

1Potentially Toxic Elements in Solid Waste Streams: Fate and Management Approaches

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28Abstract:

29Solid wastes containing potentially toxic elements (PTEs) are widely generated around the globe.
30Critical concerns have been raised over their impacts on human health and the environment,
31especially for the exposure to PTEs during the transfer and disposal of the wastes. It is important to
32devise highly-efficient and cost-effective treatment technologies for the removal or immobilisation
33of PTEs in solid wastes. However, there is an inadequate overview of the global flow of PTEs-
34contaminated solid wastes in terms of geographical distribution patterns, which is vital information
35for decision making in sustainable waste management. Moreover, in view of the scarcity of resources
36and the call for a circular economy, there is a pressing need to recover materials (e.g., precious
37metals and rare earth elements) from waste streams and this is a more sustainable and
38environmentally friendly practice compared with ore mining. Therefore, this paper aims to give a
39thorough overview to the global flow of PTEs and the recovery of waste materials. This review first
40summarises PTEs content in various types of solid wastes. Then, with respect to their patterns of
41transport transformation and risks in the changing environment, toxic metal(loid)s, radioactive
42elements, and rare earth elements are critically reviewed. Different treatments for the management of
43these contaminated solid wastes are discussed. Based on an improved understanding of the dynamics
44of metal(loid) fates and a review of existing management options, new scientific insights are
45provided for future research in the development of high-performance and sustainable treatment
46technologies for PTEs in solid wastes.

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48Keywords: metals/metalloids; rare earth elements; radioactive elements; sustainable waste
49management; environmental and human health risk; green and sustainable remediation.

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51 Table of Contents

521.	Introduction.....	5§§
532.	Flow of metal-containing solid wastes.....	7§§
543.	Fate of toxic metal(oids) and their treatment.....	11§§
55	3.1. Chromium.....	11§§
56	3.2. Arsenic.....	13§§
57	3.3. Selenium.....	15§§
58	3.4. Cadmium.....	18§§
59	3.5 Mercury.....	22§§
60	3.6. Lead.....	24§§
61	3.7 Treatment of metal(loid)-containing wastes.....	26§§
624.	Fate of radioactive elements and their treatment.....	31§§
63	4.1 Uranium.....	31§§
64	4.2. Radium.....	35§§
65	4.3. Potassium.....	37§§
66	4.4. Treatment of radioactive elements.....	39§§
675.	Fate of rare earth elements (REEs) and their treatment.....	40§§
68	5. 1. Natural existence of REEs in mineral deposits.....	40§§
69	5. 2 Recycling from coal combustion residues.....	42§§
70	5.3. Recycling from other end-of-life products.....	43§§
716.	Prospects and future research.....	45§§
727.	Conclusions.....	46§§
738.	Acknowledgment.....	48§§
749.	References.....	48§§

75

76List of Abbreviations

77APCDs Air Pollution Control Devices

78CCC Chronic Criterion Concentration

79CCRs Coal Combustion Residues

80EPA United States Environmental Protection Agency

81ESP Electrostatic Precipitators

82E-waste Electronic Wastes

83FGD Flue Gas Desulfurization

84HREE Heavy Rare Earth Elements

85IAEA International Atomic Energy Agency

86LREE Light Rare Earth Elements

87L/S Liquid to Solid

88MSW Municipal Solid Wastes

89PRB Permeable Reactive Barrier

90REEs Rare Earth Elements

91S/S Stabilization/Solidification

92TCLP Toxicity Characteristic Leaching Procedure

93TENORM Technologically Enhanced Naturally Occurring Radioactive Materials

94WEEE Wastes of Electrical and Electronic Equipment

95

96 **1. Introduction**

97Solid wastes, including municipal, electronic, and industrial wastes, lead to global environmental
98problems. Metal(loid)s (e.g., chromium, arsenic, selenium, cadmium, mercury, and lead) are widely
99used in industry and present a major concern in solid waste management in view of their acute and
100chronic risks to human health and ecosystems (Song and Li, 2014; Marra et al., 2017). Similarly,
101radioactive elements (e.g., uranium, radium, and thorium) in wastes generated from nuclear
102applications for energy, research, and clinic purposes should be cautiously treated to avoid leaching
103and subsequent environmental risks. For rare earth elements (e.g., yttrium and scandium), there are
104economic incentives for people to recover these precious resources from solid wastes due to their
105limited availability in the Earth's crust (Binnemans et al., 2013; Tunsu et al., 2015; Işıldar et al.,
1062019). Studying the fate and transportation of these toxic elements in solid wastes are of great
107significance to better understand the pathways of contaminants and their interactions with the
108environment. This is essential for the design of effective treatment processes based on sound
109scientific understanding.

110

111In recent years, increasing attention has been drawn to the environmental fate of potentially toxic
112elements (PTEs) from wastes. PTEs refer to chemical elements including both metals and non-metals
113that may potentially cause harmful effects on the organisms in the environment if present in high
114concentrations (Antoniadis et al., 2019). It has been recognized that municipal solid wastes are a
115source of toxic elements produced in waste-to-energy incineration and found in flue gas and fly ash
116(Hu et al., 2018). The leachability and mobility of PTEs in flue gas and fly ash show sensitivity to
117pH (Izquierdo and Querol, 2012; Chen et al., 2019). In addition, the PTEs in wastewater accumulate
118in sewage sludge. Upon thermal treatment of sewage sludge, PTEs are distributed in different
119fractions (i.e., ashes, chars, and bio-oils), largely depending on the conditions of temperature and
120humidity (Fang et al., 2018; Udayanga et al., 2018). The ultimate destination of PTEs is

121accumulation in soil, sediment, and surface water or groundwater systems, depending on various
122physical and chemical characteristics of the wastes (Tabelin et al., 2018). Previous reviews have only
123considered the association of PTEs with certain kinds of solid waste streams such as fly ash.
124However, the quality of solid wastes derived from different industries and under different regulatory
125contexts can have considerable variations, and few studies consider that.

126

127In management of hazardous waste, stabilisation/solidification (S/S) using cementitious materials or
128other stabilisers is the most common approach, which covers approximately 40% of the commercial
129treatment market (Wang et al., 2015; Wang et al., 2018a; Chen et al., 2019). Recent research efforts
130have explored innovative treatment technologies for PTEs in solid wastes, such as
131biohydrometallurgy (Sethurajan et al., 2018), electro dialytic treatment (Kirkelund et al., 2018), and
132sorbent capture (Ke et al., 2019). There is an emerging perspective for solid waste management,
133which is to valorise the wastes for different applications, e.g., dredged sediment as construction
134materials (Wang et al., 2015), blast furnace slag as cementitious materials (Wang et al., 2018a), red
135mud or calcined alum sludge as a metal sorbent (Ahmed and Ahmaruzzaman, 2016; Jeon et al.,
1362018), and incinerator sludge ash for zeolite catalyst synthesis (Feng et al., 2018; Zhang et al.,
1372018a).

138

139In contrast to previous reviews on PTEs which tend to focus on specific waste streams (Hu et al.,
1402018; Udayanga et al., 2018; Işıldar et al., 2019), this review aims to provide a PTEs-oriented and
141comprehensive overview of the fate of metal(loid)s, radioactive elements, and rare earth elements in
142a wide range of solid wastes to elucidate their potential environmental and human risks, and to
143identify the opportunities for new developments in solid wastes management including recycling and
144valorisation. State-of-the-art treatment technologies are discussed to assess their feasibility on the
145field scale and facilitate decision making by industrial practitioners. Moreover, this review identifies

146existing managerial and technological limitations, which should shed light on the directions of future
147research for the scientific community.

148

149 **2. Flow of metal-containing solid wastes**

150Solid waste is a summative term covering all the discarded materials in the solid or semi-solid forms
151which come from municipal, industrial, commercial, mining, and agricultural activities (McDougall
152et al., 2001). Metal-containing solid wastes, encompassing those from municipal to industrial
153sources, have been extensively concerned all over the world (Table 1) and their fates are summarised
154in Figure 1. Derived from household activities, municipal solid waste (MSW) is one of the largest
155components of wastes streams and includes food wastes, paper, textile, rubber, plastic, glass, metals,
156wood, and miscellaneous inorganic wastes. The composition of MSW varies from district to district,
157which influences the PTEs composition in the waste stream. MSW is predominately treated by
158incineration, landfill, and sometimes, composting. MSW incineration has grown steadily in recent
159years worldwide. For example, the amount of incinerated municipal solid wastes in the 27 EU
160Member States has risen by 32 million tonnes in ten years, reaching 64 million tonnes in 2014
161(Abramov et al., 2018). In China, approximately 40% of MSW in metropolitan areas is incinerated,
162and this figure is expected to reach 60% by 2020. As a result, the generation of incinerator fly ash in
163China exceeded 5 million tonnes in 2016 (Du et al., 2018).

164

165During MSW incineration, organometallic compounds volatilise along with volatile metals (e.g., Hg
166and Cd). As temperature decreases in the downstream units, volatilised metallic compounds
167experience condensation and nucleation reactions to form submicron particles. Most organic-based
168materials can be converted to carbon dioxide and water vapour by complete oxidation at high
169temperature, whereas the metal fractions still remain in the ash particles. Ashes consisting of metals,
170salts, and organic contaminants either aggregate in the heat recovery phase (coarse particles) or are
171removed from the flue gas by fabric filters or electrostatic precipitators (ESPs) (fine particles)

172(Weibel et al., 2017). Boiler and ESP ashes constitute the fly ash, while the incombustible residue
173that accumulates at the bottom of incinerators is the bottom ash. These incineration solid residues
174have gained increasing concern due to their leaching potential to soil and water after disposal in
175landfill sites. Volatile components are separated from the flue gas via a flue gas control process;
176metals can be present in flue gas (e.g., Fe-oxides and brass) as well as mineral aggregates (e.g.,
177quartz, feldspar, and glass), or may be vaporised and condensed as water-soluble chlorides
178(K_2ZnCl_4) or sulphates ($PbSO_4$). Metal mobilisation and leaching are significantly dependent on
179these metal associations (Weibel et al., 2017). As for disposal of hazardous wastes such as
180incineration fly ash, significant effort should be made in developing metal stabilisation technologies
181(e.g., S/S, vitrification, sintering, and thermocatalytic and hydrothermal treatment). On the other
182hand, it is estimated that the value of precious metals, light and heavy rare earth elements (REEs),
183and other technology-critical elements including Cu in incinerated residues can reach \$1,916 per ton
184of material (Abramov et al., 2018). Therefore, the recycling of metals from incineration ash is a
185promising future approach.

186

187The deposition and accumulation of PTEs during MSW disposal in landfill sites is a major concern;
188PTEs have complex pathways into the environment and potentially pose high risks to living
189organisms surrounding landfill areas. The content of major metals in landfill material is reflected by
190waste precursors, which are the solid wastes formed from heterogeneous interactions between
191disposed wastes, climatic conditions, and the management practices of the landfill (Adelopo et al.,
1922018). Waste degradation reduces the weight of landfilled waste, but metal concentrations increase.
193Unlike organic pollutants, metals do not degrade in landfills and their residence time is
194approximately 150 years (EU, 2002). Metal leachability from landfills results in the pollution of
195farmlands, surface waters, and underground waters. Higher levels of metal contamination have been
196found in closed landfills than active ones due to on-going degradation. However, the metal

197bioavailability might reduce with ageing processes as a result of the stronger binding formed with
198soil components (Tye et al., 2003).

199

200In comparison, composting is an alternative approach to treating the organic fraction of MSW
201(Godlewska et al., 2017), but this can also pose a threat to aquatic and edaphic systems. For example,
202open-air windrow composting piles has resulted in the uncontrolled emission of metal contaminants
203and considerable ecological risks in the surrounding watershed (Sharifi et al., 2016). Natural events
204such as rainfall also tend to accelerate the leaching of PTEs if inadequate control is in place to isolate
205open windows or stockpiled composting sites.

206

207Electronic wastes have recently drawn extensive concern due to the short lifespan of electrical and
208electronic products nowadays. With the development of the electronics industry, a significant amount
209of electronic waste (e-waste) is released into the environment at the rate of 20–50 million tonnes per
210year. Common metals found at e-waste sites are both valuable (e.g., Cu and platinum groups) and
211hazardous (e.g., Pb, Sb, Hg, Cd, and Ni) (Beiyuan et al., 2016; Jiang et al., 2019). After e-waste
212disposal in landfill sites, PTEs are not reduced as other organic contaminants, but are accumulated,
213transported, and diffused into soil pore water, thereby threatening soil ecosystems. Furthermore,
214PTEs are released in the form of airborne, size-fractionated bounded particles. Research has found
215that metals with carcinogenic effects pose severe health risks to humans by means of inhalation,
216particularly for children in the vicinity of e-waste recycling areas (Huang et al., 2016).

217

218Industrial wastes produced by numerous manufacturing and processing activities are also critical
219sources of PTEs. Among them, coal combustion is an essential means of electricity and heat
220generation, generating significant volumes of coal combustion residues (CCRs) and coal combustion
221products (CCPs). Globally, coal-based power generation produces over 750 million tonnes of coal
222ash per year (Izquierdo and Querol, 2012). It has been estimated that over 130 million tonnes of

223CCRs are generated by US power plants every year (Ruhl et al., 2012). Composed of fly ash, bottom
224ash, and flue gas desulphurisation (FGD) materials from coal combustion, CCRs contain numerous
225crystalline oxide phases of silicon, aluminium, calcium, and iron as well as metals and trace elements
226(Schwartz et al., 2018). Similar to MSW incineration fly ash, the fly ash from coal combustion can
227be collected from flue gas by ESP or bag filters. Coal ash is regarded as a major potential source of
228PTEs in the environment. Hence, its disposal has drawn great attention. The characteristics of the
229coal source significantly influence the variations in PTEs content. For example, it is estimated that
230coal-combusted fly ash contains 2.3 to 6,300 mg/kg arsenic, 0.02–0.36 mg/kg mercury, 1.2–236 mg/
231kg molybdenum, and 0.2–134 mg/kg selenium, and these concentrations are approximately 10 times
232higher than those of the original source (Feng et al., 2018). Due to the high content of organic and
233inorganic contaminants, EU directives (91/689/EEC) classify coal fly ash as a hazardous waste. In
234contrast, in the United States (Resource Conservation and Recovery Act), coal fly ash is considered a
235non-hazardous material if leaching rates pass the TCLP (toxicity characteristic leaching procedure)
236(Tsang et al., 2014; Kirkelund and Jensen, 2018). Furthermore, the water used for producing steam,
237cooling, and transportation—which may also be contaminated by CCRs—poses a threat to the
238natural water environment.

239

240The generation of sewage sludge from wastewater treatment plants has been steadily increasing over
241time. It is estimated that America generates 71 million tonnes of sewage sludge per year, and in
242Europe, the average annual outputs of sewage sludge in Germany, England, and France are 22, 12,
243and 8.5 million tonnes, respectively. In China, over 20 million tonnes of sewage sludge are generated
244each year (Xu et al., 2014). Sewage sludge is a heterogeneous mixture of microorganisms and
245undigested organic (e.g., proteins, polysaccharides, and lipids) and inorganic (e.g., minerals and
246metals) components with high moisture content (Kwon et al., 2018; Yang et al., 2017). The disposal
247and treatment of sewage sludge is a global environmental challenge. Recently, fuel generation (i.e.,
248heat, hydrochar, bio-oil, and syngas) from sewage sludge via thermal processes (i.e., incineration,

249gasification, pyrolysis, and hydrothermal treatment) has gained increasing attention (Kwon et al.,
2502018). However, the elevated concentrations of metals that result from thermal processes has
251presented environmental challenges to their increasing application (Udayanga et al., 2018).

252

253 **3. Fate of toxic metal(loids) and their treatment**

254In the following subsections, the emission and transfer of commonly encountered toxic metal(loids)
255(i.e., chromium, arsenic, selenium, cadmium, mercury, and lead) from solid waste streams are
256explained (Section 3.1 to 3.6), in an order with respect to the periodic table, and relevant research is
257summarised in Table 2. Afterwards, their decontamination approaches are elaborated and discussed
258in Section 3.7.

259

2603.1. Chromium

261Chromium is a PTE that primarily exists in trivalent and hexavalent forms. Because of its strong
262oxidising and toxicity properties, hexavalent chromium compounds can easily contaminate soil,
263groundwater, and rivers through leaching, which leads to serious ecological problems (Rajapaksha et
264al., 2013; Rajapaksha et al., 2018). The toxicity of chromium varies depending on the valence state
265and its morphology. Hexavalent chromium is a toxic carcinogenic element, and its toxicity is one
266hundred times greater than that of trivalent chromium (Mandal et al., 2017). Metal chromium and
267chromium salts are regarded as important strategic resources, widely used in leather, electroplating,
268chemical, ceramics, advanced alloy materials, spices, pigments, printing and dyeing, anti-corrosion
269treatment, medicine, and other industries (Tsang and Yip, 2014). Chromium may derive from e-
270waste such as data tapes and floppy-disks and has a typical concentration of 9,900 mg/kg in e-waste
271with a global generation rate of 198,000 tonnes (Gaidajis et al, 2010). Landfill, as the most common
272approach for e-waste disposal, limits the recycling and reuse of Cr-containing materials from this
273waste stream (Ashraf et al., 2017).

274

275Leather production enterprises are important Cr sources; Cr is produced from chromium powder, and
276chromium-containing wet blue and other pigments in the tanning process (Wu et al., 2015). The
277concentration of Cr predominantly depends on the production conditions, the types of raw materials
278used, and the level of production management. Tannery sludge is a toxic industrial solid waste
279produced during the physical treatment of tannery wastewater. By treating tannery sludge with ladle
280furnace slag and organoclay, chromium and dissolved organic carbon concentrations can be lower
281than the specified limits for non-hazardous waste for landfill disposal (Rajapaksha et al., 2013). In
282addition, bioleaching has been developed as a successful and cost-effective method for removing
283Cr(III) from tannery sludge. Mixed inoculations of primary iron sulphur-oxidising bacteria, Fe²⁺,
284and elemental sulphur are used, which accelerate the bio-oxidation of Fe²⁺ and elemental sulphur to
285produce acid and increases the redox potential, reaching a maximum removal rate at 95.6% (Zhou et
286al., 2006).

287

288Electroplating production also generates large amounts of sludge containing 3% to 5% chromium,
289which mainly exists as hydroxide (Yuan et al., 2013; Yi et al., 2014). There are three main treatment
290methods for electroplating sludge: (1) stabilisation treatment; (2) thermochemical treatment; and (3)
291resource utilisation (Yuan et al., 2017). Cement and fly ash can be used to immobilise high
292chromium plating waste by S/S process (Dayananda et al., 2013). The MSW incineration fly ash-
293based Friedel matrix also can be used to stabilise industrial chromium-containing electroplating
294sludge. The formation of ettringite and Friedel plays an important role in the fixation of metals in
295electroplating sludge (Qian et al., 2009). Recently, the possibility of using stabilised and solidified
296sludge in the construction and building industry has been explored (Kanwan et al., 2017). During
297plasma in-flight treatment, hexavalent Cr is converted into trivalent, leading to a decrease in toxicity,
298especially at high temperatures. In addition, incineration can effectively enrich Cr in electroplating
299sludge, and 90% of the chromium is retained in the ash (Espinosa and Tenório, 2001). For resource
300utilisation, Cr in electroplating sludge can be recovered by alkaline oxidation of hypochlorite.

301Hypochlorite can completely oxidise chromium(III) hydroxide in suspension to chromate under
302alkaline conditions, and can then pass sludge roasting and aqueous leaching (Matthew et al., 2000).
303The element can then, finally, be filtered to separate the chromate solution. The recovery efficiency
304of chromium can reach 92% under the optimal conditions (Silva et al., 2005). Furthermore,
305sulphidation and oxidation treatment can be used. Firstly, mixed electroplating sludge is
306simultaneously extracted and copper vulcanised at pH 1.5 to dissolve the metals, and Cu^{2+}
307precipitates in the form of CuS . Secondly, Zn is subjected to sulphurisation treatment at pH 4.5, and
308then at pH 10, and the remaining liquid is oxidised with H_2O_2 . The oxidation rate of Cr increases
309when both the temperature and the amount of added H_2O_2 increase (Kuchar et al., 2010). Metal
310phosphate and chromium oxide nanocomposites have been prepared from Cr-III electroplating
311sludge using an extraction-precipitation-calcination process. Metal-phosphate/chromium oxide
312nanocomposite has a good potential to be used as a functional pigment in energy-efficient buildings
313(Li et al., 2015).

314

3153.2. Arsenic

316Arsenic widely exists in contaminated soil and tailings produced from metallurgical processes. Some
317sediments and bio-wastes also contain high concentrations of arsenic, which can cause significant
318harm to the environment. In recent years, due to irrigation and the use of a large amount of
319groundwater in industrial processes (such as mining, non-ferrous metal smelting, coal mining) as
320well as the use of arsenic-based pesticides and municipal sewage sludge, arsenic concentrations in
321soils have increased (Tsang et al., 2014; Niazi et al., 2018; Shakoor, et al., 2018). Groundwater
322contaminated with arsenic has been a serious problem. When arsenic in soil enters plant tissue,
323metabolism is affected; arsenate weakens phosphorylation in mitochondria. Arsenite inactivates
324many enzymes by reacting with sulfhydryl proteins, hindering the moisture uptake in the plant. It
325also affects the absorption of water and nutrients by plants and causes arsenic to accumulate in plant
326tissue, resulting in damage to chlorophyll. Arsenic-containing tailings often contaminate soil,

327groundwater, and surface water, causing further damage to plants, animals, and humans (Mandal and
328Suzuki, 2002).

329

330Arsenic is ubiquitous in the environment and highly toxic to all forms of life. Trivalent arsenic is
331more toxic its pentavalent form, and inorganic arsenic is more toxic than organic arsenic (Niazi et al.,
3322018). High levels of organic arsenic compounds such as arsenic betaine and arsenic choline are
333found in marine organisms, and they show good resistance to chemical degradation. Arsenic can
334accumulate in organisms and then slowly excreted. In general, organic arsenic compounds are
335excreted more quickly than inorganic arsenic, and pentavalent arsenic is removed from the body
336faster than trivalent arsenic (Mandal and Suzuki, 2002). Arsenic enters the body of organisms
337through water pollution, and through the consumption of As-containing plants, dairy products, and
338meat. Arsenic poisoning can cause melanosis, keratosis, hyperkeratosis, back edema, gangrene, skin
339cancer, and other diseases (Singh et al., 2015b). Arsenic in the human body distributes in hair, nails
340and teeth, generally in the order of 3-4 mg/kg, increases with age. The absorption of arsenic in the
341human body is high for anionic and soluble species, but very low for insoluble species (Singh et al.,
3422015b).

343

3443.3. Selenium

345Selenium (Se) is a non-metal with intermediate properties between sulphur and tellurium. Being
346prone to mobilisation and bioaccumulation in aquatic organisms, selenium is toxic at high levels,
347although it is an essential dietary micronutrient for humans and animals at trace concentrations. It
348also acts as a fertiliser additive to enrich selenium-poor soils for plant growth. The Se species
349partitioning plays a significant role in its mobility and bioavailability. Oxidised Se predominately
350exists as oxyanions dissolved in solution or adsorbed to mineral surfaces. Oxidised Se (i.e., selenite
351and selenate) are highly soluble and more mobile than the less soluble elemental Se₀ (Liu et al.,
3522013).

353

354 Selenium originates from various industrial solid wastes, such as those resulting from glass
355 manufacturing, selective oxidation catalysts, plating, rubber compound synthesis, and electrolytic
356 manganese production. It is estimated that the world's consumption of Se consists of 40%
357 metallurgy, 25% glass manufacturing, 10% agriculture, 10% chemicals, 10% electronics, and 5%
358 other uses (United States Geological Survey, 2018). Selenium can be recovered from the electrolytic
359 refining of copper as well as from photocopiers and electronic materials. Coal is also a major source
360 of selenium (0.5–12 mg/kg) (United States Geological Survey, 2018), which is 80–90 times higher
361 than the average concentration in copper deposits. Selenium in coal is released during coal
362 combustion, but it has a potential for recovery from coal fly ash. Selenium in coal ashes (including
363 fly ash and bottom ash) is a mixture of elemental Se₀ and Se oxyanions, mostly existing as sulphide
364 and organic associations, among which selenite is the major dissolved species, constituting
365 approximately 75–94% of the total Se (Liu et al., 2013). Selenium associates with coal ash by the
366 adsorption of Se oxyanions (selenite and selenite) on mineral surfaces such as Fe-, Al-, and Mn-
367 (hydr)oxides. In addition, solid solutions can be formed with minerals such as gypsum (CaSO₄) and
368 hydrocalumite (Ca₄[Al(OH)₆]₂(OH)₂·6H₂O). Selenium in fly ash also coprecipitates with
369 multivalent cations including Ca²⁺, Pb²⁺, and Ba²⁺ (Liu et al., 2013). The weak bonding with the
370 coal solid phase means that Se is easily released and almost completely volatilised during
371 combustion (Izquierdo and Querol, 2012). As an important component of e-waste, Se enters the soil
372 after industrial or municipal solid waste disposal. Its transport involves metal complexation and
373 adsorption, which are closely related to landfill age (Adelopo et al., 2018), but does not appear to be
374 influenced by depth (Huang et al., 2016).

375

376 Leaching potential is regarded as an important parameter when elucidating the mechanisms of Se
377 migration and its impact on aquatic ecosystems. It has been reported that during leaching from coal
378 ash samples, dissolved Se generally increases with a decreasing liquid-to-solid (L/S) ratio and

379increasing reaction time, as well as a pH-dependent pattern (Liu et al., 2013). In a fly ash impact
380assessment for groundwater quality, most elements including Se and Pb showed high leaching effects
381at a L/S ratio of 5 (Dandautiya, et al., 2018). In these cases, the concentrations of Se in groundwater
382samples from nearby the fly ash dumping site were up to 0.077 mg/L and higher than the World
383Health Organization's desirable drinking water limit (0.04 mg/L). When pH is less than 2.5, the
384dominant species (H_2SeO_3) is not easily adsorbed on the surface of ash particles and so remains in
385solution. It has been reported that over 70% of Se can be removed from coal fly ash at $\text{pH} < 1$
386considering its primary surface association (Izquierdo and Querol, 2012). However, as pH increases,
387the dominant anionic species HSeO_3^- and SeO_3^{2-} can be adsorbed onto the surface of ash
388particles, from which desorption of Se oxyanions plays a vital role in Se release (Izquierdo and
389Querol, 2012). Low proton levels under alkaline conditions promote the oxidation of elemental Se⁰,
390thus boosting desorption or dissolution of Se oxyanions into the leaching solution. The amount of
391leachable Se from coal fly ash increases with pH, and dissolved Se concentrations at pH of 13 are
39213–81 times higher than at pH 5 (Liu et al., 2013). Therefore, high Se leaching under alkaline
393conditions becomes a major concern about combustion wastes, and dynamic variations in ecosystems
394(notably pH increases) can be conducive to higher rates of Se mobilisation.

395

396It has been reported in northern California that CCR from electrical power plants has had an impact
397on the surrounding waterways (i.e., lakes and rivers) (Ruhl et al., 2012); in comparison to upstream,
398Se was enriched at outfalls with concentrations as high as 87.2 $\mu\text{g/L}$, i.e., 17 times Chronic Criterion
399Concentration (CCC) of Se for aquatic life of (5 $\mu\text{g/L}$) as identified by the US EPA. Seasonal
400thermal stratification (i.e., depletion of oxygen in bottom water during summer and overturning
401during autumn) also induces spatial and temporal changes in water chemistry, which can lead to Se
402adsorption onto and desorption from suspended solids in rivers and its accumulation in bottom
403sediments. Nonetheless, Se can be immobilised in sediment by forming elemental selenium and
404metal–selenium complexes and is not incorporated into pore water (Ruhl et al., 2012).

405

406 Other important factors for Se are the nature of the combustion facilities at the source as well as the
407 chemistry of the receiving environment. Effluent from flue gas desulphurisation (FGD) systems,
408 which is usually added to ash pond water for dilution, has a higher enrichment of contaminants
409 compared to ash discharged at outfalls. FGD disposal systems effectively remove volatile elements
410 associated with SO_x, and significantly accumulate contaminants in solid wastes and wastewater
411 discharged from power plants. Selenium concentrations are also higher at FGD outfalls compared to
412 outfalls that discharge ash only (without FGD). Anoxic conditions in water lead to lower dissolved
413 Se concentrations. The oxidised species selenate (Se(VI)) is less reactive to adsorption onto
414 oxyhydroxides in aerobic conditions, while anoxic conditions result in the conversion of Se into
415 reduced forms (e.g., elemental Se⁰ and FeSe) that possess strong adsorption affinity towards both
416 oxyhydroxides and clay minerals (Ruhl et al., 2012). Flow rate also plays a significant role in the
417 dilution effect and thereby influences the contamination level of the receiving waters. During periods
418 of drought, the dilution factor is reduced, e.g., a five-fold reduction in water flow would raise the
419 CCR contribution up to 22%, thus increasing the concentrations of contaminants in downstream river
420 water (Ruhl et al., 2012). In addition, changes in the ambient conditions of lake sediments (e.g., pH
421 and redox state) tend to result in the release of Se metalloids into pore water.

422

423 3.4. Cadmium

424 Cadmium (Cd) is considered an extremely toxic element for human and aquatic life. It is normally
425 immobile under near neutral and alkaline conditions (Rizwan et al., 2016). Exposure to high
426 concentrations of Cd can damage the reproductive system, the lungs, DNA, and the kidneys, and can
427 cause problems for learning, cognition, behaviour, and neuromotor skills in children (Rizwan et al.,
428 2018). Consumption of cadmium and its compounds is linked to the wide use of industrial materials
429 such as alloys, coatings, batteries, pigments, and plastic stabilisers (United States Geological Survey,

4302018). For Cd in sewage sludge, concentrations ranging from 0 to 110 mg/kg were identified in
431different sewage sludge around the world (Udayanga et al., 2018). An average Cd concentration of
4322.13 mg/kg dry matter was reported across different seasons in sewage sludge from Greece (Spanos
433et al., 2014), while a concentration of 1.7 mg/kg was reported for sludge in a sewage treatment plant
434in France (Manon et al., 2019). In the latter case, a large proportion of Cd was associated with Fe-Mn
435oxides (52%) and oxidisable fractions (21%), with the rest being associated with the residual
436fraction. The pH-dependent leaching of Cd showed a bell-shaped relationship, which may be
437attributed to Cd desorption from Fe-Mn oxides the dissolution of Fe-oxide bearing phases, and the
438leaching of organic ligands from particulate organic matter (Manon et al., 2019).

439

440Disposal of e-waste, such as batteries and paint residual, is an important source of Cd in the aquatic
441and soil environment in the vicinity of municipal landfill areas. Cadmium is reported as a potential
442contaminant in both active and closed landfills, presenting the highest health risk potential index
443rating among other metals including Cu, Pb, Zn, and Ag (Adelopo et al., 2018). Depending on their
444mobility, these metals can penetrate through the soils and disperse vertically/horizontally across
445subsurface layers. A reduction in pH decreases Cd adsorption and increases its mobility through the
446enhanced competition for negative surfaces between H⁺ and dissolved metals (Rizwan et al., 2017).
447Metal concentrations generally decrease with increasing depth, because more sorption sites are
448provided by higher organic matter content in surface soils and higher pH conditions reduce metal
449mobility in deeper soil profiles (Wu et al., 2015). As Cd mainly exists in an extractable form, it
450shows a relatively weak vertical mobility compared with other metals (Jiang et al., 2019). It was
451found that illegal e-waste recycling activity in south China caused severe contamination of
452surrounding surface soils (Cd > 0.39 mg/kg, exceeding the guideline level of 0.3 mg/kg) and pond
453water (Wu et al., 2015; Zhu et al., 2018). Furthermore, contaminated pond water has often been used
454for farmland irrigation, resulting in the metal contamination of paddy soil and significant
455environmental damage (Ok et al., 2011). It has been shown that closed landfill sites suffer more

456severely from metal contamination than active landfill sites. In addition to landfill age, metal
457concentrations also increase with a decrease in particle size, which corresponds to the degree of
458waste degradation. As discussed in Section 2, metals are not reduced but are redistributed or released
459during waste degradation in landfill sites. E-waste recycling/disposal also promotes the release of Cd
460into the atmosphere as well as the formation of airborne particle-bound metals that can be deposited
461in the respiratory systems of humans living in the vicinity of recycling sites. For example, airborne
462particle-bound Cd with an average concentration of 6.1 ng/m³ displayed a spatial distribution with
463peaks at a distance of 0.56–1.0 m and 1.0–1.8 m from a recycling site, resulting in an incremental
464lifetime cancer risk that was higher than the acceptable cancer risk threshold (Huang et al., 2016).

465
466Different from e-waste recycling zones, general MSW landfill appears to have less of an impact,
467with typically lower Cd contents. In a life cycle assessment of MSW management system in Turkey,
468Cd derived from industrial hazardous waste or household solid waste was found to be negligible (\leq
4690.05 mg/kg oven dry waste) (Yay et al., 2016). One study detected 1.0–3.0 mg/kg Cd in the fine
470fraction (< 10 mm) of landfilled wastes, where Cd dominantly existed in its reduced form, but was
471found in relatively low concentrations along with other potentially toxic elements such as Pb and Cr
472(Burlakovs et al., 2018). These metals were mostly bound in environmentally stable compounds,
473meaning there was potential to recover materials from the landfilled wastes after appropriate safety
474evaluations. It was found that Cd had little effect on bacterial communities (microbial distributions)
475in a landfill area, as was the case for other sulphurophile elements (Cu and Zn) (Liu et al., 2018).
476Metals such as Cd are suggested to have a limited influence on soil bacterial composition and
477diversity, even at e-waste sites (Jiang et al., 2018). It appears that soil bacterial taxonomic
478composition and microbial diversity are more influenced by soil texture, organic carbon content, and
479pH conditions (Sun et al., 2016; Jiang et al., 2019).

480

481 Volatile Cd species are largely transferred from the MSW sources to fly ash during the incineration
482 process. MSW and coal combustion releases Cd to flue gas and fly ash; the most of gaseous forms
483 are condensed to or adsorbed by particles and thus retained in the fly ash (Zhang et al., 2018b).
484 Cadmium accounts for approximately 20% of the metal availability in fly ash and mostly exists in
485 the non-mineral phase, resulting in its higher mobility and release than other metals (Du et al., 2018).
486 Cadmium mostly exists as particulate matter in flue gas, which is collected in the fabric bag or
487 electrostatic precipitator as fly ash (Ahmad et al., 2018). Metals with lower boiling points, such as
488 Hg (357 °C) and Cd (767 °C), are distributed more commonly in fine fly ash particles. In addition to
489 incineration, other thermal processes (such as pyrolysis, gasification, and hydrothermal treatment)
490 are also employed for the treatment of sewage sludge. Cadmium in sewage sludge was reported to
491 vary from below detection limits to 110 mg/kg and was more commonly found in acid-soluble and
492 Fe-Mn oxides fractions (Udayanga et al., 2018). Cadmium has a greater tendency for volatilisation
493 during incineration and gasification but exhibits moderate volatilisation during pyrolysis and
494 hydrothermal process.

495

496 Incineration temperature has an important influence on the metal composition of fly ash. For
497 example, it was found that higher incineration temperatures (between 1003 °C and 1036 °C) led to
498 235% higher Cd concentrations in the fly ash, as the transfer rates of volatile metals were increased
499 (Shi et al., 2018). Cadmium exhibits a pH-dependent leaching pattern in fly ash, which is soluble in
500 acidic conditions but becomes immobile towards near-neutral conditions (Izquierdo and Querol,
501 2010). Since Fe is oxidised to form (hydr)oxides in the early stage of combustion, the resulting
502 alkalinity of the solid residue delays Cd mobilisation. However, with ageing, neutralisation of
503 alkalinity and dissolution of carbonates and oxides occur and cause a reduction in pH and the
504 subsequent mobilisation of metals. Therefore, industrial sources (e.g., wastewaters and uncontrolled
505 chemical pollution), acid rain, and contact with soil solutions that lead to a lowering of pH may pose
506 the threat of metal release during the storage or disposal of fly ash (Abramov et al., 2018). In

507addition, chloride may enhance Cd solubility via complexation and shift the solubility curve to a
508higher pH. During aging, fly ash forms a high carbonate content, which effectively immobilises Cd
509in the form of otavite (CdCO_3), and the formed calcite and hydrous Fe/Al oxides also present strong
510adsorption, thus controlling the release of Cd (Du et al., 2018). Such adsorption also occurs in the
511case of other mineral phases, such as Fe and Al (oxy)(hydr)oxides and amorphous aluminosilicates.
512Particle size is another factor that effects the leaching behaviour. For example, fine particles (< 2
513 μm) of fly ash were found to contain 77% of the total Cd in all fractions below 16 μm , and thus
514leached more extractable Cd and other metals (Abramov et al., 2018).

515

5163.5 Mercury

517Mercury (Hg) is a heavy metallic element that exists in liquid form under standard conditions. The
518most common forms of mercury include the less reactive volatile Hg^0 species as well as soluble and
519reactive Hg(II) forms, the latter tends to form soluble complexes with numerous ligands and surface
520complexes on solids such as HgS and HgSe (Bartov et al., 2013). With toxic, persistent, and bio-
521accumulative properties, mercury emission may pose a severe risk to the environment and human
522health (Beckers et al., 2019). Mercury is widely used in various household items, such as alkaline
523batteries, button cells, fluorescent lamps, thermometers, and sphygmomanometers, as well as in
524some dental, electronics, and fluorescent-lighting manufacturing industries (Hu et al., 2018; United
525States Geological Survey, 2018). Mercury released into the environment derives from various
526sources, including mercury-containing car switches, coal-fired power plant emissions, and
527incineration of mercury-containing medical devices (United States Geological Survey, 2018). The
528amount of mercury found in sewage sludge is relatively small. For example, less than 0.2 mg/kg Hg
529was measured in several different sources of sludge (Spanos et al., 2014). However, direct mercury
530emissions through gas or water from incineration or landfill of sewage sludge have significantly
531contributed to environmental toxicity. Hence, integrated sewage sludge treatment is environmentally
532and economically recommended (Xu et al., 2014)

533

534Waste-to-energy (WTE) incineration is one of the major ways in which mercury in MSW is released
535into the environment. In China, incineration constitutes 21% of the anthropogenic mercury emissions
536in the Pearl River Delta district, while in north China, much more mercury derived from the use of
537coal (Hu et al., 2018). Mercury in flue gas is mostly present in its oxidised form (75–85% Hg^{2+}),
538with only a small fraction existing in its elemental form (1–20 % Hg^0) (Hu et al., 2018). During
539incineration, the high temperatures in combustion chambers lead to mercury emission from the waste
540as well as the supplementary fuel (i.e., coal) (O'Connor et al., 2019). Other elements present in the
541MSW, such as chloride, cause mercury oxidation and promote the formation of chloromercury
542compounds in the flue gas. After cooling, the reactive and oxidised form (Hg^{2+}) in flue gas can be
543adsorbed onto unburnt carbon particulates and subsequently removed by particulate matter control
544devices or captured by wet scrubbers. In contrast, volatile, chemically inert, and poorly water-soluble
545 Hg^0 cannot be removed effectively from flue gas but can be oxidised to Hg^{2+} by selective catalytic
546reduction devices (Hu et al., 2018). Devices for capturing particulate matter, primarily fabric filters
547and electrostatic precipitators, perform well simultaneously removing mercury bound to particulates.
548Activated carbon is often used as an effective adsorbent for mercury-specific control in incineration
549flue gas in combination with other devices (Hu et al., 2018).

550

551Similarly, Hg derived from coal combustion in coal-fired power plants involves a similar process to
552MSW incineration, and the resulting concentrations in fly ash and the flue gas desulphurisation
553products are also similar (O'Connor et al., 2019). Mercury, ranging from 0.001 to 10 mg/kg in CCRs,
554is mainly bonded to sulphur, chlorine, and carbon as Hg(I) and Hg(II) species (Deonarine et al.,
5552013). Mercury capture increases with a decrease in flue gas temperature and increases with an
556increase in the amount of activated carbon (Hood et al., 2007). Improper disposal of these CCRs
557would potentially transfer mercury into ecosystems such as waterways. For example, substrates in
558ash (such as sulphate) could stimulate the biomethylation of mercury to form methylmercury

559(MeHg) via anaerobic bacteria in redox conditions. As a result, sediments in surrounding aquatic
560ecosystems can be damaged as a result of this bioaccumulative and neurotoxic organomercury
561compound. Research has shown that coal ash disposal has led to elevated concentrations of total
562mercury and methylmercury in the sediments of receiving rivers compared to upstream sediments
563(Deonarine et al., 2013).

564

5653.6. Lead

566Lead (Pb) is a heavy metal with a high density and low melting point. Pb and its compounds are
567poisonous, which may cause atmospheric, aquatic, and soil pollution, and have a significant effect on
568animals and plants. Industrial wastes, commercial wastes, institutional wastes, and residential wastes
569disperse Pb-bearing waste into the environment (Igalavithana et al., 2019; Li et al., 2019).
570Automobile exhaust emissions are the most important sources of Pb in the dust collected from the
571roadside, while non-exhaust emissions are also important sources (Li et al., 2018a). Mine tailings,
572coal ash, smelting waste, and clinker are the main five solid waste. Of these, mine tailings are the
573main source of solid wastes but have the lowest utilisation (Xu, 2018).

574

575Lead smelting slag is mainly derived from pyrometallurgy (De Angelis and Medici, 2012). At
576present, research on the resource utilisation of lead-smelting slag has mainly focussed on the
577recovery of valuable elements, the use of building materials, and the preparation of mine filling.
578Waste lead-acid batteries with lead grids and battery paste are a significant source for Pb recovery.
579Lead grids generally have relatively stable components and can be directly recovered under low-
580temperature smelting, but battery paste needs a more complex recovery process. Pre-treatment
581involves direct and indirect melting, where the battery paste of waste lead-acid batteries is melted at
5821000 °C, or the sulphur in waste lead-acid batteries is removed by adding desulphuriser at relatively
583low temperatures before smelting. The lead sludge or lead paste from desulphurisation is then put
584into the furnace. Lead-contaminated gas, dust emissions, and other forms of pollution may occur if

585this material is not properly handled (Zhang et al., 2019). Typical hydrometallurgy includes leaching
586or desulphurisation, electrowinning or cementation, and subsequent smelting and casting. In the
587leaching process, inorganic acids (such as hydrochloric acid and nitric acid) are used to dissolve
588lead-containing substances or for desulphurisation, after which electrolysis is carried out. After
589electrolysis, more than 99.9% of lead can be obtained on the cathode. Recovery of lead oxide from
590waste lead-acid batteries can be achieved as follows: inhomogeneous surface of active material
591crystals is first removed by oxidation or reduction, and then desulphurised; lead oxide is the
592produced by calcining at 400–500 °C; hydrogen chloride can then be used to recover battery
593materials, while sodium chloride can promote the dissolution of lead sulphate. The leaching rate of
594this method can reach 99%, but its corrosiveness to equipment and impurity removal remains a
595concern.

596

597Lead-containing solid wastes in some developing areas are still accumulating, which results in
598excessive occupation of land (Zhang et al., 2019). Open air stacking of fine Pb-bearing tailings and
599soot particles is likely to release dust and cause air pollution. Piled Pb may be eroded by rainfall into
600the surrounding soils. Even after MSW deposition in the landfill, airborne particulates may contain
601Pb depending on meteorological factors, landfill activity, and waste degradation (Bodihardjo et al.,
6022018). After erosion by rainfall and surface runoff, discharge and dispersion of contaminated water
603can cause soil contamination and subsequent damage to local ecological systems. The Pb-laden
604particulates in the air can cause widespread contamination through atmospheric deposition (Zhou et
605al., 2018). Lead has a highly negative charge and can easily form covalent bonds with iron,
606aluminium, manganese and other oxides, organic matter, and carbonates. When Pb enters plants, it
607firstly affects cell membranes and then influences a series of physiological and biochemical
608processes in cells, which all lead to metabolic disorders. Factors affecting the absorption of Pb by
609plants are diverse and determined by the ecological characteristics and external environment (e.g.,
610pH, oxidation-reduction potential, and chemical interactions). In addition, Pb-entrained particulates

611in the air can enter plants via deposition (Zhou et al., 2018). Human may intake Pb via the
612respiratory tract, digestive tract, and skin. Children are particularly susceptible to atmospheric Pb.
613Lead causes damage to all organs, especially during the childhood growth period. The effects of lead
614on tissues and organs are permanent and irreversible even if environmental pollution is rectified.

615

6163.7 Treatment of metal(loid)-containing wastes

617Conventional waste treatment such as incineration and landfill would cause diverse adverse impacts
618on human health as well as freshwater, marine, and terrestrial ecotoxicity due to the release of metals
619in the waste streams. To address these problems, advancement has been made based on conventional
620treatment facilities. Air pollution control devices (APCDs) are installed to mitigate metal pollution
621from MSW incineration process. Electrostatic precipitators, bag filters, and wet scrubbers are used to
622capture fine fly ash (0.1–1.0 μm) and toxic gaseous compounds in the exhaust, while the coarse ash
623particles (1–20 μm) present in flue gas stream are generally captured by cyclones (Udayanga et al.,
6242018). Fabric filter shows high efficiency in capturing most Cd, Cr, Cu, Ni, Pb, and Zn (Ahmad et
625al., 2018). As the majority of Hg in flue gas passed through the fabric filter to the upstream of the
626wet scrubber and existed in an oxidized form, the wet scrubber could efficiently remove oxidized Hg
627by reduction to elemental Hg, achieving 75-93.2% Hg removal with injection of activated carbon
628(Ahmad et al., 2018; Hu et al., 2018). Nano-structured zinc sulphide as a novel adsorbent also
629effectively captured Hg from flue gas to fly ash with less than 0.01% leaching (Li et al., 2018b).

630

631The addition of absorbent materials during combustion process effectively reduces the metal content
632in the solid residues. For example, the calcium-based adsorbents (i.e., CaO and CaCO₃)
633accomplished volatilization rate of Cr and Zn up to 35.7% and 84.0% for food waste combustion (Ke
634et al., 2019). During the process, the CO₂/O₂ ratio, furnace temperature, and Ca type make a crucial
635influence on the metal removal. Metal recovery from flue gas scrubber sludge by hydrometallurgical
636process obtained recovery rate of Pb, Hg, and Se of 99.6%, 98.9%, and 95.5%, respectively (Xing et

637al., 2018). As for dumping sites with buried wastes, rapid and high contamination in the surrounding
638aquifer is often observed. The installation of permeable reactive barriers can relieve the groundwater
639contamination by adsorption and precipitation, which largely reduced metal concentrations in the
640outflow (Kumarasinghe et al., 2017).

641

642Emerging mitigation and treatment approaches to overcome these problems are summarized in Table
6433. From the perspective of sustainable waste management, waste separation and metal recovery are
644important to alleviate the negative effects from metals in the wastes. Pre-sorting prior to incineration
645can effectively decrease the metal content in source materials and increase the combustion efficiency
646as well as heat values for incineration. A system of source-classified collection of MSW was proved
647to decrease metal content from 1.22% to 0.17% and substantially reduced the leachate toxicity of
648bottom ash (Shi et al., 2018). Policies and regulations contribute significantly to the mitigation of
649PTEs in waste streams. The global Minamata Convention on Mercury since 2013 has facilitated the
650reduction of anthropogenic emissions and releases of mercury over the 140 countries in the world.
651For example, China has implemented several national plans for gradually phasing out the mercury-
652containing products from the markets by limiting mining activities and usage of mercury industrial
653productions (Hu et al., 2018).

654

655In addition to mitigate PTEs contamination in the system design level, metal immobilization is also
656an essential solution if there is further reuse of such waste materials. Stabilisation (e.g., carbonation
657and chemical stabilisation) (Li et al., 2017) and extraction (e.g., acid washing and chemical
658extraction) are frequently used (Beiyuan et al., 2017a; Kirkelund and Jensen, 2018). As an example,
659for chemical extraction, As can be extracted from the solid waste by adding a weak acid salt such as
660ammonium acetate or oxalate or alkaline solutions (Beiyuan et al., 2017b; Kim et al., 2016). High
661liquid-to-solid ratio, pH, and oxidizing conditions are effective for facilitating metal depletion from
662fly ash (Weibel et al., 2017). Chemical leaching and metal recovery can be combined with waste-to-

663energy utilization. Metal desorption via electro-dialytic treatment is also used for PTEs removal from
664incinerator fly ash, which employed ion exchange membrane to remove anions and cations from the
665ash suspension to the concentrate by the applied current (Kirkelund and Jensen, 2018). This process
666was pH dependent, where Cd was only desorbed at acidic conditions. Besides, biosorption involving
667chelation, ion exchange, complexation, adsorption, absorption or micro-precipitation can also act as a
668potential way for remediation. Indigenous fungi achieved a cadmium removal of 72.4% from
669municipal solid wastes leachate at spore concentration of 10^5 , pH of 6.0 and temperature of 45 °C
670(Awasthi et al., 2017). Bioleaching or microbial reduction can be applied for As treatment using
671exogenous bacteria, but the treatment cycle is long (Lee et al., 2015; Yamamura et al., 2008).

672

673In recent years, the concept of green remediation is being extensively explored through stabilization/
674solidification, where contaminated wastes (such as ashes) can be utilized and recycled for secondary
675uses (Wang et al., 2018b; Wang et al., 2019). These practices effectively minimize the cost of waste
676disposal and resource extraction, thus sustainably reducing the environmental footprints and closing
677the waste loop. For example, synthesis of zeolite from coal combustion residues or incinerator sludge
678ash is considered as a safe application to immobilize toxic elements (Feng et al., 2018; Zhang et al.,
6792018a). Besides, diverse combustion ashes such as coal or incinerated ash as well as sewage sludge
680ash, can be utilized for blended cement or concrete block production (Wang et al., 2018c; Yin et al.,
6812018). Cement-solidified fly ash via natural ageing forms the high contents of carbonates and
682iron/aluminium hydroxides, which significantly controlled leaching of Cd, Cu, and Zn (Du et al.,
6832018). Recent research studied contaminated sediment and fly ash for producing non-load-bearing
684masonry units or fill materials for construction uses (Wang et al., 2015; Wang et al., 2018b). The
685microporous structure and high surface area of sediment blocks has effectively immobilized metals
686(such as Ni, Pb, Cu) by the C–S–H gel and hydration products with minimal leachability. However,
687in some cases, mobility of Cr, Cu, Pb and Zn would increase after cement stabilization due to
688dissolution of hydroxides in alkaline condition and presence of chloride, sulphate, and organic

689fractions (Yin et al., 2018). Cement-based materials can greatly stabilize the Pb-smelting slag,
690however, Pb may be released under weak acidic conditions (Angelis et al., 2012). When Pb-smelting
691slag is used as fine aggregate of road material, the mixture of asphalt and slag is formed and the
692shear resistance, drainage capacity, and percolation characteristics basically meet the requirements of
693environmental protection (Saikia et al., 2012).

694

695As an alternative, recycled solid wastes can serve as sorbent material for metal sequestration in
696wastewater treatment. To utilize the high affinity of PTEs to organic matter, a study explored to use
697MSW organic windrow compost to remediate contaminated soil and achieved Cd (89.8%) and Zn
698(64.0%) adsorption capacities (Lima et al., 2018). Industrial wastes such as blast furnace slag, fly
699ash, and red mud are also studied for metal removal from aqueous solutions (Ahmed and
700Ahmaruzzaman, 2016). Biochar, as a waste biomass-derived material, is emerging as a low-cost
701adsorbent for removal of potentially toxic elements such as Cr, Cu, As, and Pb from wastewater and
702stormwater (Zhou et al., 2017; Mohanty et al., 2018; Rajapaksha et al., 2018). In addition to sludge-
703derived biochar which proved to stabilize metal-contaminated soils (Fang et al., 2016; Beiyuan et al.,
7042017c; Yoo et al., 2018), recent advances on tailoring biochar into functional carbon composites
705(Xiong et al., 2017; Sun et al., 2019) shed light on synthesis of novel and sustainable materials for
706environmental remediation.

707

708 **4. Fate of radioactive elements and their treatment**

7094.1 *Uranium*

710With the development and utilisation of nuclear energy, the uranium mining and metallurgy
711industries have developed rapidly. The mining and smelting of uranium have produced a large
712amount of uranium-containing waste rock and tailings, which have become the most serious source
713of uranium pollution. Currently, the world's accumulated more than 20 billion tonnes of uranium
714tailings and more than 40 billion tonnes of uranium waste rock. Uranium has caused relatively

715complicated radioactive pollution; as the grade of uranium ore continues to decrease, the amount of
716waste rock and tailings produced will increase (Bowker et al., 2007). Uranium tailings ponds are
717sites of recovery for slag and other low-level radioactive wastes, which contain a considerable
718amount of radionuclide uranium (Table 4). Thus far, 22 countries in the world are engaged in
719uranium mining and metallurgy production, and there are more than 4,000 uranium tailing ponds.
720Large uranium tailing ponds can store tens of millions of tonnes of uranium tailings, whereas small
721ponds can also store from dozens to millions of tonnes of uranium tailings. The country with the
722largest volume of uranium tailings in the world is Kazakhstan, accounting for 23% of the global
723total, followed by Germany (17%), Ukraine (14%), and the United States (13%) (Bourva et al.,
7242014). In China, uranium resources are mainly found in East, Central, and South China, accounting
725for approximately 80% of the total uranium production. Based on this distribution, 70%–80% of
726uranium resources are in humid and rainy hilly areas. Most uranium deposits are typically located in
727temperate monsoon and subtropical monsoon climates (Déjeant et al., 2014).

728

729Solid waste containing uranium mainly exists in the form of uranium waste rock and uranium
730tailings, which are the main sources of uranium pollution. According to the pollution potential of the
731uranium tailings reservoir and the release potential of uranium from other pollution sources, uranium
732can be divided into the following six fractions: (1) exchangeable (water-soluble); (2) carbonate-
733bound; (3) organic matter-bound; (4) amorphous iron-manganese oxide/hydroxide-bound; (5)
734crystalline iron-manganese oxide/hydroxide-bound; and (6) residual (Liang et al., 2014). The
735exchangeable fraction of uranium refers to non-specifically adsorbed uranium on the surface of solid
736media by outer-sphere weak coordination. This form of uranium is most likely to be released into the
737environment through the effects of rainfall and surface runoff, presenting the greatest potential threat
738to the environment. The carbonate-bound fraction refers to uranium that is adsorbed on the surface of
739carbonates or that is co-precipitated. This form of uranium is highly susceptible to pH change and

740 release into the environment, also posing a potential threat to the environment. Organic matter-bound
741 uranium is chelated with organic matter, which can be released following the oxidative
742 decomposition of organic matter. Amorphous iron-manganese oxide/hydroxide-bound fraction refers
743 to uranium that is adsorbed or precipitated by an amorphous iron-manganese oxide/hydroxide
744 component, which has poor stability and is prone to release under reducing conditions. Crystalline
745 iron-manganese oxide/hydroxide-bound uranium refers to uranium that is adsorbed or co-precipitated
746 by a crystalline iron-manganese oxide/hydroxide component, which is released only under strongly
747 reducing conditions. Residual fraction refers to uranium that is firmly bound to primary or secondary
748 silicate minerals. This form of uranium is not easily released under natural weathering conditions and
749 can be stabilised for a long period of time without any significant impact on the environment.

750

751 A large number of uranium tailings may release pollution via rupturing of the conveying pipe,
752 leakage from the tailings pond, and flooding of the tailings mine dam by heavy rain (Zhijie et al.,
753 2014). In addition to the toxic effects of heavy metals, uranium also produces a certain amount of
754 radioactivity. Since uranium tailings contain all the decaying bodies and uranium remaining after
755 smelting, most ^{230}Th and ^{226}Ra is concentrated in the tailings. The radioactivity of uranium itself is
756 not strong, but its half-life is long and produces decaying daughter products with high radioactivity.
757 Therefore, the radioactive hazard persists for a considerable period of time after decommissioning of
758 uranium tailings. The radioactive contamination of uranium tailings is not as easily detected as other
759 types of pollution, and the sensitivity of different organisms to radioactivity varies widely. Singh et
760 al. (2015a) studied the accumulation of major pollutants in a uranium tailings pond and the affected
761 areas. Different plants grew under the same environment, while the biological concentrations and
762 elemental accumulation in some plant species were more obvious than in others (Singh et al., 2015a).
763 In general, human exposed to the same levels of radioactivity will be much more affected than other

764organisms. The latency of the carcinogenic effect of radiation is less than three years but can be more
765than 20 years. The genetic effects of radiation can also be delayed.

766

767Uranium from the tailings migrates into the soil under natural conditions, causing its accumulation in
768the soil. The concentration of uranium ions in the soil decreases with increasing depth, and the
769affected depth does not generally exceed 30 cm. Uranium is transferred to plants via microbial
770activity. Different species of plants show large differences in their accumulation of radionuclides and
771heavy metals. There are also large differences in the enrichment of uranium in different parts of
772plants. For example, stems and leaves of plants are more enriched than seeds. When uranium
773accumulates to a certain concentration in plants, it will affect normal growth and can result in
774eventual death. More importantly, uranium accumulated in plants enters the human body through the
775food chain. In an experiment, three plants (corn, sunflower, and green pea) were grown in four
776matrix variants for 40 days (Santos et al., 2011). The results showed that the concentrations of U in
777the plants were dependent on plant growth; the concentrations of uranium in the substrates were
778important, and the uranium contents in the upper part and root of sunflower, corn, and pea were
779different. The concentrations of uranium in roots were significantly higher than in the above-ground
780biomass, and the uranium content in sunflower was higher than in corn and pea; the dry matter
781quantity of sunflower was the highest, followed by corn and green pea (Stojanovic et al., 2009). The
782uranyl ion (UO_2^{2+}) that enters the human body can bind to the kidneys and other organs in
783combination with biomolecules, accumulates, and causes necrotic glomerular necrosis, tubular wall
784atrophy, and malignant bone and kidney lesions (i.e., tumours).

785

7864.2. Radium

787Naturally occurring radioactive radium may become concentrated in residues, waste materials, and
788end products of non-nuclear industrial activities such as fossil fuel burning. In particular, energy

789production from fossil fuels such as coal may lead to the enrichment of radium, which significantly
790increases human exposure to radiation. Coal-fired power plants generate large amounts of ash with
791naturally occurring radioactive radium (Figure 2). Radium-226 and Radium-228 are the main sources
792of low-dose ionizing radiation exposure in coal ash. Radium in coal ash would migrate to the soil
793under natural conditions, resulting in accumulation in the soil and subsequent absorption in plants.
794As an alkali-earth element with oxidative state of +2, radium has a higher solubility and is more
795biologically available to plants. Coal fly ash is widely used not only as supplementary material in
796concrete construction but also in road construction and asphalt, leading to a potential risk of
797radioactive contamination. All isotopes of radium are highly radioactive, with the most stable isotope
798being radium-226, which has a half-life of 1,600 years. Radium-228 is also a stable isotope, with a
799half-life of 5.75 years. It was estimated that over 130 million tonnes of coal combustion residues are
800generated in US power plants every year. Globally, coal-based power generation produces over 750
801million tonnes of coal ashes per year. It was estimated that all forms of released radium-226 amount
802to approximately 1.55 TBq y⁻¹, with gross particulate dispersion rates of 7.4 GBq y⁻¹ (18.5 MBq d⁻¹
8031) (Laura et al., 2009).

804

805For coal ashes, based on the modified sequential extraction method, radium can be divided into the
806following six fractions: (1) water-soluble; (2) ion-exchangeable; (3) carbonate-bound; (4) free oxide;
807(5) sulphide-bound; and (6) residual. Approximately 74% of Radium-226 exists in the form of
808residues, a certain amount exists in the carbonate-bound fraction, a small amount in the water-
809soluble, free oxide, sulphide-bound fractions, and almost none in ion-exchangeable fraction.
810Radium-228 also primarily exists in the form of residue (approximately 48%). There is a moderate
811amount in sulphide-bound and free oxide fractions, a small amount in carbonate-bound fraction, but
812none in water-soluble and ion-exchangeable fractions. Although some fractions of the isotopes also
813occur in association with vertical migration and root uptake, these nuclides are generally associated
814with the soil material. Radioactivity decreases with an increase in soil pH value. The highest

815activities of natural ^{226}Ra have been found in soils where carbonates have been leached, while lower
816radioactivity has been found in soils with the highest content of carbonates (Xin et al., 2015).

817

818The release of coal ashes with high levels of radioactivity into the environment has the potential to
819generate resuspended ambient fine particles ($< 10\ \mu\text{m}$). Inhalation of fly ash emitted from the stacks
820and through the ingestion of foodstuffs receiving atmospheric deposition are the main pathways of
821human exposure to radiation. Leaching of contaminants from coal ashes causes heavy contamination
822of surface waters in areas of restricted water exchange, but only trace levels are found downstream
823due to dilution. However, the accumulation of coal ash in river sediments has a potential impact on
824downstream ecology through fish poisoning in anaerobic river sediments. Similarly, the
825accumulation of radioactive material in the ground near to coal-fired plants also results in higher
826levels of radioactivity in soils compared to soil located further away. This affects plant growth and
827possibly further enhances human uptake through the food chain. Emissions from coal-fired power
828plants in gaseous and particulate forms containing nuclides probably accumulate in the surrounding
829environment, thus exposing local populations to radiation (Bozena et al., 2017).

830

8314.3. *Potassium*

832Potassium is an active reducing agent, which is easily oxidized to +1 valence of stable state. Since
833potassium has a high hydration energy, most potassium compounds are soluble in water. Naturally
834occurring potassium (K) is composed of three isotopes, namely K-39 (93.258%), K-40 (0.012%), K-
83541 (6.730%), of which K-40 is radioactive (Meija et al., 2016). Plants cannot distinguish isotopes
836similar in chemical properties when absorbing essential nutrients, so K-40 will be undertaken by the
837plant's roots along with non-radioactive potassium and distributed throughout the plant (James et al.,
8382011). The soil-to-plant transfer factor of potassium in leaves was in the range of 0.49-5.6 with a
839mean of 1.4 (IAEA, 2010). Leached radioactive material may directly or indirectly enter human body

840through drinking water and food, and result in radiation contamination of terrestrial and freshwater
841ecosystems (Aliyu et al., 2015). In fact, potassium is an essential element for human and its average
842concentration in a male adult reaches approximately 2 g per kg of body weight (Gautam et al., 2015).
843Most of the internal radiation dose received by human is attributed to the consumption of food
844contaminated with different radionuclides (Khan et al., 2010). The total ingestion dose from natural
845radioactivity is about 0.21 mSv/y, of which more than 90% is attributed to K-40 (Verma, 1999).
846Potassium-40 is uniformly distributed throughout the tissues and organs of the human body, with a
847biological half-life of 30 days (Rabitsch and Pichl, 2008). The potassium content of the body is
848constant and is not affected by variations in environmental levels due to homeostatic control.

849

850With a half-life of 1.248×10^9 years, K-40 can stably exist in the natural environment. It is,
851therefore, a major source of natural background radiation. There is no commercial or medical use of
852potassium radionuclides, however, K-40 is widely present in various types of solid wastes, mainly
853due to industrial production and construction activities (Sas et al., 2017; Schroeyers et al., 2018).
854Only a very small proportion of solid waste has a concentration of K-40 that exceeds the suggested
855limit of 10,000 Bq/kg (IAEA, 2014). This portion of solid waste is categorised as “technologically
856enhanced naturally occurring radioactive material” (TENORM), i.e., naturally occurring radioactive
857materials that have been concentrated or exposed to the environment as a result of human activities
858such as manufacturing, mineral extraction, and water processing.

859

860Solid wastes containing K-40 are mainly derived from two sources — oil and gas resources
861exploitation and thermal power generation. In the process of oil and gas resource exploitation,
862radioactive potassium is gradually released from the rock matrix and is pumped to the surface with
863water, this finally precipitates as scale and sludge (Gazineu and Hazin, 2008). The largest volumes of
864scale occur in three areas: (1) water lines associated with separators; (2) heater treaters and gas
865dehydrators; and (3) oil stock and water storage tanks. Sludge mainly occurs in tanks, oil and gas

866 separators, dehydrators, storage tanks, and at the bottom of slush pits. According to the US EPA, oil
867 production processes used in conventional drilling generate an estimated 230,000 MT or 141 m³ of
868 TENORM sludge each year. The global average activity concentration of K-40 in soil is 412 Bq/kg,
869 and in the Earth's crust is 750 Bq/kg (UNSCEAR, 2008). The activity concentrations of solid waste
870 generated by the oil and gas industry fluctuate depending on geological factors. For example, in
871 Brazil, the maximum radioactivity of K-40 in scales and sludge reached 144.6 kBq/kg and 53.8 kBq/
872 kg, respectively, while the lowest values could not be detected in the states of Sergipe and Alagoas
873 (Gazineu and Hazin, 2008).

874

875 4.4. Treatment of radioactive elements

876 In summary, the main technologies for controlling radioactive elements are S/S method, backfilling
877 method, containment method, biochemical repair method, and integration of the above. For S/S
878 method, cementitious materials could be employed as agents for the immobilisation of radioactive
879 elements. Supplementary cementitious materials such as fly ash, silica fume, calcined clay were used
880 to reduce carbon footprint and improve the S/S efficiency for radioactive wastes. Utilization of
881 tailings or waste rock containing radioactive elements as backfilling material is also an option. It can
882 not only reduce the cost of using other materials, but also reduce the accumulation of radioactive
883 waste on the ground which expose harms to people. Nevertheless, backfilling method is more likely
884 to contaminate the groundwater body than other treatment methods (Mishra et al., 2014).
885 Containment by covering the tailings can also prevent pollution. It has a wide application range and
886 is suitable for long-term treatment of radioactive tailings (Tan et al., 2012). However, there are
887 problems associated with ecological restoration of the surface and potential harm to animals and
888 plants. Biochemical repair of radioactive waste is a method of adding microorganisms and chemicals
889 (e.g., carbonate/bicarbonate) or planting plants in the tailings (Santos et al., 2011). Choosing suitable
890 plants and harvesting the above-ground parts with excessive accumulation characteristics of specific
891 nuclide relieve the radioactive pollution in the soil (Li et al., 2011). Unlike the backfilling method

892and containment method, the biochemical repair method removes the contaminant instead of
893isolating it. In an integrated approach, the synergistic effects of different methods improve the ability
894to remove/stabilise harmful substances and reduce long-term hazard.

895

896 5. Fate of rare earth elements (REEs) and their treatment

8975. 1. Natural existence of REEs in mineral deposits

898Rare earth elements (REEs), as defined by the International Union for Pure and Applied Chemistry
899(IUPAC), incorporate a group of 17 elements consisting of 15 lanthanides as well as yttrium and
900scandium, covering Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Pm, Sm, Sc, Tb, Lu, Tm, Yb, and Y.
901The term “REY” is sometimes used to denote rare earth elements plus yttrium. REEs are usually
902divided into the light rare earth elements (LREEs), including La through Sm, and heavy rare earth
903elements (HREEs), including Eu through Lu. LREEs are abundant in bastnäsite
904[(Ce,La)CO₃(F,OH)] and monazite [(Ce,La,Nd,Th)PO₄], while HREEs are usually present in
905apatite, cheralite, eudialyte, loparite, phosphorites, rare-earth-bearing (ion adsorption) clays,
906secondary monazite, spent uranium solutions, and xenotime (Duta et al., 2016). Their states vary
907from REY-bearing minerals and glass to fine nanoparticles embedded within the amorphous carbon
908surrounding the glassy particles (Hood et al., 2017). HREEs are preferentially desorbed by acidic
909solutions, have a stronger organic affinity, and present more stability in their complex forms than
910LREEs (Hower et al., 2016). Furthermore, based on the abundance and industrial demand, REEs can
911be classified as critical (Y, Nd, Eu, Tb, Dy, and Er), uncritical (La, Pr, Sm, and Gd), and excessive
912(Ce, Ho, Tm, Yb, and Lu) (Hower et al., 2016).

913

914Their unique structures (4f orbitals) and distinctive properties render REEs critical for various
915industrial applications, including uses in superconductors, photonic device permanent magnet
916materials, etc. (Chen et al., 2019). Moreover, REEs show great prospects for sustainable energy
917fields such as wind power, electric vehicles, and energy-efficient lighting (Tan et al., 2017). REEs

918 show a scattered geological distribution with generally low abundance, unevenly distributed around
919 the world. It is estimated that the total demand for REEs will grow by over 50% by 2020 (UNEP,
920 2009). As the largest export source of REEs, China is meeting more than 90% of the global REE
921 demand, and this monopoly is prompting other countries to develop techniques for identifying and
922 recovering REEs. Recent studies on the fate of REEs in solid wastes are summarised in Table 6.

923

924 Conventional mining includes ore deposits, grinding, and cracking of minerals to produce mixed rare
925 earth oxide concentrates, separation and purification of oxides, and refining to produce industrial
926 grade REEs (Duta et al., 2016). Conventional REEs production requires high temperature and
927 stringent treatment conditions, consuming a high amount of energy as well as generating significant
928 volumes of toxic wastes and acidic wastewater. Moreover, a high content of thorium and uranium in
929 REEs-bearing ores (up to 10%) may result in the emission of radioactive wastes during REEs
930 mining. This appeals for comprehensive disposal regulations (Fathollahzadeh et al., 2018a).
931 Exploitation for REEs is challenging as REE-containing minerals seldom occur in concentrated
932 forms but are usually present together in the same deposits (Tunsu et al., 2015). Similar chemical
933 properties of REEs further add to difficulties in their separation and purification.

934

935 That said, the chemical behaviours of sub-structures, known as the tetrad-effect, are utilised in the
936 recovery of REEs in groups of adjacent elements. This is because the stabilisation energy related to
937 the inter-electron repulsion energy of the electrons can cause the ionic radii to decrease within the
938 lanthanide series, thus resulting in variation in the strength of the complexes that REEs form in
939 aqueous solutions. Hence, heavier REEs create stronger complexes with extractant molecules
940 compared to lighter REEs, while yttrium complexes have an ionic radius comparable to heavy
941 lanthanoids. Nevertheless, it is still challenging to accomplish complete separation of adjacent
942 elements on a large scale (Tunsu et al., 2015). Biotechnology is currently emerging as an
943 environmental alternative for REEs extraction from minerals, with the release of less hazardous

944waste. Bioleaching of monazite has recently been reported using a combination of heterotrophic and
945autotrophic acidophilic microorganisms, which solubilised REEs (Ce, La, Nd, Pr, and Y) up to a
946final concentration of 40 mg/L (Fathollahzadeh et al., 2018b).

947

9485.2 *Recycling from coal combustion residues*

949As REEs ore deposits are often laced with radioactive elements such as uranium and thorium,
950extraction would cause radioactive stockpiles. Other environmental impacts include soil erosion,
951biodiversity loss, land use change, flooding; pollution of air, soil, and water; and crop uptake of
952REEs. All pose severe threats to human health. As a result, waste-derived sources of REEs are
953attracting attention as an alternative approach. Coal fly ash and other combustion/incineration
954residues serve as promising secondary sources for REEs recovery, showing both economic feasibility
955and environmental sustainability. The content depends on the characteristics of coal. For example,
956more than 1,000 mg/kg of REEs has been reported in heating plant stoker ash from Kentucky State
957University, while up to 1,220 mg/kg of REEs was found in stoker ash from a Kentucky power plant
958burning Appalachian Basin coal (Taggart et al., 2016; Hower et al., 2018). Nitric acid digestion is
959generally used for REE extraction. Research has shown that pretreatment with an alkaline agent (e.g.,
960lime roasting) tended to liberate REEs without resorting to hydrogen fluoride digestion, which
961mitigates environmental hazard as well as increasing REEs extractability (Taggart et al., 2016).
962Bioleaching using H₂SO₄ and mixed culture of acidophilic bacteria achieved recovery rates of 76%
963Nd, 52% Sb, and 48% Ce from MSW incineration fly ash (Funari et al., 2016).

964

9655.3. *Recycling from other end-of-life products*

966Urban mining is defined as the "systematic management of anthropogenic resources stocks and waste
967(products and buildings), in the view of long-term environmental protection, resource conservation,
968and economic benefits" (Cossu, 2013). As such, urban mining is emerging as an approach to REEs
969recycling from e-waste and other wastes streams including construction and demolition wastes,

970 combustion residues, sludge, exhausted oil, previously landfilled wastes, and industrial residues
971 among others (Tunsu et al., 2015).

972

973 Notably high concentrations of REEs were detected in the surface waters of the San Francisco Bay
974 area, presumably due to the discharge of refractory magnetic resonance imaging contrast agents
975 (used in hospitals and medical research centres) via wastewater treatment plants. Urban mining
976 might be a useful means of tackling some of the undesired effects of REEs discharge. Unlike the
977 primary REEs ores, recycled REEs material does not contain radioactive substances, implying that
978 health problems can be partially mitigated (Duta et al., 2016). In construction and demolition waste
979 management, up to 100% metal recycling performance is expected for the potential closed-loop
980 waste lifecycle (Kucukvar et al., 2016). Urban mining recovery of technology-critical elements from
981 waste sources such as spent batteries, mobile phones, and magnet motors is a promising strategy to
982 relieve pressure on both resource depletion and environmental pollution (Zuo et al., 2019). Future
983 perspectives may focus on overcoming disadvantages such as collection and dismantling
984 requirements, difficulty of separation by conventional milling operations, and matrix heterogeneity
985 (Tunsu et al., 2015). Furthermore, the adoption of geographic information systems for a
986 comprehensive overview may offer a potential means of developing urban mining (Koutamanis et
987 al., 2018).

988

989 Currently, some chemical and electrical companies (e.g., Umicore and Solvay Rhodia, Mitsubishi
990 Electric, and Veolia Environmental Services) are making efforts to develop REEs waste recycling
991 processes (Duta et al., 2016). Permanent magnets are an important means of recycling REEs.
992 Conventional hydrometallurgy employs chemicals such as strong mineral acids, where magnet alloys
993 are dissolved and the REEs are selectively precipitated with sulphates, oxalates, or fluorides. In
994 contrast, a pyrometallurgical approach requires high-temperature routes and includes electroslag
995 refining, liquid metal extraction, glass slag method, direct melting, and gas-phase extraction methods

996(Binnemans et al., 2015). It is estimated that REEs constitute over 23 wt% of phosphors in
997fluorescent lamps (Tan et al., 2017). Innovative use of mechanical activation as a pretreatment for
998the recovery of REEs from waste phosphors has increased the leaching rates of terbium (Tb),
999lanthanum (La), and cerium (Ce), achieving a significant increase to approximately 90% (Tan et al.,
10002017). Recovery of REEs from other wastes, such as nickel-metal-hydride batteries and lamp
1001phosphors, is technically feasible (Binnemans et al., 2013). For example, ceria can be separated and
1002extracted from glass polishing powder wastes; surfactant collector (sodium oleate) at a neutral pH
1003recovered up to 97.1% from a ceria-based abrasive (Wang et al., 2018b).

1004

1005In addition to conventional hydrometallurgical or pyrometallurgical processes, biotechnologies are
1006emerging as promising alternatives that have less environmental impacts, greater practicality with
1007respect to operation, and higher cost-effectiveness (Işıldar et al., 2019). Biologically induced
1008leaching (bioleaching) from various matrices, biomass-induced sorption (biosorption), and
1009bioelectrochemical systems are being explored for REEs recovery from WEEEs. For example, a
1010bioleaching process employing acidophilic and cyanogenic bacterial strains achieved lanthanum and
1011yttrium extraction at a yield of 80% from shredded WEEE material (Marra et al., 2017).

1012

1013 **6. Prospects and future research**

1014As improper storage and disposal of potentially toxic elements-containing solid wastes could lead to
1015severe degradation of the environment and deleterious consequences to human health, strengthening
1016waste management systems, as well as developing alternative techniques in recycling valuable
1017resources, are required to tackle this problem. With regard to waste streams, measures could be
1018implemented from source reduction, contaminant isolation, resource recycling, and waste
1019stabilization. Therefore, an integrated and sustainable waste management system is recommended, to
1020maximize resource utilization efficiency and minimize the impact on the environment. Research into
1021more effective contamination mitigation measures during waste treatment (such as air pollution

1022control devices for incineration and permeable reactive barrier for landfill) are encouraged, while
1023more advanced techniques are urged to stabilize or safely dispose of these secondary products (e.g.,
1024incineration ashes and combustion residues).

1025

1026Although numerous treatment methods have emerged in recent years, environmental impact and
1027economical cost may still be the major concerns. The prospect for future development should focus
1028on green and sustainable remediation, where implementation is incorporated into sustainable
1029strategies, to restore contaminated solid wastes for productive use, with reduced associated costs and
1030environmental burdens. Green and sustainable remediation also help formulate strategies in
1031exploring the relationships among regulation, economics, and technology. Besides, prior to final
1032waste disposal, the technology-critical, radioactive, and rare earth elements should be prioritised for
1033resource recovery. The development in both technology and management/policy aspects are
1034necessary for meeting the United Nation's Sustainable Development Goals.

1035

1036In addition, the valorisation or re-exploration of valuable resources from existing waste is attracting
1037more attention. Some innovative waste recycling approaches, such as landfill mining (Burlakovs et
1038al., 2018) and sustainable biorefinery from waste biomass (Chen et al., 2017; Cao et al., 2018; Yu et
1039al., 2019), are now emerging to substitute conventional resource exploration and chemical
1040production. In this regard, prudent safety assessment and measures for landfill mining and industrial
1041operation are in need to avoid possible potentially toxic elements leaching to water streams or
1042aerosols during excavation, sorting/separation, and extraction/conversion. Life cycle assessment and
1043techno-economic investigations into various approaches are needed to reduce economic cost,
1044decrease environmental impacts, and devise sustainable strategies for the management of potentially
1045toxic elements-containing solid wastes.

1046

1047 7. Conclusions

1048 This paper critically reviews the fate, risks, and management approaches of potentially toxic
1049 elements in solid waste streams. It summarises the flows and contents of potentially toxic elements in
1050 three typical solid wastes, specifically municipal solid wastes, electronic wastes, and industrial solid
1051 wastes. In particular, toxic metal(loid)s, radioactive elements, and rare earth elements are critically
1052 reviewed with respect to their patterns of transport, transformation, risks in a changing environment,
1053 as well as their corresponding management approaches.

1054

1055 It should be recognised that municipal solid waste is one of the largest components of waste streams.
1056 Incineration and landfill are two major treatment methods for municipal solid waste. For incineration
1057 of municipal solid waste, the potentially toxic elements in the waste are concentrated in fly ash and
1058 bottom ash, whereas for landfill method, the concentrations of the toxic materials also increased in
1059 landfill sites after waste degradation, especially in closed landfill sites. Electronic waste has drawn
1060 increasing attention due to the short lifespan of electrical and electronic products. The potentially
1061 toxic elements in e-waste are not reduced with other organic wastes during landfilling, but instead,
1062 are accumulated, transported, and diffused into soil pore water and thereby threaten the soil
1063 ecosystem. Industrial solid wastes derived from numerous manufacturing and processing activities
1064 are also critical sources of potentially toxic elements. Industrial solid wastes usually contain more
1065 homogeneous forms of potentially toxic elements albeit at high concentrations. Therefore, specific
1066 treatment methods should be designed for different industrial solid wastes.

1067

1068 The commonly encountered toxic metals, chromium, arsenic, selenium, cadmium, mercury, and lead
1069 are comprehensively reviewed in this paper. These elements have similar but distinctive physical,
1070 chemical, and toxic characteristics. Different treatments and recycling methods for toxic metals are
1071 discussed, including metal immobilisation, chemical extraction, electrochemical treatment,
1072 biosorption, and transformation into valuable materials. In addition, radioactive wastes including

1073uranium, radium, thorium, and potassium have become one of the most serious sources of pollution
1074for ecosystems. To tackle the grave environmental risks of radioactive wastes,
1075stabilisation/solidification, backfilling, containment, and biotechnology methods as mainstream
1076technologies are used, whereas recycling methods are not yet mature. For radioactive tailings with
1077relatively low risks, biochemical repair, phytoremediation methods, and other comprehensive
1078methods can be potential treatment technologies. Rare earth elements are mainly produced from
1079natural rare earth ore and industrial wastes (e.g., coal combustion residues and industrial sewage
1080sludge). However, rare earth element mining and extraction processes are usually accompanied by
1081emission of radioactive wastes, which leads to potential environmental risks. As rare earth elements
1082are valuable materials, urban mining is emerging as a means of recycling rare earth elements from
1083solid wastes rather than disposing of them.

1084

1085Based on a holistic understanding of the dynamic fate of potentially toxic elements and a review of
1086existing management options, the development of an integrated and sustainable waste management
1087system is required for resource recovery, waste minimisation, and environmental decontamination. It
1088is imperative to develop novel and advanced treatment technologies for achieving high-efficiency
1089and cost-effective potentially toxic elements removal, recycling, and/or immobilisation methods for
1090different solid wastes. Furthermore, life-cycle analysis and cost-benefit assessment can advise further
1091improvements in energy consumption and the associated carbon footprint. Sustainable remediation,
1092low-carbon treatments, and resource recovery of solid wastes are recommended for meeting the
1093sustainable development goals.

1094

1095 **8. Acknowledgment**

1096The authors appreciate the financial support from the Hong Kong Research Grants Council (E-
1097PolyU503/17 and PolyU 15223517) for this study.

1098

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