Chapter 1
Sources and Health Risks of Rare Earth Elements in Waters

Willis Gwenzi, Nyarai M. Mupatsi, Munyaradzi Mtisi, and Allan A. Mungazi

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Abstract Anthropogenic rare earth elements widely used in high-technology applications are prevalent in the aquatic environment, thus constituting emerging contaminants. Yet reviews on the anthropogenic sources, behavior, and potential health risks of rare earth elements remain limited. The current chapter seeks to (1) highlight anthropogenic sources, behavior, and human intake pathways of rare earth elements, (2) discuss the human and ecological health and exposure risks of rare earth

W. Gwenzi (✉) · N. M. Mupatsi · M. Mtisi · A. A. Mungazi
Biosystems and Environmental Engineering Research Group, Department of Soil Science and Agricultural Engineering, Faculty of Agriculture, University of Zimbabwe, Harare, Zimbabwe
e-mail: wgwenzi@agric.uz.ac.zw

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elements, (3) present a conceptual outline for assessing and mitigating health risks, and (4) identify the key thematic areas for further research.

Anthropogenic hotspot sources of rare earth elements include wastes and wastewaters from medical facilities, rare earth elements mining and mineral processing, high-technology electrical and electronic industries, petroleum refineries, rare earth elements-enriched fertilizers and livestock feeds, and recycling plants for postconsumer electronic and electrical goods. The dissemination of rare earth elements from sources into the various environmental compartments is controlled by anthropogenic (industrial discharges) and hydrological processes. Human exposure occurs via occupational inhalation in rare earth elements-based industries, ingestion of contaminated food, and medical applications including magnetic resonace imaging. To date, evidence exists documenting rare earth elements in human body parts including the brain, hair, nails, milk, serum, and sperms. High concentrations of rare earth elements reduce plant growth and nutritional quality, impaired biochemical function in plants, and induce neurotoxicity, acute and chronic health effects, and genotoxicity in aquatic animals. The uptake, partitioning, and bioaccumulation of rare earth elements may also occur along the trophic levels in aquatic ecosystems. Human health risks include (1) severe damage to nephrological systems and nephrogenic systemic fibrosis induced by gadolinium-based contrast agents used in medical applications, (2) induced sterility and anti-testicular effects in males, (3) dysfunctional neurological disorder and reduced intelligent quotient, (3) fibrotic tissue injury, (4) pneumoconiosis, and (5) oxidative stress and cytotoxicity. In developing countries, the health risks of rare earth elements may be considerably high due to the following: (1) weak and poorly enforced environmental and public health regulations, (2) overreliance on untreated drinking water, and (3) lack of human health surveillance systems for early detection and treatment of human health effects. However, limited empirical data exist to establish the relationship between rare earth elements in the aquatic environments and their health effects. A conceptual outline for assessing and mitigating the health risks and thematic areas for further research were highlighted.

Keywords Anthropogenic sources · Ecological effects · Ecotoxicology · Environmental reservoirs · Human health effects · Human intake pathways · Lanthanides

Acronyms

Ce	Cerium
$CeCl_3$	Cerium trichloride
Cu	Copper
Dy	Dysprosium
Er	Erbium
Eu	Europium
Gd	Gadolinium

Ho	Holmium
La	Lanthanum
$LaCl_3$	Lanthanum trichloride
Lu	Lutetium
Nd	Neodymium
P680	Photosystem II, one of the photosynthetic reaction centers
Pm	Promethium
Pr	Praseodymium
REE(s)	Rare earth elements
Sc	Scandium
Sm	Samarium
Tb	Terbium
Tm	Thulium
Y	Yttrium
Yb	Ytterbium
Zn	Zinc

1.1 Introduction

Rare earth elements consist of 17 elements, which include 15 lanthanides and 2 other elements (scandium, Sc, and yttrium, Y) (Li et al. 2013a, b). The 15 lanthanides are lanthanum (La), cerium (Ce), dysprosium (Dy), europium (Eu), gadolinium (Gd), lutetium (Lu), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), terbium (Tb), holmium (Ho), erbium (Er), thulium (Tm), and ytterbium (Yb). Yttrium (Y) and scandium (Sc) exhibit physicochemical properties and behavior similar to that of lanthanides, hence are included among rare earth elements. Promethium, a radioactive element, has no naturally occurring long-lived and stable isotope; thus, it is excluded in environmental studies (Hu et al. 2006). Thus, the current chapter only focuses on lanthanides (lanthanum to lutetium) and yttrium and scandium. Although the term "rare earth elements" is used in this chapter for consistency with other literature, the term is a misnomer because the crystal abundances of rare earth elements are often comparable to, and in some cases even higher than, those of common metals such as silver and gold (Wedepohl 1995; Schüler et al. 2011; Henderson 2013). In this context, "rare" implies that rare earth elements occur in very low concentrations but are highly dispersed in most geological systems compared to common elements such as metals.

The unique properties of rare earth elements, including permanent magnetism and high reactivity, make them ideal for high-technology applications including electrical and electronic devices and equipment, advanced ammunition systems and platforms, renewable energy, and medical applications. The increasing industrial applications of rare earth elements could lead to a corresponding increase in the release of rare earth elements into the environments via industrial wastes and wastewaters, thus raising human and ecological health concerns. An increasing body of literature has reported anthropogenic rare earth elements in aquatic systems in various countries (Hatje et al. 2016; Hissler et al. 2016). Anthropogenic rare earth elements, specifically gadolinium, have been used as transient chemical tracers for anthropogenic pollution in aquatic systems (Brünjes et al. 2016). Fraum et al. (2017) and Rogowska et al. (2018) present recent reviews on gadolinium and gadolinium contrast agents, while a review on the occurrence, behavior and fate, and health risks is presented in one of the current author's papers (Gwenzi et al. 2018). Thus, rare earth elements can be considered as among emerging contaminants posing human and ecological health risks. Yet the ecotoxicology, mechanisms of action, and epidemiology of emerging contaminants are still poorly understood; thus, emerging contaminants remain unregulated (Petrović et al. 2003). In addition, until now, appropriate analytical methods were unavailable to detect the occurrence of emerging contaminants in the environment due to very low concentrations (Petrović et al. 2003; Gwenzi et al. 2018).

Until recently, literature on emerging contaminants was dominated by harmful biological agents including antimicrobial resistance genes and the host antimicrobial-resistant microbes and synthetic organic contaminants particularly industrial solvents, fire retardants, illicit drugs, food additives and colorants, pharmaceuticals, and personal care products (Petrie et al. 2015; Gwenzi and Chaukura 2018). However, recent studies including one review by the current author have drawn attention to rare earth elements as emerging contaminants of human and ecological health concerns (Gwenzi et al. 2018). Rare earth elements qualify to be classified among emerging contaminants for the following reasons: (1) maximum guideline limits to safeguard human and ecological health are nonexistent, rendering the regulation of rare earth elements problematic (Kulaksız and Bau 2013); (2) routine environmental and human health monitoring and surveillance systems often exclude rare earth elements; (3) the concentrations in environmental media are often extremely low (ng/mL to μg/ml), which can only detected using highly sensitive advanced analytical equipment; and (4) the ecotoxicology and human toxicity including modes of actions and epidemiology, which are important for formulating maximum guideline limits, are still poorly understood.

Recent field studies and reviews have documented the occurrence and health risks of rare earth elements of anthropogenic origin in aquatic systems (Tepe et al. 2014; Hatje et al. 2016; Gwenzi et al. 2018). These studies further point that rare earth elements are a new group of contaminants, which is currently overlooked by policy makers, the public, and researchers. The purpose of this chapter is to present an overview of anthropogenic sources, behavior and fate, and potential health risks associated with rare earth elements. Figure 1.1 presents an overview of the focus of the chapter.

The specific objectives of the current chapter are to (1) highlight anthropogenic sources, behavior, and human intake pathways of rare earth elements, (2) discuss the human and ecological health and exposure risks of rare earth elements, (3) present a

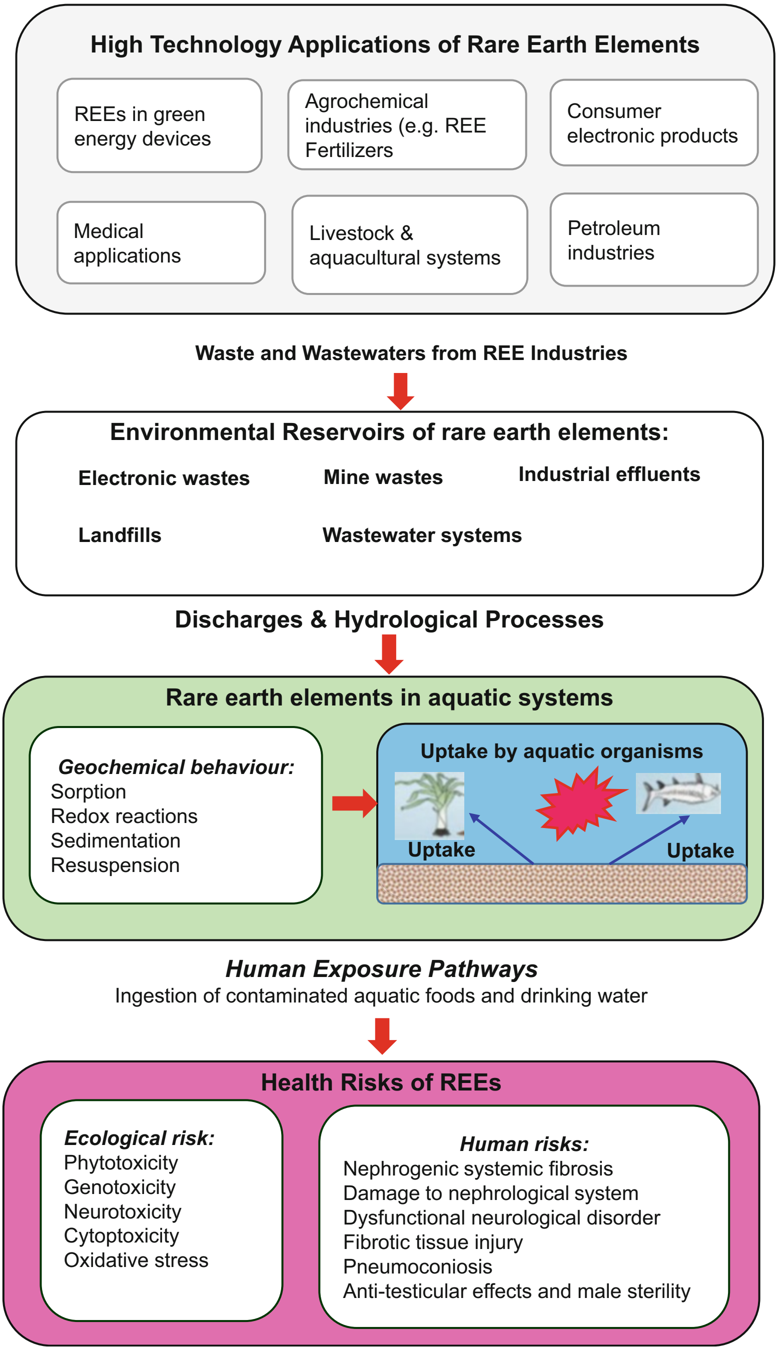

Fig. 1.1 Summary depiction of the high-technology applications, environmental reservoirs, behavior, and health risks of rare earth elements in aquatic systems (modified after Gwenzi et al. 2018). REE(s), rare earth element(s); MRI, magnetic resonace imaging

conceptual outline for assessing and mitigating health risks, and (4) identify the key thematic areas for further research.

1.2 Rare Earth Elements

1.2.1 Global Occurrence and Production of Rare Earth Elements

Rare earth elements occur in geological systems and undergo mobilization and are transported as a coherent group; thus, the geogenic concentrations of rare earth elements exhibit the characteristic saw-tooth behavior (Gupta et al. 2014). Rare earth elements are highly reactive; hence, they do not occur as pure elements or uninterrupted ore bodies, but exist within other host mineral ores (Charalampides et al. 2015). The predominant rare earth oxides are xenotime, monazite, and bastnasite (Humphries 2012), while less common ones are gadolinite, allanite, ancylite, euxenite, parisite, lanthanite, yttrotungstite, yttrotantalite, stillwellite, fergusonite, samarskite, yttrialite, loparite, chevkinite, cerite, britholite, fluocerite, and cerianite (Haque et al. 2014). Rare earth elements may also coexist with base metals in mineral ores.

The recovery of rare earth elements from the host ores is achieved through costly extractive and metallurgical processes that can release the rare earth elements from mineral ores and subsequently isolate them from the complex solution of various elements. Rare earth oxide reserves exist in Australia, Brazil, the Dominican Republic, the United States, Russia, and several African countries, including South Africa, Burundi, Kenya, and the Democratic Republic of the Congo, among others (Zhanheng 2011; Massari and Ruberti 2013). Globally, China has nearly half (48%) of the total reserves of rare earth elements, followed by the United States (12%), Commonwealth States (17%), India (3%), and then Australia (1%), while other countries including those in Africa account for the remainder 19% (United States of America Geological Survey 2010).

The global supply chain of rare earth elements is dominated by China, which accounts for 98% of the total production, while other countries contribute the remainder (2%) (Alonso et al. 2012). Mining of rare earth elements in China started around 1990 and since then has played a dominant role in the rare earth elements' supply chain, and several countries including the United States are wholly dependent on imports from China (United States of America Geological Survey 2008; Stone 2009). The global consumption of rare earth elements in 2015 was about 119,650 metric tons, and the annual growth rate is estimated to rise to 5% in 2020 as the global demand increases (Zhou et al. 2017). Rare earth elements are regarded as critical resources for high-technology applications because of their strategic and economic importance, coupled with the high risk of the global supply chain (Du and Graedel 2011; Graedel et al. 2015). Substantial literature exists on the global occurrence, production, and applications of rare earth elements in high-technology engineering (Du and Graedel 2011; Massari and Ruberti 2013). Recent advances in high-technology applications have witnessed an upsurge in mining, production, and industrial applications of rare earth elements.

1.2.2 Properties and High-Technology Applications of Rare Earth Elements

Rare earth elements and their compounds exhibit unique physicochemical properties, which are critical for their high-technology applications. These properties include the following: (1) rare earth elements with odd atomic numbers have lower relative abundances than those with an even ones, a phenomenon referred to as the "Oddo–Harkins rule" (Binnemans et al. 2013a, b); (2) unique electronic configuration, which accounts for the high reactivity of rare earth elements; thus, they readily react with several nonmetallic elements such as sulfur, oxygen, and hydrogen, thereby forming ionic complexes of rare earth elements with high coordination number often exceeding 6 and in some cases 12 (Tang and Johannesson 2003; Nockemann et al. 2006); (3) an increase in atomic number is accompanied by a decrease in the cationic radius, a phenomenon termed "lanthanide contraction" (Ramos et al. 2016); and (4) rare earth elements are considered as soft elements, which are malleable and ductile, with excellent catalytic, chemical, electrical, and permanent magnetism and optical properties including high luminescence (Redling 2006; Gai et al. 2013).

Rare earth elements are essential components of high-technology electronic and engineered applications (Kulaksız and Bau 2011a, b). In summary, Table 1.1 shows that rare earth elements are used in various industrial applications, including (1) miniaturized electronic devices and appliances including mobile phones, (2) advanced weapon systems and platforms, (3) solar panels and wind turbines for renewable energy, (4) electronic and electronic applications including conductors and supercapacitors, and (5) contrast agents in magnetic resonance imaging. According to literature, the proportion of rare earth elements used in various industrial applications decreases in the order: 21% lightweight permanent magnets, followed by 20% catalysts, 18% alloys, 12% powders, and 7% phosphors (Paulick and Machacek, 2017). Specifically, neodymium is a critical component of high-performance neodymium (Nd)–iron (Fe)–boron (B) permanent magnets for generators and motors, while yttrium (Y) is used for the production of superconductors and laser technology (Du and Graedel 2011; Charalampides et al. 2015).

Neodymium, yttrium, and other rare earth elements are also used in laser technology as dopants, while yttrium is also used to develop yttrium–aluminum–garnet lasers (Ancsombe 2002). Moreover, rare earth elements are key components of autocatalysts, lighting and display systems, wind turbines, and solar panels (Du and Graedel 2011; Haque et al. 2014) and are also widely applied in pharmacology and

Table 1.1 An overview of industrial applications of high-technology rare earth elements

Rare earth elements	Industrial applications	References
Ce, Eu, Gd, Tb, tm	Flat-screen displays	Resende and Morais (2010) and Humphries (2012)
Er, Yb, Eu, Y	Optical fibers	Eliseeva and Bünzli (2011)
Ce, La, Nd, Pr, Sc, Yb	Metallurgy	Stegen (2015), Paulick and Machacek (2017)
Dy, Gd, Nd, Pr, Tb, Tm,	Medical imaging	Naumov (2008), Xie et al. (2014), Stegen (2015)
Pr, Nd, Gd, Tb, Dy, Sm	Lightweight strong permanent magnets	Haque et al. (2014), Xie et al. (2014), Charalampides et al. (2015) and Stegen (2015)
La, Ce, Pr, Nd	Auto-catalysts in petroleum refining	Navarro and Zhao (2014)
Ce, Dy, Er, Gd, Ho, La, Pr, Nd, Y,	Ceramics and glass additives	Naumov (2008), Campbell and Keane (2010), United States of America Geological Survey (2014) and Stegen (2015)
Ce, Dy, Eu, Gd, La, Tb, Y	Phosphors	Tan et al. (2015)
Y	Oxygen sensors, medicines and drugs, radar	Townley (2013), Charalampides et al. (2015), Lu et al. (2017)
Er, Ho Nd, Y,	Lasers, superconductors and capacitors	Ancsombe (2002)
Ce, La, Nd, Pr, Sm	Battery alloys	Charalampides et al. (2015)
Y, Sm	Microwave filters	Naumov (2008)
Sc, Tm	Electron beam tubes	Naumov (2008)
Lu, Gd	Crystal scintillators	Naumov (2008)

biomedical applications (Thomsen 2017). For example, gadolinium is a well-known component of gadolinium-based contrast agents that are widely utilized for clinical and diagnostic medical applications including magnetic resonance imaging (Lu et al. 2017). For example, yttrium is utilized in the development of anticancer drugs such as TheraSphere®, which contain microspheres of yttrium-90 (Kulik et al. 2006; Lu et al. 2017). In agriculture, low doses of rare earth elements are used in fertilizers and livestock feeds as crop and livestock growth promoters (He et al. 2008, 2010; Wu et al. 2013). For example, on an annual basis, fertilizers doped with nitrates of lanthanum, neodymium, and cerium are applied to approximately more than six million hectares of agricultural land to increase crop yields and quality (Wang et al. 2001; Migaszewski and Gałuszka 2015a, b). The increase in mining, production, and subsequent applications of rare earth elements in various high-technology systems could be accompanied by increased release of rare earth elements in industrial wastes and wastewaters. In fact, the increased occurrence of rare earth elements in aquatic environment has been attributed to the increased production and applications of rare earth elements in high-technology applications (Klaver et al. 2014; Khan et al. 2016).

1.3 Occurrence and Behavior of Rare Earth Elements in Aquatic Systems

1.3.1 Anthropogenic Sources

Although the current chapter focuses on aquatic systems, it is noteworthy that such aquatic systems are connected to other environmental compartments (contaminated soils, tailings, wastewaters) through material flows and hydrological processes. Thus, the connectivity between aquatic systems and the contiguous environmental compartments may form a continuum of rare earth contamination. Accordingly, the concentrations of rare earth elements in mine wastes such as tailings (Li et al. 2010) and other rare earth reservoirs (Sloof 1995) exceed the baseline concentrations in the earth crust by several orders of magnitude. Once in the environment, hydrological processes (e.g., runoff) and anthropogenic activities (e.g., wastewater and effluent discharges) play a key role in the dissemination of rare earth elements into aquatic environments.

To date, anthropogenic rare earth elements have been detected in aquatic environments in several countries such as Australia, Europe, the United Kingdom, and North America, with gadolinium being the most documented rare earth elements (Bau et al. 2006; Kulaksız and Bau 2011a, b; Hatje et al. 2016). Table 1.2 presents an overview of some of the rare earth elements detected in aquatic systems. Several anthropogenic sources emit rare earth elements into solid waste repositories (landfills, non-engineered waste dumps, mine waste rock dumps, and tailings dams and wastewater treatment plants, which in turn act as rare earth reservoirs). Specific anthropogenic hotspot sources of rare earth elements include (1) mining and processing operations of rare earth elements (Paulick and Machacek 2017; Victoria et al. 2017); (2) hospitals and clinical and diagnostic medical imaging facilities (Lawrence et al. 2009; Lawrence, 2010; Kulaksız and Bau 2011a, b; Möller et al. 2011, 2014); (3) pharmaceutical and drug industries (Hutchinson et al. 2004); (4) rare earth elements-enriched livestock feeds and fertilizers applied to croplands and aquacultural systems (Wang et al. 2001; Redling 2006; He et al. 2010; Migaszewski and Gałuszka 2015a, b); (5) recycling plants for waste electronic and electrical equipment such as computer monitors, television screens, and fluorescent tubes (Resende and Morais 2010; Erfort et al. 2017); and (6) petroleum refineries where rare earth elements are used in catalysts (Kulaksız and Bau 2013). A review of the anthropogenic sources of rare earth elements is presented in an earlier paper (Gwenzi et al. 2018).

Table 1.2 Nature and sources of rare earth elements reported in various aquatic systems

Rare earth elements	Source	Aquatic system	References
Eu	Fiber optics	Sedimentary rocks, bed sediments, seawater rivers in the Chambal River (1.296 μg/g) and Deccan Trap basalts	Allègre et al. (1996) and Kümmerer and Helmers (2000)
Gd	Gadopentetic acid, Gd $(DTPA)_2^-$, which is used in magnetic resonance imaging	Oceans, river water, and seawater in Yamuna River at a concentration of 4.8 μg/g	Knappe et al. (1999)
La	Metallurgy	Oceans, river water, and seawater in Yamuna River (39.6 μg/g)	Knappe et al. (1999)
Heavier rare earth elements		Zircon and garnet in Yamuna River (14.6 μg/g)	Byrne and Kim (1990)
Lu	Crystal scintillators	Sediments such as quartzites, zircon, monazite, and water in Kali Sindh (0.48 μg/g) and Chambal River (0.38 μg/g)	Johannesson et al. (1995) and Aubert et al. (2001)
Ce	Polishing powders and metallurgy	Bed sediments in Yamuna and Chambal rivers and fresh water, granites in Hanuman, Chatti, and Deccan Trap basalts (76.6 μg/g)	Braun et al. (1990), Sholkovitz (1995) and Dalai et al. (2004)

1.3.2 Dissemination and Behavior of Rare Earth Elements in Aquatic Systems

Rare earth elements may be disseminated into aquatic systems as a coherent group via hydrological processes and anthropogenic activities (Weltje et al. 2002). Specifically, hydrologically driven processes such as infiltration, groundwater recharge, runoff, and erosion processes mobilize and transfer rare earth elements from hotspot sources into various environmental compartments (Cao et al. 2017). The release of untreated and partially treated effluents and wastewaters from rare earth element industries contributes to the occurrence of rare earth elements in aquatic systems. For example, excessive precipitation and overtopping of tailing dams transfer rare earth elements from catchment to surface aquatic systems through runoff (Khan et al. 2016). Rare earth elements may enter groundwater via three pathways (Keasler and Loveland 1982): (1) mobilization and transport of readily soluble rare earth elements by infiltration and recharge, (2) as rare earth element-contaminated water migrating through soil layers into groundwater, and (3) via surface–groundwater interactions. Some studies report that the loading of rare earth elements in aquatic systems decreases in the order: riverine systems followed by brackish and then seawater (Herrmann et al. 2016). This trend reflects the hydrochemical processes such as dilution and mixing of fresh and salty water in estuaries (Herrmann et al. 2016). In addition, coagulation and subsequent sedimentation of rare earth elements bound to particulate and colloidal materials such as organic matter and iron and manganese oxyhydroxides may also occur.

Gadolinium originating from gadolinium-based contrast agents widely applied in magnetic resonace imaging has been detected in aquatic systems in several developed countries. These include Germany (Kulaksız and Bau 2013), Australia (Lawrence et al. 2009; Lawrence 2010), Switzerland (Vriens et al. 2017), Canada (Macmillan et al. 2017), the United Kingdom (Thomsen 2017), and the United States (Bau et al. 2006; Hatje et al. 2016). Gadolinium contrast agents have a low residence time in the human body; thus, they are rapidly excreted into wastewater conveyance and treatment systems and other aquatic systems. Gadolinium contrast agents are highly stable, and almost inert; thus, they are not effectively removed by most unit operations in wastewater treatment processes, hence the occurrence of gadolinium contrast agents in surface and groundwater systems (Knappe et al. 2005). Rare earth elements have also been reported in drinking water systems including tap water (Kulaksız and Bau 2011a, b; Lindner et al. 2015) and some aquatic organisms such as sea urchins (Merschel et al. 2015). In the Netherlands and Germany, anthropogenic rare earth elements detected in aquatic systems include samarium and lanthanum, which are used as catalytic cracking catalysts in petroleum refineries (Kulaksız and Bau 2013). In the same studies, gadolinium largely occurred in a dissolved form, while samarium and lanthanum were in nanoparticulate or colloidal forms depending on pH and the available ligands (Kulaksız and Bau 2011a, b, 2013). However, currently missing in existing literature are data pertaining to rare earth elements in aquatic systems in developing regions including Africa, Asia, and South America.

Rare earth elements in aquatic systems may exhibit complex behaviors, which are controlled by several biogeochemical conditions. These conditions include type and speciation of the rare earth elements and geochemical conditions including solution pH, ionic strength, ligands, redox potential, natural organic matter content, and the presence of aquatic plants (Johannesson et al. 2004; Wilke et al. 2017). In aquatic environments, rare earth elements may undergo sorption–desorption processes, ion exchange, plant uptake and bioaccumulation, and liquid–solid phase partitioning among colloidal materials such as organic matter and minerals and pore and bulk water and plant uptake (Chakhmouradian and Wall 2012; Verplanck 2013; Klaver et al. 2014). The biogeochemistry of rare earth elements in aquatic systems, including solution chemistry, and complexation processes with both inorganic and organic ligands has been discussed in an earlier papers (Johannesson et al. 2004; Migaszewski and Gałuszka 2015a, b). Mineral phases including manganese and iron oxyhydroxides and organic matter exhibiting high surface areas behave as efficient scavengers for rare earth elements, thus controlling the adsorption–desorption reactions. The adsorption of rare earth elements on solid matrices may also occur, followed by sedimentation and resuspension through bioturbation and hydraulic drift. Rare earth elements may also undergo redox reactions, characterized by highest oxidation states in surface waters containing high concentrations of oxygen, while lower oxidation states may dominate under anoxic conditions in deepwater layers (Weltje et al. 2002). Compared to lighter rare earth elements, heavier rare earth elements have a tendency to form complexes with inorganic anions; thus, they remain in the solution, while chlorine complexes often mobilize the rare earth elements (Williams-Jones et al. 2012).

Aquatic plants may take up rare earth elements, which may then undergo trophic bioaccumulation and biotransformation, phenomena similar to those occurring in soils (Li et al. 2013a, b; Lindner et al. 2013; Khan et al. 2017; Amyot et al. 2017). Plants tend to have a differential uptake of rare earth elements, characterized by a relatively higher affinity for trivalent lanthanides than divalent rare earth elements (Weltje et al. 2002). However, the effects of uptake and bioaccumulation of rare earth elements on plants appear to depend on the type and speciation of the rare earth elements and plant species. One study evaluated the ability of four aquatic plants (*Elodea nuttallii*, *E. canadensis*, *Ceratophyllum demersum*, *Lemna gibba*) to act as biofilters and take up two gadolinium-based contrast agents (i.e., Dotarem, Omniscan) from aqueous systems (Braun et al. 2018). Although the study showed no significant bioaccumulation of gadolinium-based contrast agents, tissue concentration reached at peak between days 1 and 4, before being released back into water (Braun et al. 2018). In the same study, uptake by the four plant species had negligible effect on the removal of gadolinium in water at concentrations investigated (i.e., 1–256 μg/ L). Accordingly, Braun and co-workers concluded that biofiltration by macrophyte species studied had limited capacity to remove gadolinium-based contrast agents in aquatic systems. Overall, the biogeochemical behavior and fate of rare earth elements in aquatic environments are governed by complex processes involving surface chemistry, solution complexation processes, and plant uptake, which in turn, depend on various environmental and biotic factors.

1.3.3 Environmental Health Risks

Ecological Health Risks

Compared to studies documenting the occurrence of rare earth elements in aquatic systems, limited data exist on the effects of rare earth elements on aquatic ecology at species, population, community, and ecosystem levels. However, the few ecotoxicological laboratory studies and data largely drawn from other terrestrial ecosystems in China (e.g., agroecosystems) point to potential ecological effects (Liang et al. 2014; Zhuang et al. 2017a, b). Rare earth elements and their ecological effects have been reported in soils, soil organisms, and plants (Zhang and Shan 2001; Liang et al. 2014) and aquatic systems (Yuan et al. 2003; Trifuoggi et al. 2017). The dominance of literature drawn from China may reflect the country's major role in the production and industrial applications of rare earth elements. The ecotoxicological effects of rare earth elements can be traced back to the late 1940s and 1970s (Burkes and

Table 1.3 Summary ecological health risks of rare earth elements

Rare earth elements	Health risks	Remarks	References
Several rare earth elements	Affects embryo development in zebra fish	Rare earth elements attach in place of Ca^{2+} and affect physiological functions regulating Ca^{2+} in zebra fish	Tang and Johannesson (2003) and Cui et al. (2012)
La^{3+}, Yb^{3+}	Delayed larval development	La^{3+} and Yb^{3+} delayed larval development, reduced hatching and survival rates, and induced tail deformities in zebra fish	Cui et al. (2012)
Yb^{3+}	Induced mortality in goldfish	Reduced activity of enzyme catalase	
Ce^{4+}	Induces mortality and infertility of sea urchin embryo	Sea urchin experience mortality when exposed to Ce^{4+} and sperm development are also affected	Oral et al. (2010), Martino et al. (2017)
Gd	Neurotoxicity in rats	Rats' exposure to different doses of Gd resulted in neuronal death	Xia et al. (2011)
Ce	Reduced life expectancy in fruit flies	Ce reduced the lifespan of *Drosophila melanogaster* at high exposure rates	Huang (2011)
CeO_2	Contamination of food chain	In its nanoparticle form, CeO_2 accumulates in zucchini, crickets, and wolf spiders food chain. This results in trophic transfer and food chain contamination by CeO_2	Hawthorne et al. (2014)
Nd	Decreased growth in wheat and rice	A decreased growth was observed in rice and wheat grown in Nd concentration of 10 and 25 mg/L of Nd	Basu et al. (2016)
Li	Necrosis of older leaves	Death of older leaves was observed in lettuce when tissue concentration above 1000 mg was detected	
La and Ce	Decreased growth in maize	Application of high concentrations of La and Ce reduced growth of maize relative to the control	
La^{3+}	Inhibits seed germination	Pre-soaking of seeds in 1–10 mM of La^{3+} and rare earth element nitrate solution for 4 h inhibited seed germination in plants	D'Aquino et al. (2009)
N, Er, and Y	Reduced seed germination rate	Higher concentrations reduced seed germination of *R. sativus* and *S. lycopersicum*	Carpenter et al. (2015); Thomas et al. (2014)

McCleskey 1947; Gale 1975). The bacteriostatic effects of cerium and lanthanum were first reported as early as 1947 by Burkes and McCleskey (1947). Since then, evidence showing the adverse effects of rare earth elements in both terrestrial and aquatic organisms has been increasing (Table 1.3).

High concentrations of rare earth elements in aquatic systems may have adverse ecological effects (Table 1.3). Rare earth elements have been reported to inhibit plant uptake of essential nutrients such as calcium, an effect attributed to the fact that calcium cationic radius and those of rare earth elements are similar (Hu et al. 2003). In some plant species, rare earth elements also alter the channels associated with the endoplasmic reticulum responsible for calcium release (Klüsener et al. 1995). In turn, the reduced intake of calcium may interfere with the biochemical functions of calcium in plants, including formation of cell walls, root growth, photosynthesis, and flowering. Cerium has similar physical properties as calcium; hence, it is easily absorbed by plants and is more phytotoxic as compared to the rest of rare earth elements. According to Burda et al. (1995), cerium competes with calcium for the same binding sites in P680 or photosystem II, one of the photosynthetic reaction centers. In the symplast of roots, ions of rare earth elements disrupt ionic channels responsible for xylem exudation (Schwenke and Wagner 1992).

Lanthanum reversibly inhibits the phototaxis and photophobic response in some green and blue–green algae via blockage of the calcium pump and reduction of conductivity (Herrmann et al. 2016). Lanthanum also reduces calcium and magnesium concentrations in plants, which in turn reduce the concentrations of carotenoids and chlorophylls a and b while enhancing the activities of antioxidant enzymes (Xu et al. 2012). In addition, at a concentration of 11.1 mg/L, lanthanum is reported to induce alterations of the ultrastructure of cell organelles such as mitochondria, chloroplasts, and nucleus (Xu et al. 2012). The same studies attributed the reduction in chlorophyll synthesis to lanthanum inhibiting the functions of magnesium and calcium. In algae (*Chlorella pyrenoidosa*), which is a common aquatic plant, high concentrations of rare earth elements inhibited both growth and reproduction (Hu et al. 2003). In the same study, the toxicity decreased in the order: neodymium followed by praseodymium, cerium, and lanthanum and then the mixtures of the four rare earth elements. The uptake and bioaccumulation of rare earth elements have documented several aquatic plant species such as water hyacinth, a process also common with metals (Singh and Kalamdhad 2012; Zheng et al. 2016). Other mechanisms accounting for the toxicity of rare earth elements in plants include generation of reactive oxygen species causing oxidative stress and alteration of photosystem II (Pang et al. 2002; Xia et al. 2011). In some plant species, cerium, lanthanum, and praseodymium concentrations exceeding 50 mg L^{-1} inhibit photosynthesis via reduction of the activity of photosystems (Pang et al. 2002).

The toxicity of rare earth elements may also depend on geochemical conditions such as redox potential, pH, ionic strength, and the prevailing ligands. A study conducted by Thomas et al. (2014) on seed germination showed that at low pH, cerium had harmful effects on *A. syriaca*, *P. virgatum*, *R. sativus*, and *S. lycopersicum*, while Y appeared to have phytotoxic effects on *D. canadense* and *S. lycopersicum* at higher doses. However, at high pH, lanthanum and cerium had no significant effect on the percentage germination at all doses, but lanthanum reduced plant biomass relative to the control (Thomas et al. 2014). In the same study, all five plant species that were studied showed that Ce accumulated in both plant shoot and root, with the roots accounting for the most accumulation. Oxygen evolution rate in seeds treated with rare earth elements was higher than in non-treated seeds, indicating greater metabolism. Application of low rates improved root growth and fresh weight in *Eriobotrya japonica*; thus, low rates were found to increase the chlorophyll content and vice versa. Application of lanthanum at high rates was found to have an inhibitory effect. Yb^{3+} and Eu^{3+} inhibit shoot growth and even cause death at high concentrations. Lanthanum was observed to indirectly reduce algal growth by forming an insoluble phosphate precipitate, which reduced plant uptake and growth due to shortage of phosphate (Stauber and Binet 2000). Jin et al. reported that a lanthanum concentration of 7.2 μmol L^{-1} significantly reduced the growth of cyanobacteria and microalgae. One beneficial effect of rare earth elements to plants is that rare earth elements can inhibit bioavailability and uptake of toxic metals possibly via competition. For example, lanthanum inhibits uptake of Pb^{2+} in plants grown in lead-polluted areas. However, the ecological effects of rare earth elements may also depend on other stress factors, plant species, and growth stage.

Rare earth elements may also adversely affect aquatic animals including fish and crustaceans, similar to the ecological effects reported in terrestrial animal bioassay species (e.g., rats, mice (Pagano et al. 2012, 2015). In one study conducted on water fleas (*Daphnia carinata*), lanthanum delayed maturation (Barry and Meehan 2000). Another study reported lack of antagonistic and synergistic effects in fish (carp) exposed to rare earth elements (Qiang et al. 1994). High concentrations of gadolinium in aquatic systems were positively correlated with increased mortality of the daphnids (Perrat et al. 2015). The inhalation of cerium dioxide (CeO_2) nanoparticles obtained from diesel fuel catalysts promoted IFN-γ and IL-12 production by alveolar macrophages (Ma et al. 2014).

Although data pertaining to aquatic animals is limited, the age dependence of the toxicity of rare earth elements has been reported in terrestrial animals. For example, in one study, 200 or 500 mg cerium trichloride ($CeCl_3$)/kg body weight was administered via gavage for adults, milk for neonatal mice, and placenta transfusion for fetal mice (Kawagoe et al. 2008). On the one hand, multiple toxicity effects manifested in adult mice, including neutrophil infiltrations, necrosis, pulmonary venous congestion, pulmonary hemorrhage, thickened alveolar septae, and hepatic. On the other hand, toxicity in fetal mice occurred in the form of hepatic and pulmonary congestion (Kawagoe et al. 2008). The same authors reported that cerium trichloride ($CeCl_3$) administered via gavage resulted in damage of the liver and respiratory system. These effects were more severe in adult mice that in neonatal and fetal mice. In some studies, 40 mg $LaCl_3$/kg body weight administered to rats via gavage caused behavioral changes and promoted enzyme activity of Ca^{2+} ATPase in the hippocampal cells (He et al. 2008). On the contrary, significant decreases were noted in the activities of catalases, glutathione peroxidase, and superoxide dismutase relative to the control (Feng et al. 2006).

Rare earth elements are highly redox reactive; thus, they may induce oxidative stress that damages biomolecules including deoxyribonucleic acid. As reported in plants, oxidative stress has also been reported in bioassay animals subjected to gavage administration of La^{3+}, Ce^{3+}, and Nd^{3+} (Huang 2011). In ecotoxicological studies, oxidative stress is evaluated using endpoints such as lipid peroxidation, concentrations of reactive oxygen species, and changes in antioxidant activities of associated enzymes such as superoxide dismutase, glutathione peroxidase, and catalases (Liang and Wang 2014; Pagano et al. 2015). Some recent studies report that gadolinium originating from gadolinium-based contrast agents is toxic to marine organisms including bivalves and sea urchins (Perrat et al. 2017; Martino et al. 2018; Rogowska et al. 2018). Some ecotoxicological studies show that rare earth elements are characterized by biphasic or hermetic dose–response relationships (Pagano et al. 2015). Such dose–response relationships show beneficial or stimulatory effects at low doses, while toxic or inhibitory effects are observed at high doses (Pagano et al. 2015). This biphasic or hormetic behavior forms the basis for the applications of rare earth elements as growth promoters in plants, aquaculture, and livestock production (Wang et al. 2000). Accordingly, low doses of rare earth elements are used in livestock feeds and fertilizers as substitutes for zinc and copper in the pretext that the use of rare earth elements reduces the health risks of zinc and copper (Redling 2006).

At ecosystem level, studies show that high concentrations of rare earth elements alter ecological functions and reduce biodiversity of insects of the order Dermaptera and family Carabidae (Li et al. 2010). However, some species (e.g., *Stibaropus formosanus*) and genera (e.g., Formicidae) are less sensitive (Li et al. 2010), suggesting that the ecotoxicological effects are species- and genera-dependent. Rare earth elements potentially alter biogeochemical cycling in aquatic systems, but limited data exist on this aspect. Liu and Wang (2001) showed that soil application of rare earth elements at a rate of 5 mg/kg significantly decreased the concentration of available nitrogen, possibly by inhibiting mineralization of nitrogen. This could in turn induce nitrogen deficiency and alter ecosystem function by reducing primary productivity in aquatic systems. Rare earth elements may also pose a radioactivity hazard to aquatic organisms (Akiwumi and D’Angelo 2017). In summary, rare earth elements may cause diverse ecological effects, which depend on speciation, recipient organism and intake route, environmental conditions, nature, and concentration of rare earth elements in aquatic systems.

Human Exposure and Health Risks

Human intake of rare earth elements occurs via medical applications in magnetic resonace imaging and administration of rare earth element-based pharmaceuticals, occupational exposure, and dietary intake (Kanda et al. 2017; Zhuang et al. 2017a, b; Khan et al. 2017). A number of studies have reported rare earth elements and their health risks in the human body including the brain, hair, nails, serum, milk, and sperms (Gomez-Aracena et al. 2006; Wei et al. 2013; Poniedziałek et al. 2017). For example, four rare earth elements (praseodymium, erbium, neodymium, lanthanum) were observed in human milk sampled from women in a hospital in Poznań, Poland (Poniedziałek et al. 2017). Occupational exposure to rare earth elements may occur via inhalation during mining, refining, and industrial production of rare earth elements-based products (Rim et al. 2013; Gambogi 2016). Once in the human body, rare earth elements pose several human health risks (Table 1.4). Human health risks associated with occupational exposure to rare earth elements include

Table 1.4 Summary of human health risks associated with rare earth elements

Rare earth elements	Health risks	Remarks	References
Eu, Dy, Pr	Breast cancer	Rare earth elements were detected in neoplastic cells	Roncati et al. (2018)
Gd, La	Nephrogenic systematic fibrosis	Gd and La damage the structure of the renal tubule. Persistence of Gd in the body resulted in the release of toxic Gd from gadodiamide transmetalation, which triggers nephrogenic systemic fibrosis	Thomsen (2006)
Ce	Myocardial infarction	Elevated Ce concentrations in toe nails increase the risk of myocardial infarction	
Several rare earth elements	Genotoxicity	Rare earth elements accumulate in the bone structure, changes the bone texture, and enhances bone marrow micronucleus	Chen and Zhu (2008) and Zaichick et al. (2011)
Several rare earth elements	Pneumoconiosis	Rare earth elements cause damage to the cells of the lungs, resulting in pneumoconiosis	Pagano et al. (2015)
Several rare earth elements	Sterility	Accumulate in serum and decrease percentage of normal sperm cells	Marzec-Wróblewska et al. (2015)
Several rare earth elements	Arteriosclerosis	Rare earth elements in blood raise cholesterol levels and production of a lot of lipoprotein	Migaszewski and Gałuszka (2015a, b)
Several rare earth elements	Memory loss	Rare earth element accumulation in brain tissues lowers intelligence quotient levels. Rare earth elements also cause dysfunctional neurological behavior, leading to impaired learning ability	Zhu et al. (1996) and Zhuang et al. (2017a, b)
Several rare earth elements	Failure for blood to clot	Rare earth elements lower the total blood bilirubin, glucose, and albumin	

pneumoconiosis and interstitial lung disease which have been reported to be prevalent among movie projectionists and photogravers. A detailed database of the case reports of the human health risks associated with occupation exposure is presented in an earlier study (Pagano et al. 2015). A study applying a scanning electron microscopy coupled to energy-dispersive X-ray detected cerium particles in the lungs of polishers for optical lens (Yoon et al. 2005).

Dietary intake of contaminated drinking water and foods also contribute to human intake of Rare earth elements. In China, two studies investigated the occurrence of rare earth elements in vegetable and cereal crops and the human health risks of rare earth elements in mining relative to non-mining (control) areas (Zhuang et al. 2017a, b). In the same studies, the total concentrations of rare earth elements in cereals were 74.22 μg/kg for mining area and 47.83 μg/kg for the control (Zhuang et al. 2017a), while the corresponding concentrations in vegetables were on average 94.08 μg/kg (mining area) versus 38.67 μg/kg (control) (Zhuang et al. 2017b). In terms of total rare earth elements, this resulted in maximum permissible human daily intakes of approximately 70 μg/kg body weight per day for both vegetables and cereal crops. The same authors hinted that human health risks could be particularly high in children due to continuous exposure. In another study, the total concentrations of rare earth elements in taro and water spinach treated with rare earth elements exceeded Chinese food standards (Li et al. 2013a, b). The same authors noted that the concentrations of rare earth elements were significantly lower in a non-leafy vegetable (taro) than a leafy one (water spinach). Consequently, the concentrations of rare earth elements detected in the human hair and blood were significantly higher in areas contaminated with rare earth elements (424.76–1294.8 μg/L) than in the corresponding samples from the control (Li et al. 2013a, b). This study shows that continuous application of rare earth elements to vegetables may enhance uptake and accumulation in edible parts and transfer of rare earth elements into the human food chain.

Hutchison and Albaaj estimated that acceptable rare earth element nitrate concentration in humans range from 0.2 to 2 mg/kg, beyond which adverse human health effects may occur. The human health risks of rare earth elements via dietary intake depend on the concentrations of rare earth elements in food and age and body weight of individuals. Although aquatic systems are sources of human food and water, limited data exists linking human health conditions to rare earth elements in aquatic systems. Thus, further research is required to investigate this aspect. Therefore, the bulk of literature on human health risks of rare earth elements is drawn from literature on medical applications and applications of rare earth elements-enriched fertilizers in crop production systems.

Once in the human body, rare earth elements may induce a wide range of human health effects (Table 1.4). Nephrogenic systemic fibrosis is the most severe human health risk attributed to gadolinium-derived gadolinium-based contrast agents used in medical applications (Broome 2008; Thomsen 2017). Gadolinium applications in magnetic resonace imaging and the associated human health effects are among the most studied rare earth elements of anthropogenic sources. Gadolinium-linked incidences of nephrogenic systemic fibrosis, including gadolinium deposition in the human body, and immediate toxicity during pregnancy and lactation and global best practices to minimize the human health risks have been discussed in an earlier review (Fraum et al. (2017).

Gadolinium from gadolinium-based contrast agents may migrate across the blood–brain barrier, resulting in gadolinium accumulation in the brain, where it may cause severe damage to the nephrological system (Kanda et al. 2017; Vergauwen et al. 2018). The speciation, partitioning, and human health risks of gadolinium derived from gadolinium-based contrast agents in the brain are discussed in an earlier review (Kanda et al. 2017). Other human health risks such as genotoxicity, fibrotic tissue injury, and bone alteration attributable to rare earth elements have also been reported in literature (Chen and Zhu 2008; Jenkins et al. 2011). Lanthanum causes neurological disorders and reduced the intelligence quotient particularly in infants (Zhu et al. 1996; Gwenzi et al. 2018), while cerium induces pneumoconiosis (Porru et al. 2000).

In males, rare earth elements have been reported to contribute to male sterility and anti-testicular effects (Chen et al. 2015; Marzec-Wróblewska et al. 2015). Zhu et al. (1996) report that high concentrations of rare earth elements in the human blood system promote the formation of arteriosclerosis via immunogenic damage to the vascular wall. Accumulation of rare earth elements in the human body alters blood properties such as serum triglyceride, β globulin, and albumin while decreasing glutamic pyruvic transaminase and total protein and increasing levels of cholesterol (Khan et al. 2017). However, some studies reporting concentrations of rare earth elements exceeding background values in human tissues due to environmental and dietary exposures failed to detect any corresponding apparent abnormalities in human health conditions (Li et al. 2013a, b; Zhuang et al. 2017a, b). As Pagano et al. (2015) indicated, case–control epidemiological evidence derived occupational exposure to rare earth elements is scanty. Therefore, the determination of maximum permissible concentrations of rare earth elements in food and environmental media, and the corresponding acceptable human intake values of rare earth elements, and chronic effects warrant further research.

1.4 Assessment and Mitigation of Health Risks

1.4.1 Assessment of Human and Ecological Health Risks

Health risk assessment is a critical step for evaluating human and ecological health risks of rare earth elements. According to Peduzzi et al. (2009) and Cardona et al. (2012), the notion of health risk entails three aspects: (1) the existence of a potential health threat or hazard, derived from the toxicity of rare earth elements; (2) risk of exposure to the hazard, including the exposure routes and daily intake, which are related to the occurrence and concentrations of anthropogenic rare earth elements in the environment; and (3) vulnerability, which is indicative of the predisposition or propensity of human and ecological populations to a hazard or harm, and other adverse health outcomes such as morbidity and mortality (Gwenzi and Chaukura 2018). These aspects underpin the approach for evaluating the health risks of rare earth elements in aquatic environments (Gwenzi et al. 2018). Health risk assessments include risk analysis, involving the identification and determination of concentrations of rare earth elements in aquatic media such as sediments, pore water, aquatic organisms, and surficial and deep bulk waters (Gwenzi and Chaukura 2018; Gwenzi et al. 2018). Subsequently, the evaluation of health risk may involve qualitative and quantitative analysis, including ecotoxicological studies, modelling, and rating to develop a priority list of rare earth elements that warrant mitigation (Milić et al. 2012). Qualitative risk evaluation or ranking entails the estimation of the probability, likelihood or frequency of occurrence of the event, and the magnitude of the harm or consequences on ecological or human health (Gwenzi and Chaukura 2018). Qualitative ranking and classification of the health risks use categories such as: “low”, “moderate”, “high”, and “extremely high” risk (Gwenzi and Chaukura

2018; Gwenzi et al. 2018). Such ranking facilitates the development of a priority list, which then assist in identifying health risks that warrant mitigation and target resources.

Quantitative risk assessment involves modelling and determination of quantitative metrics for determining the human health risks, which are often probabilistic results. However, in the case of rare earth elements, and other emerging contaminants, quantitative risk assessments are constrained by lack of maximum permissible guideline limits for both humans and the environment. Detailed discussions of health risk assessment procedures for various contaminants including emerging ones are presented in earlier papers and environmental guidelines (Organisation for Economic Co-operation and Development 2003, 2007; Gwenzi and Chaukura 2017; Gwenzi et al. 2018; Sanganyado and Gwenzi 2019). Indeed, several health risk assessment protocols exist at country and regional levels. These protocols include the US Environmental Protection Agency (2017) environmental risk assessment and Organisation for Economic Co-operation and Development (2015) guidelines widely used in the United States and the European community, respectively. These environmental risk assessment protocols apply known sensitive bioassay organisms to determine the dose–response relationships, including ecotoxicological threshold values (Organisation for Economic Co-operation and Development 2015). However, such protocols should be extended to include mixtures of rare earth elements and potential synergistic interactions among rare earth elements and other health stressors using realistic and environmentally relevant concentrations. Such risk assessment should consider daily intakes, multiple exposure pathways, nature and speciation of the rare earth elements, and age and nature of the target organisms exposed to rare earth elements.

1.4.2 Prevention and Control of Health Risks

Given that aquatic systems provide ecological services, and act as sources of human food and water, the remediation of rare earth elements in aquatic systems is a high priority. Potential preventative and control measures may be grouped into three classes: (1) "soft" engineering interventions, (2) "hard" engineering interventions, and (3) strategies aimed at minimizing the demand for raw rare earth elements. "Soft" engineering interventions include raising awareness on the human and ecological health risks of rare earth elements associated with their industrial applications and subsequent disposal. Such awareness may include educational campaigns, policy briefings, and mass media programs targeting policy makers, environmentalists, researchers, and the general public. Practicing proper housekeeping including implementing proper occupational hygiene and safety, health, and environmental programs in the rare earth element supply chain system may minimize waste and wastewater discharges and anthropogenic releases of rare earth elements into aquatic systems. At industrial plant level, principles of cleaner production can also be adopted to recover rare earth elements from industrial waste and wastewaters, thus attaining a closed-loop system and zero waste discharge scenario.

“Hard” engineering interventions include the development and applications of synthetic materials in engineered systems designed to remove rare earth elements. Such “hard engineering technologies include those based on chemical, biological, biochemical, and biosorption processes and (3) physicochemical processes. Among these technologies, the most prominent ones are (1) adsorption using natural or synthetic adsorbents (Tay et al. 2018; Zhang et al. 2018; Barros et al. 2019; Madbouly et al. 2019); (2) ion exchange using natural and synthetic ion-exchange resins (Wang 2018); (3) biosorption and biorecovery using live organisms such as fungi, bacteria, and microalgae (Kang et al. 2019; Furuhashi et al. 2019) and reverse osmosis (Lawrence et al. 2010); and (4) solid–liquid (Kumar et al. 2010, 2011) and liquid–liquid extraction using ionic liquids (Atanassova et al. 2018; Hunter et al. 2018; Habib et al. 2019).

The highlighted technologies have some limitations, including sludge production, some requiring large amounts of chemical reagents, membrane fouling, formation of secondary pollutants, and challenges in separation of adsorbents from aqueous solution. Due to low cost, environmental friendliness, simplicity of application, and readily available feedstocks for the development of adsorbents, adsorption is the most dominant and widely used process for removal of rare earth elements in aqueous systems (Fiket et al. 2018). However, one drawback of adsorption is that it cannot be used for many cycles due to the degradation of the adsorbent structure by the stripping solutions. To overcome this limitation, immobilization on silica and polymers has been used to render adsorbents more efficient and reusable (Gupta et al. 2019). The capacity of several methods to remove rare earth elements occurring in aqueous systems, including the mechanisms and the drawbacks involved, is discussed in detail in the respective papers (Atanassova et al. 2018; Furuhashi et al. 2019; Hunter et al. 2018; Kang et al. 2019; Tay et al. 2018; Wang 2018; Zhang et al. 2018; Madbouly et al. 2019). For example, Lawrence et al. (2010) reported that reverse osmosis removed more than 99% of gadolinium in aqueous systems. Moreover, to attain even higher removal efficiencies, several methods may be combined in tandem in a train consisting of various unit operations (Carolin et al. 2017). In addition, extractive and separation processes are the methods which may be adapted to recover rare earth elements from ores in aquatic systems. These methods include precipitation and selective leaching targeting specific rare earth elements (Hidayah and Abidin 2017; Innocenzi et al. 2018), solid–solid extraction techniques (Hidayah and Abidin 2017), the use of supercritical fluids (e.g., carbon dioxide) (Sinclair et al. 2017), membrane technologies (Liu et al. 2018), plasma separation methods (Gueroult et al. 2018), and ultrasonic extraction techniques (Diehl et al. 2018). However, limited data exists on the application of membrane filtration, electrochemistry, oxidation, photocatalysis, and adsorption using emerging biomaterials such as biochars and biochar–metal oxide composites.

The environmental footprint of the mining, processing, and industrial applications of rare earth elements could also be reduced via various interventions (Binnemans et al. 2013a, b). Specifically, the overreliance on rare earth elements for high-technology applications can be minimized by reducing the amount used in high-technology devices and equipment. Future demand for raw rare earth elements can also be reduced by exploring new and emerging industrial applications for rare earth elements which are most readily available and cheap to recover relative to less abundant ones. For example, the high concentrations of the costly dysprosium used in neodymium–iron–boron lightweight permanent magnets may be significantly minimized by using samarium–cobalt magnets, whose performance is comparable to that of the neodymium–iron–boron magnets. Another option is transmaterialization or substitution of rare earth elements with other materials, coupled with the elimination of rare earth elements in products by developing alternative non-rare earth element products. In this regard, transmaterialization may entail the use of carbon nanomaterials derived from abundant carbon, thus reducing the amount of rare earth elements required in the development of high-technology devices and applications (Arvidsson and Sandén 2017).

The demand for raw rare earth elements can also be reduced via recovering, recycling, and reuse of rare earth elements occurring in post-consumer high-technology products and nonconventional sources of rare earth elements including e-wastes and mine wastes such as coal ashes (Dent 2012; Tan et al. 2015). For rare earth elements in motors and generators, an option exists to substitute machines relying on permanent magnet with those based on coil-wound induction. In catalysis, scope exists to develop non-rare earth element catalysts, although such options may require considerable time, financial resources, and research effort (Dent 2012). These interventions highlighted here can be used as part of a conceptual framework for the mitigation of health risks of rare earth elements. Overall, a combination of "soft" and "hard" engineering interventions, coupled with minimizing the demand for raw rare earth elements via the various options highlighted, may reduce the global production and applications of rare earth elements and the associated health risks.

1.5 Future Research Directions

Specific knowledge gaps are highlighted under the following focal areas: (1) source partitioning and environmental behavior, (2) ecotoxicology, (3) human toxicology and epidemiology, and (4) human exposure and health risks in developing regions.

1.5.1 Source Partitioning and Behavior in Aquatic Systems

Literature on rare earth elements in aquatic systems is dominated by only a few elements (e.g., lanthanum, cerium, europium, gadolinium, lutetium), thus monitoring of the other understudied rare earth elements is required. Although several anthropogenic hotspot sources may contribute to rare earth elements detected in aquatic systems, limited data exists on source partitioning of rare earth elements among various sources. Moreover, barring studies documenting the concentrations of rare earth elements in aquatic systems, few studies have investigated the following: (1) the dominant hydrological and wind-driven processes accounting for the dissemination of rare earth elements in the environment; (2) the relative contribution of biogeochemical processes including sorption–desorption, complexation, ion exchange, precipitation, and plant uptake and bioaccumulation on the partitioning of rare earth elements in bulk water, pore water, the solid phase, and aquatic organism; and (3) trophic bioaccumulation in aquatic food webs including aquatic foods such as fish and crustaceans.

1.5.2 Ecotoxicology

Existing ecotoxicological data on rare earth elements are drawn from studies investigating the effects of single elements on a target bioassay species, while the synergistic interactions among rare earth elements, and between rare earth elements and other health stressors, remain understudied. Future research should investigate the effects of rare earth elements and the interactions between rare earth elements and other stressors on ecosystem services (e.g., biogeochemical cycling) and population, community, trophic, and ecosystem diversity and functions. Studies are also needed to estimate the ecotoxicological threshold points for various environmental media (e.g., median effect concentration, no observable effect concentration). To facilitate the determination of maximum allowable guideline limits, such studies should be conducted at concentrations considered to be environmentally relevant.

1.5.3 Human Toxicology and Epidemiology

Evidence exists on the human toxicology and health effects of rare earth elements in medical applications particularly gadolinium contrast agents used in magnetic resonace imaging (Thomsen 2006), while other studies conducted in China have reported rare earth elements in scalp hair sampled from children in rare earth element mining sites (e.g., Tong et al. 2004). However, further research is required to better understand the human exposure routes and daily intakes and behavior and fate of rare earth elements once in the human body. Moreover, further epidemiological research is required to establish the relationship between rare earth elements detected in aquatic environments and human health effects such as incidences of morbidity and mortality.

1.5.4 Human Exposure and Health Risks in Developing Countries

Developing regions, including Africa, are poorly represented in literature on anthropogenic rare earth elements in the environment including aquatic systems. Yet several developing countries, including Kenya, the Dominican Republic, and the Democratic Republic of Congo, among others have large reserves of rare earth elements. Moreover, developing countries also have anthropogenic hotspot sources of rare earth elements including medical facilities, waste repositories, and petroleum refineries. Other sources include recycling plants for waste electrical and electronic equipment and mining and mineral processing industries. In these regions, the bulk of monitoring data in aquatic systems excludes rare earth elements and is often limited to conventional toxic contaminants including toxic metals, pesticides, geogenic arsenic, and fluoride. As highlighted in one of the author's earlier papers (Gwenzi and Chaukura 2017; Gwenzi et al. 2018), the potential health risks of rare earth elements could be higher in these regions as in developed countries due to several risk factors. These risk factors include (1) weak and poorly enforced human health and environmental regulations, causing the discharge of partially treated and untreated effluents and wastewaters into aquatic environments; (2) the continued use of medical devices, equipment, and reagents long banned elsewhere in developed countries; (3) lack of regular human health surveillance systems for early detection and treatment of human health effects; and (4) unavailability of clean treated drinking water, thus forcing people to rely on potentially contaminated shallow groundwater and surface water sources. Therefore, comprehensive studies are required to investigate the occurrence of rare earth elements and their health risks in these regions. In addition, developing countries provide ideal sites to investigate the capacity of conventional low-cost water treatment (sand filtration, boiling) and emerging technologies such as zero-valent iron (Fe^0) and biochar water filters (Gwenzi et al. 2018) to remove rare earth elements in drinking water.

1.6 Summary, Conclusions, and Future Directions

This chapter summarizes the key sources, environmental behavior, human intake pathways, and health risks of anthropogenic rare earth elements as emerging contaminants. Anthropogenic rare earth elements (i.e., lanthanum, cerium, europium, gadolinium, lutetium) are widely reported in aquatic environments in several countries. Dominant hotspot sources of rare earth elements include mining and processing of rare earth elements, medical facilities, petroleum refineries, recycling industries for waste electrical and electronic equipment, industries producing high-technology products, pharmaceutical industries, and rare earth elements-enriched livestock feeds and fertilizers. The mobilization and transport of rare earth elements from sources and reservoirs occur via anthropogenic activities and hydrologically and wind-driven processes. Human exposure occurs via occupational inhalation, intake during medical applications, and consumption of contaminated water and foods. Evidence exists demonstrating the chronic and acute adverse effects of some rare earth elements in humans and aquatic organisms. Rare earth elements occur in human hair, nails, and bio-fluids (i.e., milk and serum), pointing to human intake and potential human health risks. Gadolinium contrast agents used in magnetic resonace imaging may cross the human brain–blood barrier and induce severe damage to nephrological systems and nephrogenic systemic fibrosis. In humans, rare earth elements also reduce intelligence quotient particularly in infants and cause dysfunctional neurological disorders, pneumoconiosis, fibrotic tissue injury, cytotoxicity, and oxidative stress. In males, rare earth elements induce anti-testicular health effects and sterility. However, barring gadolinium in gadolinium contrast agents used in magnetic resonace imaging, epidemiological evidence relating anthropogenic rare earth elements detected in aquatic systems to adverse human health effects is still weak. In plants, rare earth elements decrease both root function and growth and nutritional quality, while genotoxicity, oxidative stress, and neurotoxicity may occur in aquatic animals. Rare earth elements may bioaccumulate in aquatic organisms along trophic levels and cause chronic and acute toxicities and alter ecological functions. Although data drawn from developing countries including Africa are still missing, the human health and ecological risks could be higher in these regions than currently perceived. The risk factors predisposing human and ecological health in developing countries relative to their counterparts in developed countries were discussed. A conceptual outline for assessing and mitigating health risks of rare earth elements was highlighted. Specific remediation techniques for the removal of rare earth elements in aqueous systems were also discussed. Future research directions were highlighted to better understand the following: (1) hotspot reservoirs; (2) behavior and fate of rare earth elements in aquatic systems, including solid–liquid phase partitioning; (3) human toxicology and epidemiology and aquatic ecotoxicology; and (4) remediation of rare earth elements in aqueous systems, including drinking water and wastewaters. An improved understanding of the sources, behavior, and health risks of anthropogenic rare earth elements could contribute toward the development of environmental maximum guideline limits.

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