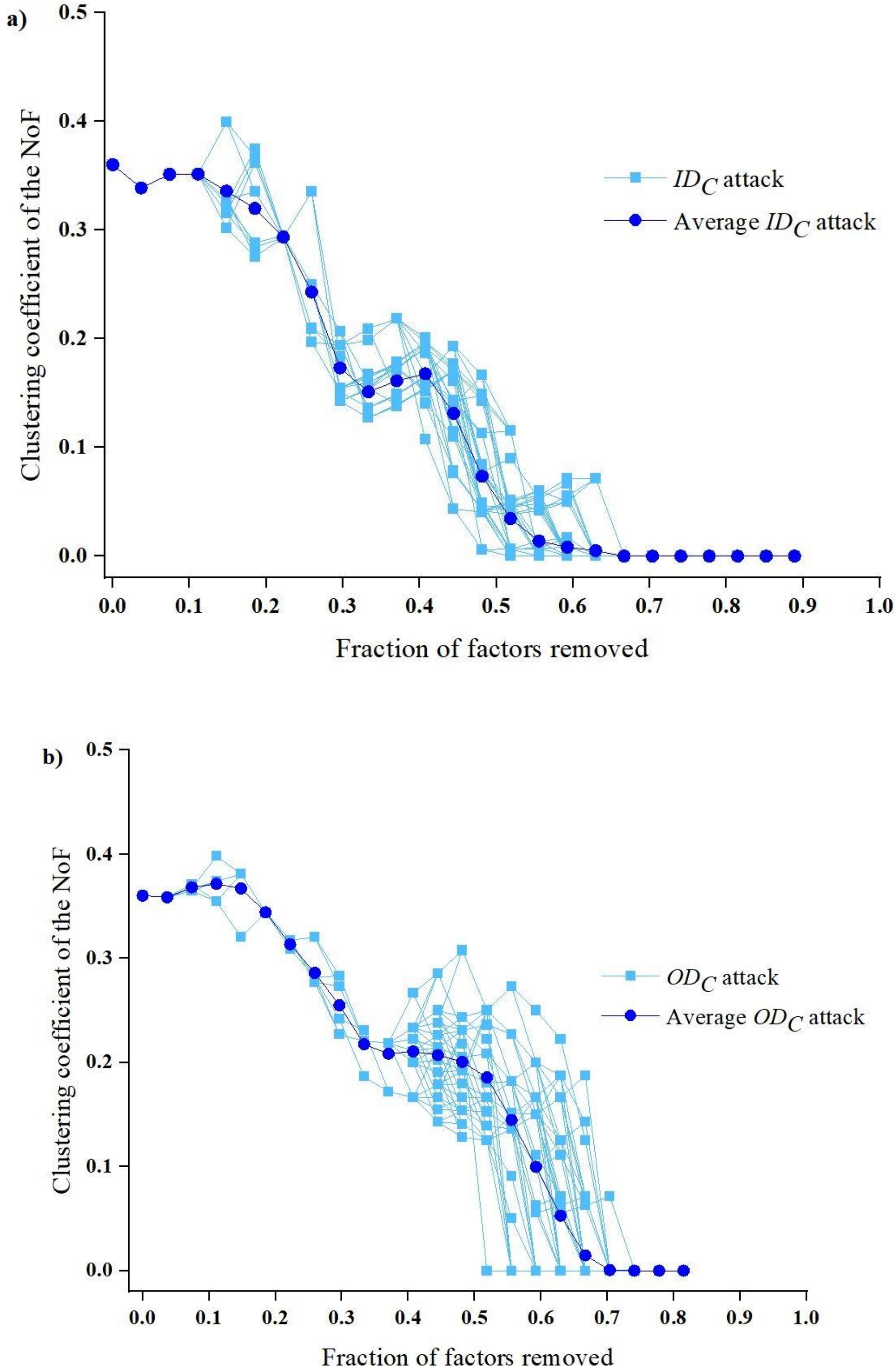
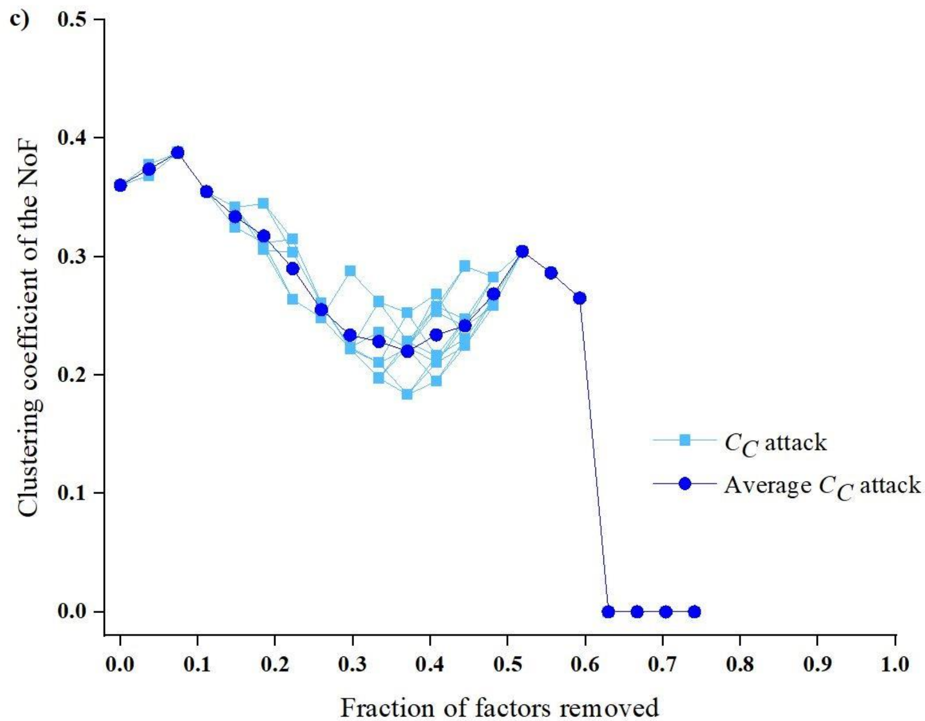
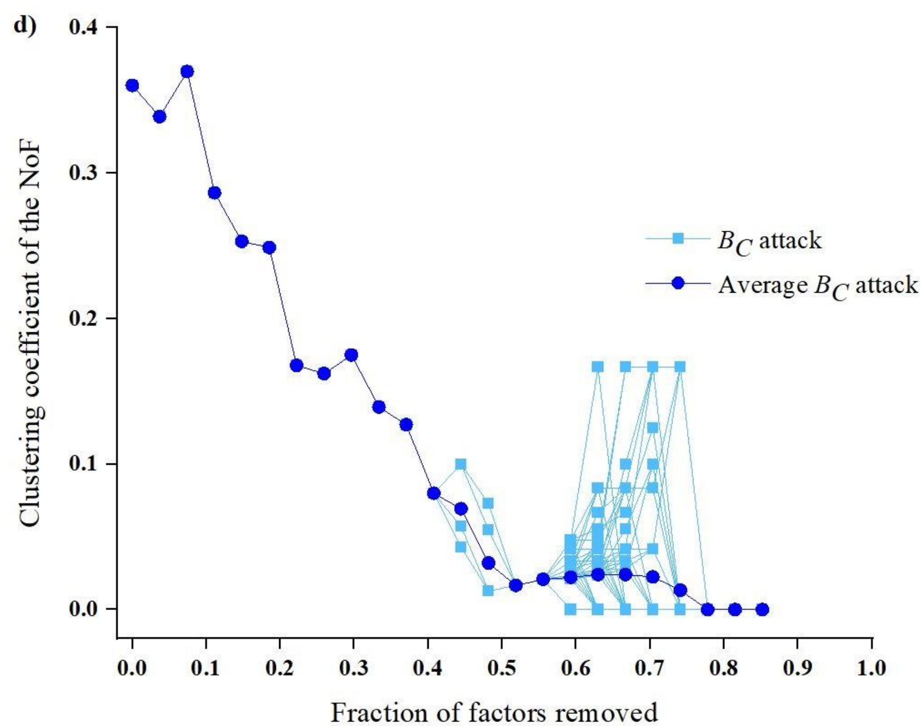


Fig. B. 2. Variation in the network's characteristic pathlength (d) due to removal of factors based on a)ID_C; b)OD_C; c)C_C; d)B_C; and e)E_C


a)
Clustering coefficient of the NoF
Fraction of factors removed
ID_C attack
Average ID_C attack
b)
Clustering coefficient of the NoF
Fraction of factors removed
OD_C attack
Average OD_C attack


c)
Clustering coefficient of the NoF
Fraction of factors removed
C_C attack
Average C_C attack
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0


d)
Clustering coefficient of the NoF
Fraction of factors removed
B_C attack
Average B_C attack
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0

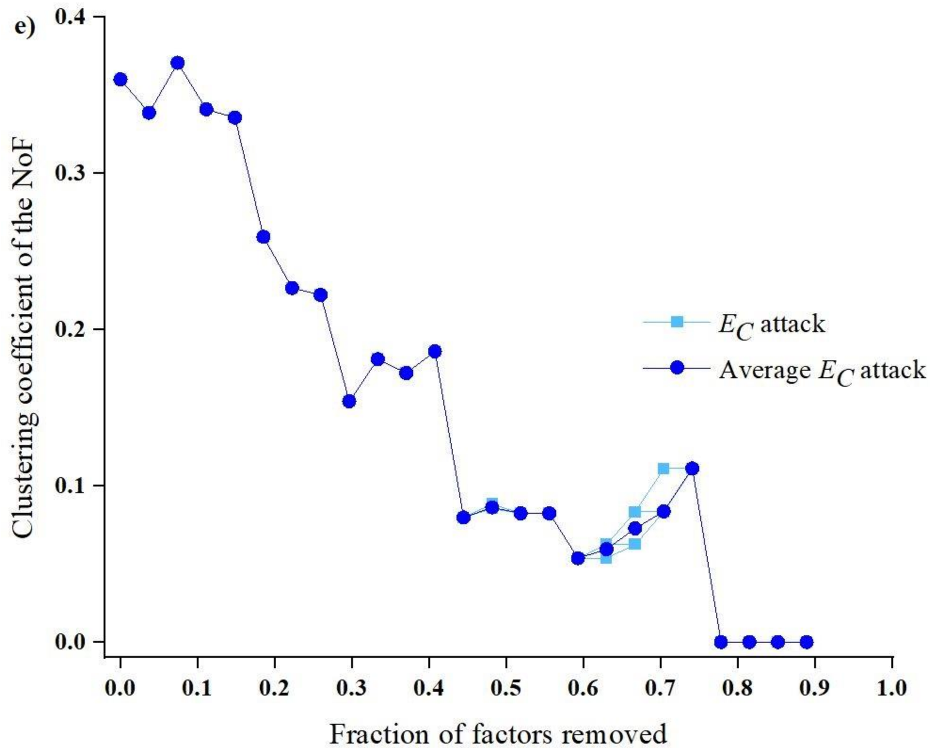


Fig. B. 3. Variation in the network's clustering coefficient (Ccoe) due to removal of factors based on a)ID_C; b)OD_C; c)C_C; d)B_C; and e)E_C

4. Conclusion

Uranium mine tailings dams (UMTD), containing the waste by-products of uranium ore mining and milling process (i.e., tailings), are of major environmental concern in the nuclear energy industry. The presence of residual uranium-238, thorium-230, and radium-226 attributes to the radioactivity of uranium mine tailings, which is equal to 85% of the radioactivity of the unprocessed ore. UMTD failures under unusual weather conditions such as heavy rainfall can lead to the distribution of radionuclides both above and below the ground surface. Seepage of radionuclide plume and its subsequent migration through the unsaturated zone located immediately below the ground surface is a phenomenon of interest requiring more attention. Hence, a mapping of literature describing the current state of knowledge in radionuclide migration in the unsaturated zone was performed using scoping review methodology.

Works of literature accumulated from scoping review described several environmental factors (i.e., chemical, and biological characteristics of soil stratigraphy, groundwater, and radionuclide plume, meteorological, and hydrogeological) influencing radionuclide behavior and migration mechanisms in the subsurface. In order to visualize the interdependence existing between these factors and their collective influence, and to identify dominant factors among them, a complex network theory (CNT) approach was applied. A network of factors (NoF) comprising of 27 factors connected by 144 links was visualized for an integrated subsurface radionuclide fate and transport model. Results from the CNT approach are summarized as follows:

- The formation of clusters between environmental factors (i.e., degree of saturation, clay fraction in the soil, fraction of organic carbon, infiltration rate and transmissivity of subsurface, and particulates in groundwater) indicate their collective influence on radionuclide migration. The collective influence should be reflected while developing radionuclide fate and transport models.

- Soil microorganisms, particulates concentration, and redox potential of groundwater are significantly influenced by other factors in the NoF, hence will experience the most uncertainty as it propagates through the model.

- Precipitation (i.e., rainfall), fraction of organic carbon, transmissivity, porosity, clay fraction, and cation exchange capacity (CEC) of stratigraphy influence most other factors in the NoF through their shorter pathlengths either directly or indirectly. Hence, any uncertainty associated with these factors will rapidly propagate throughout the model.

- Soil microorganisms and infiltration rate of the vadose zone are major connectors, as they are present in a greater number of shortest paths between any two factors in the NoF. Hence uncertainty in them will be amplified and progress to the factors present downstream of shortest paths leading to a misrepresented model.

- Results from sensitivity analysis showed that the removal of factors from the NoF based on their descending B_C values (i.e., removal of factors such as soil microorganisms, infiltration rate, fraction of organic carbon, transmissivity, and tailings microorganisms) made the NoF more vulnerable. Hence, evaluation of these factors should be performed more carefully to minimize the propagation of their uncertainty to an integrated subsurface radionuclide fate and transport model.

NoF includes unweighted and directed links, which is a limitation in this study. This limitation can be eradicated by assigning weights to the links based on the evaluation and representation of relationships between the factors using mathematical and empirical expressions. Scoping review provides information on areas that can be focussed. More studies combining different soil stratigraphy (i.e., sediment and fractured rock medium) under different saturation conditions should be initiated in the future. Inclusion of more experimental studies on field-scale should also be considered. Some of the dominant factors listed in the CNT approach were not frequently studied as indicated by the literature accumulated from scoping review. Hence, it is noted that these factors (i.e., microorganisms present in soil and tailings, fraction of organic carbon, particulates in groundwater, infiltration rate, and degree of saturation) require more detailed study in the future about their influence on radionuclide migration.

Building a reliable subsurface radionuclide fate and transport model requires a large amount of data on crucial environmental factors affecting the process to minimize uncertainty. Identifying the dominant factors in the NoF aids us in prioritizing and allocating resources for thorough data acquisition of the factors that have a major influence on the subsurface radionuclide transport. Accurate and reliable radionuclide transport models are imperative to systemize management procedures and remedial measures for the prevention of groundwater contamination in the event of UMTD failure.

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Appendix T1 – Title and Abstract screening questionnaire[2]

1) Does the article/report identify any information or present any method to understand the transport of radionuclides in the unsaturated zone?
   a) Yes, it provides a methodology to understand radionuclide transport in the unsaturated zone.
   b) Yes, it provides a methodology to understand radionuclide transport in saturated zone along with/without considering unsaturated zone.
   c) No, none of the above.
   d) Cannot decide.
2) Is it an article/report that reviews other studies with an objective to understand the transport of radionuclides in the unsaturated zone?
   a) Yes, it is a review article/report correlating the studies related to transport of radionuclides in the unsaturated zone.
   b) No, it does not deal with the transport of radionuclides in the unsaturated zone.
   c) Cannot decide.

[2] Main Research Question: What is the current state of knowledge on the radionuclides transport in the unsaturated zone?

If the answer is 'Yes' to either Question 1 or 2, the article/report will be included in further screening process. If the answer is 'Cannot decide' for either of the questions, include the article/report for the next stage of appraisal.

Appendix T2

Table T2. 1. Data charted from full-article review

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Phosphate controls uranium release from acidic waste-weathered Hanford sediments	RS[3]	Exp[4]	Lab	Flow-through columns	N/A	Uranium	UZ	SS[5]	N/A	Phosphate ions	Mineral dissolution;
Reduced-order modeling of near-field THMC coupled processes for nuclear waste repositories in shale	RS	Mod[6]	N/A	N/A	N/A	Radionuclides	Both UZ and SZ	BB[7]	FR[8]	N/A	Migration
A Case study on the soil-water characteristics of the vadose zone and the migration of intermediate-level nuclides in a planned spent-fuel reprocessing plant site	RS	Mod	N/A	N/A	N/A	Tritium; Cobalt; Cesium	UZ	SS	N/A	Infiltrating rainwater	Migration
Uranyl oxalate species in high ionic strength environments: stability constants for aqueous and solid uranyl oxalate complexes	RS	Mod	N/A	N/A	N/A	Uranium	N/A	FR	FR	Ionic strength	Complexation
Solid-liquid distribution coefficients (Kd-s) of geological deposits at the Chernobyl Nuclear Power Plant site with respect to Sr, Cs and Pu radionuclides: A short review	RV[9]	N/A	N/A	N/A	N/A	Strontium; Cesium; Plutonium	N/A	SS	SS	N/A	Sorption
Evaluation of gaseous substrates for microbial immobilization of contaminant mixtures in unsaturated subsurface sediments	RS	Exp	Lab	Column leaching	N/A	Uranium; Technetium	UZ	SS	N/A	Microbes	Reduction; Migration/retardation
Integral transform analysis of radionuclide transport in variably saturated media using a physical non-equilibrium model: Application to solid waste leaching at a uranium mining installation	RS	Mod	N/A[10]	N/A	N/A	Uranium	UZ	SS	N/A	Infiltration and recharge rates; Initial radionuclide concentrations	Advection-dispersion
Simulation of the migration of3h and14c radionuclides on the 2nd phase facility at the wolsong lilw disposal center	RS	Mod	N/A	N/A	N/A	Tritium; Radiocarbon	Both UZ and SZ	SS	SS	Integrity of the engineered barriers	Advection; Diffusion
Impact of Natural Organic Matter on Plutonium Vadose Zone Migration from an NH4Pu(V)O2CO3(s) Source	RS	Exp	Lab and Field	Batch adsorption and leaching	Lysimeter	Plutonium	UZ	SS	N/A	Natural Organic Matter (NOM)	Migration; Reduction
Fundamental Study on Transport Model for Radionuclides under unsaturated Condition around Near-Surface Underground	RS	Exp and Mod	Lab	Column leaching	N/A	General	UZ	SS	N/A	Water-saturation; Permeability	Advection-dispersion
Combining geostatistics and simulations of flow and transport to characterize contamination within the unsaturated zone	RS	Mod	N/A	N/A	N/A	Tritium	UZ	SS	N/A	Hydraulic parameters	Advection-dispersion

[3] RS - Research study
[4] Exp – Experimental
[5] SS – Soil sediments
[6] Mod - Modeling
[7] BB – Bentonite Buffer
[8] FR – Fractured rocks
[9] RV - Review
[10] N/A – Not Applicable

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Field experiments of surface water to groundwater recharge to characterize the mobility of uranium and vanadium at a former mill tailing site	RS	Exp	Field	N/A	Injection-extraction into wells	Uranium; Vanadium	Both UZ and SZ	SS	SS	Recharge; Flooding events	Mobilization; Sorption; Desorption
Dissolution-desorption dynamics of strontium during elution following evaporation: pH and ionic strength effects	RS	Exp and Mod	Lab	Flow-through columns	N/A	Strontium	UZ	SS	N/A	water content; pH; ionic strength; concentration	Desorption
Iodine immobilization by materials through sorption and redox-driven processes: A literature review	RV	N/A	N/A	N/A	N/A	Radioiodine	N/A	N/A	N/A	Organic matter; Calcite; Apatite; Iron oxides;	Precipitation; sorption; redox reaction
Deriving probabilistic soil distribution coefficients (Kd). Part 3: Reducing variability of americium Kd best estimates using soil properties and chemical and geological material analogues	RS	Meta-analysis of data	N/A	N/A	N/A	Americium	N/A	SS	SS	pH, organic matter, texture; experimental method	Sorption; Distribution coefficient
Deriving probabilistic soil distribution coefficients (Kd). Part 1: General approach to decreasing and describing variability and example using uranium Kd values	RS	Meta-analysis of data	N/A	N/A	N/A	Uranium	N/A	SS	SS	pH, organic matter, texture; experimental method	Sorption; Distribution coefficient
Deriving probabilistic soil distribution coefficients (Kd). Part 2: Reducing caesium Kd uncertainty by accounting for experimental approach and soil properties	RS	Meta-analysis of data	N/A	N/A	N/A	Cesium	N/A	SS	SS	Organic matter, texture; Experimental approach	Sorption; Distribution coefficient
Speciation of Uranium and Plutonium From Nuclear Legacy Sites to the Environment: A Mini Review	RV	N/A	N/A	N/A	N/A	Uranium; Plutonium	N/A	SS	SS	N/A	Migration; Speciation
Uranium storage mechanisms in wet-dry redox cycled sediments	RS	Exp	Lab	Sequential extraction; X-Ray Absorption spectroscopy	N/A	Uranium	UZ	SS	N/A	Water table fluctuations; Redox conditions	Adsorption; Complexation; Redox; Precipitation
A multiple lines of evidence approach for identifying geologic heterogeneities in conceptual site models for performance assessments	RS	Mod	N/A	N/A	N/A	Technetium	UZ	SS	N/A	Small-scale Geologic heterogeneities	Migration
Climate change impact on residual contaminants under sustainable remediation	RS	Mod	N/A	N/A	N/A	Tritium	Both UZ and SZ	SS	SS	Recharge; Drainage; infiltrating rainwater	Advection-dominated transport
Environmental behaviour of radioactive particles from Chernobyl	RS	Mod	N/A	N/A	N/A	Strontium; Europium;Cesium; Americium; Plutonium	UZ	SS	N/A	N/A	Dissolution/ Weathering
Determination of diffusion coefficient of Cs-137 at unsaturated zone of DH-2 site soil under delta=1.41 g.cm(-3) condition	RS	Exp	Lab	Diffusion column - Gravitation method	N/A	Cesium	UZ	SS	N/A	Initial concentration; Soil density; Sorption efficiency	Diffusion
Numerical evaluation of unsaturated-zone flow and transport pathways at Rainier Mesa, Nevada	RS	Mod	N/A	N/A	N/A	Tritium	UZ	FR	N/A	Infiltration; Layered/faulted materials; Water table; matrix in the fractures	Migration; dispersion; diffusion; decay

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Impact of spatial variability in hydraulic parameters on plume migration within unsaturated surficial formations	RS	Mod	N/A	N/A	N/A	Tritium	UZ	SS	N/A	Hydraulic conductivity; Air-entry value; Pore-size distribution	Advection-dispersion
Investigation of 3 H, 99 Tc, and 90 Sr transport in fractured rock and the effects of fracture-filling/coating material at LILW disposal facility	RS	Exp	Lab	Batch adsorption; Flow-through column; Batch diffusion	N/A	Strontium, Technetium; Tritium	UZ	FR	N/A	Fracture filling/coating - Zeolite	Sorption; Diffusion; Retardation
Plutonium binding affinity to sediments increases with contact time	RS	Exp and Mod	Lab	Batch adsorption/ desorption	N/A	Plutonium	UZ	SS	N/A	Contact time b/w plutonium and sediments	Adsorption; Desorption
Determination of iodine mobility in the soil vadose zone using long-term column experiments	RS	Exp	Lab	Flow-through column	N/A	Radioiodine	Both UZ and SZ	SS	SS	Capillary forces; DOM in groundwater	Migration; Remobilization
Modeling of radionuclide transport in porous media: A review of recent studies	RV	N/A	N/A	N/A	N/A	General	N/A	SS	SS	N/A	Advection; dispersion; diffusion; sorption
A review of the behavior of radioiodine in the subsurface at two DOE sites	RV	N/A	N/A	N/A	N/A	Radioiodine	N/A	SS	SS	Iodine concentration; redox potential; pH; organic matter; iron and manganese minerals; microbes	Complexation; Precipitation; Adsorption/desorption; Oxidation/reduction
Mobility of Aqueous and Colloidal Neptunium Species in Field Lysimeter Experiments	RS	Exp	Field	N/A	RADFLEx Field-scale Lysimeter; High-purity germanium detector; Batch desorption	Neptunium	UZ	SS	N/A	Colloids; infiltrating rainfall	Oxidation/Reduction; Sorption
Reassessment of the Goiânia radioactive waste repository in Brazil using HYDRUS-1D	RS	Mod	N/A	N/A	N/A	Cesium	UZ	SS	N/A	Preferential flow; Concrete liner degradation	Advection; Dispersion; Adsorption;
Uranium speciation in acid waste-weathered sediments: The role of aging and phosphate amendments	RS	Exp and Mod	Lab	Batch reaction	N/A	Uranium	UZ	SS	N/A	Phosphate	Precipitation; Dissolution
Modeling the sensitivity of hydrogeological parameters associated with leaching of uranium transport in an unsaturated porous medium	RS	Mod	N/A	N/A	N/A	Uranium; Thorium; Radium; Lead; Bismuth; Polonium	UZ	SS	N/A	Soil texture; moisture content; hydraulic conductivity;	Advection; Dispersion; Adsorption; Decay
Uranium(VI) adsorption and surface complexation modeling onto vadose sediments from the Savannah River Site	RS	Exp and Mod	Lab	Batch adsorption	N/A	Uranium	UZ	SS	N/A	Minerals - Kaolinite (Clay), Goethite (Clay), Quartz	Complexation; Adsorption

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Numerical and Experimental investigations of cesium and strontium sorption and transport in agricultural soils	RS	Exp and Mod	Lab and Field	Batch adsorption	Lysimeter	Cesium; Strontium	UZ	SS	N/A	Organic matter; CEC; Temperature; Saturation	Complexation; Adsorption
Neptunium(V) sorption to vadose zone sediments: Reversible, not readily reducible, and predictable based on Fe-oxide content	RS	Exp and Mod	Lab	Batch adsorption; Flow-cell experiment	N/A	Neptunium	UZ	SS	N/A	Natural organic matter; Iron oxide; Redox potential	Sorption; Oxidation/Reduction; Complexation;
One-dimensional spatial distributions of Gamma-ray emitting contaminants in field lysimeters using a collimated gamma-ray spectroscopy system	RS	Exp	Lab	RADFLEx Field-scale Lysimeter; High-purity germanium detector	N/A	Cesium; Cobalt; Barium; Europium	UZ	SS	N/A	Cementitious solid-waste forms	Adsorption; Diffusion
Coupling of unsaturated zone and saturated zone in radionuclide transport simulations	RS	Mod	N/A	N/A	N/A	Cesium	Both UZ and SZ	SS	SS	Water flux	Advection; Dispersion; Diffusion; Sorption
Uranium Release from Acidic Weathered Hanford Sediments: Single-Pass Flow-Through and Column Experiments	RS	Exp	Lab	Single-pass Flow-through and Column leaching	N/A	Uranium	Both UZ and SZ	SS	SS	Phosphate	Speciation; Precipitation; Dissolution
Radionuclide transport model for risk evaluation of high-level radioactive waste in Northwestern China	RS	Mod	N/A	N/A	N/A	Strontium; Cesium; Uranium and Plutonium	Both UZ and SZ	FR	FR	Permeability; Distribution and Diffusion coefficient	Advection; Diffusion; Adsorption; Decay
Computation of Saturation Dependence of Effective Diffusion Coefficient in unsaturated Argillite Micro-fracture by Lattice Boltzmann Method	RS	Mod	N/A	N/A	N/A	General	UZ	FR	N/A	N/A	Diffusion
Soil–Radionuclide Interaction under Varied Experimental Conditions	RS	Exp	Lab	Batch adsorption	N/A	Cesium; Strontium; Cobalt	UZ	SS	N/A	Contact time; pH; Liquid to solid ratio; CEC	Sorption; Distribution coefficient
Elements of complexity in subsurface modeling, exemplified with three case studies	RV	Mod	N/A	N/A	N/A	Uranium	N/A	N/A	N/A	N/A	Advection; Dispersion; Adsorption
Evaluation of deep vadose zone contaminant flux into groundwater: Approach and case study	RS	Mod	N/A	N/A	N/A	Technetium	UZ	SS	N/A	Recharge	Advection; Diffusion
Geological and physicochemical controls of the spatial distribution of partition coefficients for radionuclides (Sr-90, Cs-137, Co-60, Pu-239,240 and Am-241) at a site of nuclear reactors and radioactive waste disposal (St. Petersburg region, Russian Federation)	RS	Exp	Lab	Batch adsorption	N/A	Strontium; Cesium; Cobalt; Plutonium; Americium	Both UZ and SZ	SS	SS	Hydroxide and oxides of iron and magnesium; Clay minerals	Sorption; Complexation; ion-exchange
Application of vadose zone approach for prediction of radionuclide transfer from near-surface disposal facility	RS	Mod	N/A	N/A	N/A	Tritium; Radiocarbon	UZ	SS	N/A	Moisture content	Advection; Dispersion; Adsorption; Diffusion

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Role of the vadose zone in mitigating strontium transport at the near-surface disposal facility (NSDF) in Kalpakkam, India	RS	Mod	N/A	N/A	N/A	Strontium	UZ	SS	N/A	Moisture content	Distribution coefficient; Diffusion
Evaluation of distribution coefficient of vadose zone soil from proposed near-surface disposal facility at Kalpakkam, India	RS	Exp	Lab	Batch adsorption; Flow-through column	N/A	Strontium	UZ	SS	N/A	Cations	Sorption
Radionuclide removal by apatite	RV	N/A	N/A	N/A	N/A	Radionuclides	N/A	FR	FR	Cations	Sorption; Precipitation; ion-exchange
Quantifying particulate and colloidal release of radionuclides in waste-weathered Hanford sediments	RS	Exp	Lab	Column leaching	N/A	Strontium; Cesium; Iodine	UZ	SS	N/A	pH; Ionic strength; Colloids; particulates	Weathering; Speciation
pH-dependent reactive transport of uranium(VI) in unsaturated sand	RS	Exp and Mod	Lab	Flow-through Column	N/A	Uranium	UZ	SS	N/A	pH; Solution chemistry; Heterogeneous flow regime	Adsorption; Desorption; Precipitation; Complexation
Uranium fate in Hanford sediment altered by simulated acid waste solutions	RS	Exp and Mod	Lab	Batch adsorption - BenchTop, GloveBox	N/A	Uranium	UZ	SS	N/A	pH;	Adsorption; Surface complexation; Precipitation
Refining the site conceptual model at a former uranium mill site in Riverton, Wyoming, USA	RS	Mod	N/A	N/A	N/A	Uranium	Both UZ and SZ	SS	FR	Organic-rich sediments; Minerals	Evapotranspiration
Radioiodine sorption/desorption and speciation transformation by subsurface sediments from the Hanford Site	RS	Exp	Lab	Air-sealed uptake; Open air uptake; Desorption	N/A	Iodine	Both UZ and SZ	SS	SS	Organic carbon; Sediment texture	Sorption; Desorption; Redox conditions
Effects of dissolved organic matter on the co-transport of mineral colloids and sorptive contaminants	RS	Exp	Lab	Batch adsorption; Flow-through Column	N/A	Cesium	Both UZ and SZ	SS	SS	Organic matter; Clay minerals	Sorption; Desorption
The atmospheric transport of iodine-129 from Fukushima to British Columbia, Canada and its deposition and transport into groundwater	RS	Mod	N/A	N/A	N/A	Iodine	UZ	SS	N/A	Infiltrating rainwater; Organic matter	Migration; Sorption; Retardation
Integrated watershed modeling for simulation of spatiotemporal redistribution of post-fallout radionuclides: Application in radiocesium fate and transport processes derived from the Fukushima accidents	RS	Mod	N/A	N/A	N/A	Cesium	Both UZ and SZ	SS	SS		Adsorption
Actinides sorption onto hematite: experimental data, surface complexation modeling and linear free energy relationship	RS	Exp and Mod	Lab	Batch adsorption	N/A	Americium; Thorium; Uranium; Neptunium	N/A	SS	N/A	Colloids	Sorption; Surface complexation
Modelling the behaviour of uranium-series radionuclides in soils and plants taking into account seasonal variations in soil hydrology	RS	Mod	N/A	N/A	N/A	Uranium; Radium	UZ	SS	N/A	SS; Plants	Sorption; Uptake by plants; decay;

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Colloid-facilitated mobilization of metals by freeze-thaw cycles	RS	Exp	Lab	Pilot-scale - Rainfall simulator, Soil core and sampling grid	N/A	Cesium; Strontium	UZ	FR	N/A	Temperature; Rainfall; Colloids	Sorption;
Transport modeling in performance assessments for the Yucca Mountain disposal system for spent nuclear fuel and high-level radioactive waste	RV	Mod	N/A	N/A	N/A	General	N/A	Both SS and FR	Both SS and FR	N/A	Advection; Dispersion; Diffusion; Sorption
Unsaturated flow modeling in performance assessments for the Yucca Mountain disposal system for spent nuclear fuel and high-level radioactive waste	RV	Mod	N/A	N/A	N/A	General	N/A	Both SS and FR	Both SS and FR	N/A	Advection; Dispersion; Diffusion; Sorption
Mineral transformation controls speciation and pore-fluid transmission of contaminants in waste-weathered Hanford sediments	RS	Exp	Lab	Flow-through column experiments	N/A	Cesium; Strontium; Iodine	UZ	SS	N/A	Recharge; Drainage	Sorption; Desorption; Precipitation; Ion-exchange
Recent developments in assessment of long-term radionuclide behavior in the geosphere-biosphere subsystem	RV	Mod	N/A	N/A	N/A	Radionuclides	N/A	N/A	N/A	N/A	N/A
Insights into transport velocity of colloid-associated plutonium relative to tritium in porous media	RS	Exp	Lab	Flow-through column experiments	N/A	Plutonium; Tritium; Strontium	UZ	SS	N/A	Natural colloids	Sorption; Migration; Colloid-facilitated transport
A high-performance workflow system for subsurface simulation	RS	Mod	N/A	N/A	N/A	Technetium	UZ	SS	N/A	N/A	Migration
Geochemical and mineralogical investigation of uranium in multi-element contaminated organic-rich subsurface sediment	RS	Exp	Lab	Monitoring; SEM; Energy Dispersive Spectrometry (EDS);	N/A	Uranium	UZ	SS	N/A	Naturally reduced zones; Iron oxides; Clays	Sorption; Redox potential
Influence of temperature decrease and soil drought on the geochemical fractionation of 60Co and 137Cs in fluvisol and cambisol soils	RS	Exp	Lab	Extraction experiments	N/A	Cobalt; Cesium	UZ	SS	N/A	Infiltrating rainfall; Temperature	Sorption; Mobilization
Influence of alkaline co-contaminants on technetium mobility in vadose zone sediments	RS	Exp	Lab	Batch and Flow-through column experiment	N/A	Technetium	UZ	SS	N/A	Alkaline waste	Precipitation; Oxidation/ Reduction
Guidelines for thermodynamic sorption modelling in the context of radioactive waste disposal	RV	Mod	N/A	N/A	N/A	General	N/A	N/A	N/A	N/A	Sorption
Establishing a geochemical heterogeneity model for a contaminated vadose zone--aquifer system.	RS	Exp	Lab	Extraction experiments; Desorption	N/A	Uranium	UZ	SS	N/A	Spatial heterogeneity; Water table fluctuation	Sorption; Desorption; Complexation;
An Alternative Process Model of Preferential Contaminant Travel Times in the Unsaturated Zone: Application to Rainier Mesa and Shoshone Mountain, Nevada	RS	Mod	N/A	N/A	N/A	Radionuclide	UZ	FR	N/A	Preferential flow paths	Advection; Dispersion; Recharge

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Influence of acidic and alkaline waste solution properties on uranium migration in subsurface sediments	RS	Exp	Lab	Batch and Column experiment	N/A	Uranium	UZ	SS	N/A	pH; Acidic; Alkaline	Precipitation; Sorption; Complexation
Perched-water analysis related to deep vadose zone contaminant transport and impact to groundwater	RS	Mod	N/A	N/A	N/A	Technetium and Uranium	UZ	SS	N/A	Perched water	Recharge; Drainage
Environmental Impact Assessment of Liquid Waste Ponds in Uranium Milling Installations	RS	Mod	N/A	N/A	N/A	Uranium; Thorium; Radium	Both UZ and SZ	SS	SS	N/A	Advection; Dispersion; decay; Sorption
Plutonium Transport in the Environment	RV	N/A	N/A	N/A	N/A	Plutonium	N/A	Both SS and FR	Both SS and FR	Moisture content; Organic matter; Microbes; Inorganic colloids	Migration; Sorption
Characterizing particle-scale equilibrium adsorption and kinetics of uranium(VI) desorption from U-contaminated sediments	RS	Exp and Mod	Lab	Flow through reactors	N/A	Uranium	UZ	SS	N/A	N/A	Sorption; Desorption; Complexation
Effect of water content on strontium retardation factor and distribution coefficient in Chinese loess	RS	Exp	Lab	Column experiments	N/A	Strontium	UZ	SS	N/A	Moisture content; CEC	Sorption
Identifying key controls on the behavior of an acidic-U(VI) plume in the Savannah River Site using reactive transport modeling	RS	Mod	N/A	N/A	N/A	Uranium; Tritium	Both UZ and SZ	SS	SS	Mineral surfaces; pH	Precipitation; Sorption; Complexation
Transport of Europium Colloids in Vadose Zone Lysimeters at the Semiarid Hanford Site	RS	Exp	Field	N/A	Lysimeter	Europium	UZ	SS	N/A	Rainwater; Colloids	Migration
Dual permeability variably saturated flow and contaminant transport modeling of a nuclear waste repository with capillary barrier protection	RS	Mod	N/A	N/A	N/A	General	UZ	FR	N/A	Infiltration; Drainage	Dispersion
Colloid-associated plutonium transport in the vadose zone sediments at Lop Nor	RS	Exp	Lab	Column experiment	N/A	Plutonium	UZ	SS	N/A	Infiltration intensity; Particle size distribution; Ionic strengths; Water content	Sorption;
Source Term Evaluation Model for Uranium Tailings Ponds.	RS	Mod	N/A	N/A	N/A	Uranium; Thorium; Radium; Plutonium; Americium; Lead	UZ	SS	N/A	Infiltrating rainwater; Moisture content	Migration
Monte Carlo simulation of radionuclide migration in fractured rock for the performance assessment of radioactive waste repositories	RS	Mod	N/A	N/A	N/A	Plutonium	UZ	FR	N/A	Fracture-matrix interactions	Advection; Dispersion; Retardation
Environmental Speciation of Actinides	RV	N/A	N/A	N/A	N/A	Neptunium; Plutonium; Thorium; Uranium	N/A	N/A	N/A	Inorganic minerals; Organic matter; Microorganisms	Redox condition
Scale-dependent rates of uranyl surface complexation reaction in sediments	RS	Exp and Mod	Lab	Stirred flow-cell; Flow-through columns	N/A	Uranium	UZ	SS	N/A	Experiment scale	Advection; Diffusion; Sorption; Complexation

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Assessment of the Resistance to Uranium (VI) Exposure by Arthrobacter sp. Isolated from Hanford Site Soil	RS	Exp	Lab	Microbial culture; Atomic Force Microscopy	N/A	Uranium	UZ	SS	N/A	Arthrobacter strains, G975, G968, andG954; redox potential	Precipitation; Complexation; Reduction
Radionuclide migration through an unsaturated clay buffer under thermal and hydraulic gradients for a nuclear waste repository	RS	Exp and Mod	Lab	KENTEX injection phase experiment	N/A	Iodine; Cesium	N/A	BB	N/A	Temperature; Saturation	Diffusion; Sorption; Decay
Changes in the pore network structure of Hanford sediment after reaction with caustic tank wastes	RS	Exp	Lab	Flow-through column; X-Ray computed microtomograp hy	N/A	Strontium; Cesium; Uranium; Radioiodine	UZ	SS	N/A	Precipitates	Dissolution; Precipitation
Quantitative 3-D Elemental Mapping by LA-ICP-MS of a Basaltic Clast from the Hanford 300 Area, Washington, USA	RS	Exp	Lab	Laser ablation - Inductively coupled mass spectroscopy	N/A	Uranium	UZ	SS	N/A	N/A	Sorption
Using high performance computing to understand roles of labile and nonlabile uranium(VI) on Hanford 300 area plume longevity	RS	Mod	N/A	N/A	N/A	Uranium	UZ	SS	N/A	N/A	Complexation; Sorption; Diffusion; Dispersion
An experimental study of diffusivity of technetium-99 in Hanford vadose zone sediments	RS	Exp	Lab	Half-cell diffusion	N/A	Technetium	UZ	SS	N/A	Metallic iron; Moisture content	Diffusion
Simulation of radionuclide transport through unsaturated, fractured rock: Application to Yucca Mountain, Nevada	RS	Mod	N/A	N/A	N/A	Technetium; Plutonium; Neptunium	UZ	FR	N/A	Infiltration; Sorption parameters; Colloids	Advection; Dispersion; Diffusion; Sorption
Radionuclide migration at Experimental polygon at Red Forest waste site in Chernobyl zone. Part 2: Hydrogeological characterization and groundwater transport modeling	RS	Exp and Mod	Lab and Field	Batch adsorption	In-situ sampling	Strontium	Both UZ and SZ	SS	SS	Infiltrating rainwater; moisture content	Advection-Dispersion; Sorption
Numerical modeling of the radionuclide water pathway with HYDRUS and comparison with the IAEA model of SR 44	RS	Mod	N/A	N/A	N/A	Uranium; Technetium; Strontium; Plutonium; Tritium; Iodine; Cobalt; Carbon;	UZ	SS	N/A	Infiltrating rainwater	Dispersion; Sorption;
Radionuclide transport in the unsaturated zone at Yucca Mountain, Nevada	RS	Mod	N/A	N/A	N/A	Uranium; Thorium; Radium; Strontium; Cesium; Technetium; Iodine; Carbon	UZ	FR	N/A	Colloids; infiltration	Advection; Dispersion; Diffusion; Sorption
The kinetic stability of colloid-associated plutonium: Settling characteristics and species transformation	RS	Exp	Lab	Flow-through column	N/A	Plutonium	UZ	SS	N/A	Cations; pH	Sorption; Desorption

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Investigations of the Fundamental Surface Reactions Involved in the Sorption and Desorption of Radionuclides	RS	Exp and Mod	Lab	Batch adsorption; Batch desorption; Flow-through column experiment	N/A	Lead	N/A	SS	SS	pH; Temperature; Initial concentration	Adsorption; Complexation
An integrated study of uranyl mineral dissolution processes: etch pit formation, effects of cations in solution, and secondary precipitation	RS	Exp	Lab	In-situ(fluid cell) dissolution; Batch dissolution	N/A	Uranium	N/A	SS	N/A	pH; Cations; Secondary surface precipitates	Dissolution-precipitation
Interfacial reactivity of radionuclides: emerging paradigms from molecular-level observations	RV	N/A	Field	N/A	Analytical probes	Cesium; Uranium	N/A	SS	SS	N/A	Oxidation; Reduction; Ion-exchange
Stochastic simulation of uranium migration at the Hanford 300 Area	RS	Mod	N/A	N/A	N/A	Uranium	Both UZ and SZ	SS	SS	Permeability	Precipitation; Complexation
Investigations of Near-Field Thermal-Hydrologic-Mechanical-Chemical Models for Radioactive Waste Disposal in Clay/Shale Rock	RS	Mod	N/A	N/A	N/A	General	UZ	FR	N/A	N/A	Advection; Diffusion
Summary of Uranium Solubility Studies in Concrete Waste Forms and Vadose Zone Environments	RV	N/A	N/A	N/A	N/A	Uranium	UZ	SS	N/A	N/A	Solubility
Conceptual and numerical models of solute diffusion around a HLW repository in clay	RS	Mod	N/A	N/A	N/A	Uranium, Cesium, Radio-chlorine, Tritium	UZ	BB	N/A	N/A	Diffusion; Sorption
Downward migration of Chernobyl-derived radionuclides in soils in Poland and Sweden	RS	Mod	N/A	N/A	N/A	Cesium; Plutonium	UZ	SS	N/A	Organic matter; Clay minerals	Advection; Dispersion; Diffusion; Sorption
137Cs: Parametric analysis of the Darcy velocity influence on the contaminant concentration at receptor well	RS	Mod	N/A	N/A	N/A	Cesium	Both UZ and SZ	SS	SS	Darcy's velocity	Advection; Dispersion
Sequestration and remobilization of radioiodine (129I) by soil organic matter and possible consequences of the remedial action at savannah river site	RS	Exp	Lab	Soil resuspension experiment	N/A	Radioiodine	UZ	SS	N/A	Soil organic matter; pH; Colloids; Humic acid; Manganese and Iron Oxides	Precipitation;
Strontium and cesium release mechanisms during unsaturated flow through waste-weathered Hanford sediments	RS	Exp and Mod	Lab	Column leaching	N/A	Strontium; Cesium	UZ	SS	N/A	Cations; Initial concentration	Ion-exchange;
Mobilization of actinides by dissolved organic compounds at the Nevada Test Site	RS	Exp	Lab	Batch adsorption	N/A	Americium; Plutonium; Neptunium; Uranium	N/A	FR	FR	Dissolved Organic Matter	Sorption; Complexation; Oxidation
Influence of contact time on the extraction of (233)uranyl spike and contaminant uranium from Hanford Site sediment	RS	Exp	Lab	Batch extractions; Column leaching extractions	N/A	Uranium	N/A	SS	N/A	Contact time between uranium and sediments; Extractants	Sorption; Desorption

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Quantifying Differences in the Impact of Variable Chemistry on Equilibrium Uranium(VI) Adsorption Properties of Aquifer Sediments	RS	Exp and Mod	Lab	Equilibrium adsorption	N/A	Uranium	N/A	SS	SS	Cations; Seasonal flushing	Surface complexation; Desorption
Impact of phosphate on U(VI) immobilization in the presence of goethite	RS	Exp and Mod	Lab	Batch reaction	N/A	Uranium	N/A	SS	SS	Phosphate; Goethite	Complexation; Precipitation; Sorption
2D FEM analysis for coupled thermo-hydro-mechanical-migratory processes in near field of hypothetical nuclear waste repository	RS	Mod	N/A	N/A	N/A	General	Both UZ and SZ	BB	FR	Temperature field; Radionuclide decay	Diffusion; Dispersion; Sorption
Upward movement of plutonium to surface sediments during an 11-year field study	RS	Exp and Mod	Field	N/A	Lysimeter	Plutonium	UZ	SS	N/A	Root uptake by Surface plants	Advection; Dispersion; Sorption
Field-scale model for the natural attenuation of uranium at the Hanford 300 Area using high-performance computing	RS	Exp and Mod	Lab	Flow-through column	N/A	Uranium	Both UZ and SZ	SS	SS	River fluctuations	Precipitation; Complexation; Sorption
Resupply mechanism to a contaminated aquifer: A laboratory study of U(VI) desorption from capillary fringe sediments	RS	Exp	Lab	Batch adsorption and desorption; Flow-through column	N/A	Uranium	UZ	SS	N/A	Recharge; Drainage; Moisture content	Sorption; Desorption; Complexation
Simple estimation of fastest preferential contaminant travel times in the unsaturated zone: Application to Rainier Mesa and Shoshone Mountain, Nevada	RS	Mod	N/A	N/A	N/A	General	UZ	FR	N/A	Preferential flow	Advection-dispersion
The effects of preferential flow and soil texture on risk assessments of a NORM waste disposal site	RS	Mod	N/A	N/A	N/A	Uranium; Thorium; Radium; Lead	UZ	SS		Preferential flow; soil texture	Advection; Dispersion; Sorption
Modelling of a large-scale in-situ migration experiment with 14C-labelled natural organic matter in Boom Clay	RS	Exp and Mod	Field	N/A	Piezometers	Carbon labeled NOM	UZ	SS	N/A	NOM	Advection; Diffusion; Sorption
Uranium-series constraints on radionuclide transport and groundwater flow at the nopal I uranium deposit, Sierra Peña Blanca, Mexico	RS	Exp	Lab	Mass spectroscopy	N/A	Uranium; Thorium; Radium	Both UZ and SZ	FR	FR	N/A	Precipitation; Sorption
Characterization of uranium-contaminated sediments from beneath a nuclear waste storage tank from Hanford, Washington: Implications for contaminant transport and fate	RS	Exp	Lab	Isotope exchange technique; Batch Desorption	N/A	Uranium	UZ	SS	N/A	N/A	Sorption; Desorption; Precipitation
Mineralization of contaminant uranium and leach rates in sediments from Hanford, Washington	RS	Exp	Lab	Column leaching	N/A	Uranium	UZ	SS	N/A	N/A	Sorption; Precipitation
Bedrock Kd data and uncertainty assessment for application in SR-Site geosphere transport calculations	RV	N/A	N/A	N/A	N/A	Radionuclides	N/A	N/A	N/A	pH; ionic strength; redox potential; CEC; Speciation; Temperature;	Sorption
Influence of iron redox transformations on plutonium sorption to sediments	RS	Exp	Lab	Batch adsorption	N/A	Plutonium	UZ	SS	N/A	Iron oxides; Iron phyllosilicates	Sorption; Oxidaation/reduction

Title of article or report	Type	Method	Exp type	Type of lab studies	Type of field studies	Radionuclides discussed	Zone studied	UZ medium	SZ medium	Factors	Mechanisms discussed
Conceptual model for radionuclide release, Yucca Mountain, Nevada, USA.	RS	Mod	N/A	N/A	N/A	General	UZ	FR	N/A	Heat release	Advection; Dispersion
Safety implication for an unsaturated zone nuclear waste repository	RS	Mod	N/A	N/A	N/A	General	UZ	SS	N/A	Temperature	Diffusion
Factors influencing plutonium sorption in shale media	RS	Exp	Lab	Batch sorption experiment	N/A	Plutonium	Both UZ and SZ	FR	FR	Grain size; pH; water/solid ratio; temperature; ionic strength	Sorption
Colloid-facilitated transport of cesium in vadose-zone sediments: The importance of flow transients	RS	Exp	Lab	Flow-through column	N/A	Cesium	UZ	SS	N/A	Transient flow condition; Colloids	Sorption
Diffusion experiments for estimating radiocesium and radiostrontium sorption in unsaturated soils from Spain: Comparison with batch sorption data	RS	Exp	Lab	Half-cell (Diffusion tube)	N/A	Cesium; Strontium	UZ	SS	N/A	Clay minerals; Organic Matter; Organic carbon; CEC; Tortuosity	Sorption; Diffusion
Distributions of radionuclide sorption coefficients (K d) in sub-surface sediments and the implications for transport calculations	RS	Exp and Mod	Lab	Gamma spectroscopy	N/A	Cobalt; Cesium; Strontium	Both UZ and SZ	SS	SS	Clay content; Soil texture; CEC; mineral oxides	Sorption
Cation Exchange on Vadose Zone Research Park Subsurface Sediment, Idaho National Lab	RS	Exp and Mod	Lab	Batch adsorption	N/A	Strontium	UZ	SS	N/A	CEC; Ionic strength; pH	Sorption; Ion-exchange
Modeling reactive transport of strontium-90 in a heterogeneous, variably saturated subsurface	RS	Mod	N/A	N/A	N/A	Strontium	UZ	Both SS and FR	N/A	CEC; Recharge	Advection; Dispersion; Precipitation