



Recovering rare earth elements from contaminated soils: Critical overview of current remediation technologies

Lima, Ana Teresa; Ottosen, Lisbeth M.

Published in:
Chemosphere

Link to article, DOI:
[10.1016/j.chemosphere.2020.129163](https://doi.org/10.1016/j.chemosphere.2020.129163)

Publication date:
2021

Document Version
Peer reviewed version

[Link back to DTU Orbit](#)

Citation (APA):
Lima, A. T., & Ottosen, L. M. (2021). Recovering rare earth elements from contaminated soils: Critical overview of current remediation technologies. *Chemosphere*, 265, Article 129163.
<https://doi.org/10.1016/j.chemosphere.2020.129163>

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

1 Recovering Rare Earth Elements from contaminated soils: Critical overview of current
2 remediation technologies

3

4 Ana Teresa Lima*, Lisbeth Ottosen

5

6 Department of Civil Engineering, Technical University of Denmark, 2800 Lyngby, Denmark

7 *Corresponding author: atmli@byg.dtu.dk

8

9 **Abstract**

10 Rare earth elements (REE) are essential for sustainable energies such as solar and wind power, with
11 rising demand due to the ambitious goal for a circular society. REE are currently mined from virgin
12 ores while REE-rich contaminated soil is left untreated in the environment. Soil remediation
13 strategies are needed that concomitantly cleanup soil and harvest metals that contribute to process
14 circular economy. In this review we aim to (i) define REE concentrations in contaminated soils as
15 well as (ii) identify soil remediation techniques used in remediating REE from soils, emphasizing the
16 ones that extract REE. Current literature lists REE polluted soils in the vicinities of REE mines, coal
17 mines, high traffic roads and agricultural soils (due to REE association with phosphate fertilizers).
18 We first list the conventional separation methods used in the mining industry and their main strategies
19 in extracting/precipitating REE. Solvent extraction is the most commonly conventional method used
20 followed by electrodeposition of REE at high temperatures. We then highlight soil remediation
21 techniques that are used to treat REE. These techniques can be separated into two types: the ones that

22 (a) stabilize REE in soils, and the ones that (b) extract REE from soils. Bioremediation, soil
23 amendments and others offer stabilization of REE, eventually creating a legacy problem since REE
24 keep accumulating in the soil. Soil remediation techniques that achieve REE extraction are a step
25 closer to resource recovery, contributing to the circularity of REE. Techniques such as
26 phytoremediation, soil washing and electrokinetic treatment show promising extraction results.

27 **KEYWORDS:** Soil, extraction techniques, phytoremediation, electrokinetics, REE recovery

28 **1. Introduction**

29 Rare earth elements (REE) are found in “green” technologies, and both play a key role in achieving
30 the ambitious goal of a CO₂ neutral society (Dodson et al., 2012). But as they introduce a solution to
31 one problem, they pressure the mining sector and incentivize pristine mineral resource exploitation.
32 As mining increases, so does the resulting environmental impacts including soil pollution. For
33 “green” technologies to be considered truly sustainable, we need to address the impacts of their life-
34 cycle and guarantee that resource recovery is being applied when possible. From a sustainable and
35 cleaner production standpoint, all types of waste should be harvested and used in their life-cycle
36 (Čuček et al., 2012; Du and Graedel, 2013; El-Hagar, 2007; Markard et al., 2012), including
37 contaminated soil – often enriched with the very same pollutants that we are mining in the first place.

38
39 REE constitute 17 elements: ²¹scandium (Sc), ³⁹yttrium (Y), and the 15 elements of the lanthanide
40 series (Table 1). REE are soft, malleable, ductile, and good electrical conductors, making them
41 suitable for applications in a variety of essential technologies, such as automotive, nuclear, petroleum,
42 electronics (batteries, headphones, TVs), renewable energy, cars, phosphor, ceramics, fertilizers, and
43 even as biological cells stain (Alonso et al., 2012; Binnemans et al., 2013; Bünzli, 2010; Haque et

44 al., 2014; Hemmilä et al., 1984; Massari and Ruberti, 2013; Ramos et al., 2016; Scaff et al., 1969).
 45 Seredin (Seredin, 2010) defined Y, Nd, Eu, Tb, Dy and Er as *critical REE* (Table 1). Beside these,
 46 light-emitting diodes, batteries, and the glass industry are also among REE utilizations (Du and
 47 Graedel, 2013; Wu et al., 2014; W. Zhang et al., 2015). Automotive and wind energy industries are
 48 consequently big consumers of REE, where particularly Nd and Dy are expected to increase 700%
 49 and 2600% respectively over the next 25 years, in a business-as-usual scenario (Alonso et al., 2012).
 50 According to the United States Geological Survey (USGS), REE global production increased from
 51 124.000 tons in 2010 to 167.000 tons in 2018 – an increase of 35% in the last 8 years. Reserves
 52 worldwide amount to 99 million tons (USEPA, 2012). As demand rises, pressure over mining and
 53 pristine resources also increase, since REE recycling is appallingly low. In 2018, Jowitt et al. (2018)
 54 estimated that merely 2,8% of all disposed REE (10.683 out of 375.000 tons) is recycled. REE
 55 resource recovery options are paramount to make green energy truly green.

56

57 Table 1 – Summary of the 17 rare earth elements and their current market value, global production,
 58 main usages, and reuse by element in 2008

Element	Atomic number	Main Usages	% reuse in 2008 (Goonan, 2011)	Price 2015 (USD/kg)*	Price 2019 (USD/kg) ‡
Sc	²¹ scandium	Optical, electronic, aeronautical, automotive and transportation industries (Wang et al., 2011)		5100	440.02
Y	³⁹ yttrium	Ceramics, phosphors (Goonan, 2011), Aid in biochar amendments (Wu et al., 2019)	9.0	8	31.5

La	⁵⁷ lanthanum	Catalysts, battery alloys (USEPA, 2012), ceramics, metallurgy, glass industry, phosphors (Goonan, 2011)	29.9	3	5.25
Ce	⁵⁸ cerium	Catalysts, metal alloys (USEPA, 2012)	32.7	3	5.11
Pr	⁵⁹ praseodymium	Catalysts, ceramics, glass industry, metallurgy, (Neodymium) magnets, battery alloys (Goonan, 2011)	6.8	75	92.4
		Magnets, computers, audio systems, cars.			
Nd	⁶⁰ neodymium	Wind turbines (USEPA, 2012), metallurgy, battery alloys, glass, ceramics, catalysts Long-life atomic batteries (in e.g. satellites), satellite-to-submarine laser communication systems, “cosmic clocks” for the measurement of cosmic rays	18.5	48	55.72
Pm	⁶¹ promethium	lifetime, monitoring of the changes in water content of citrus leaves caused by wetting and drying cycles in the soil, radiotherapy, prevention of dandruff (Elkina and Kurushkin, 2020)	n.f.	n.f.	n.f.
Sm	⁶² samarium	Battery alloys (Goonan, 2011)	0.4	3	13.81
Eu	⁶³ europium	Phosphors (Goonan, 2011)	0.3	339	36.4
Gd	⁶⁴ gadolinium	(Neodymium) magnets, phosphors (Goonan, 2011)	0.6	47	110
Tb	⁶⁵ terbium	(Neodymium) magnets, phosphors (Goonan, 2011)	0.4	562	542.5
Dy	⁶⁶ dysprosium	(Neodymium) magnets (Goonan, 2011)	1.0	278	231.7
Ho	⁶⁷ holmium	Nuclear reactors (Smith, 2001)	n.f.	n.f.	n.f.

Used in infrared absorbing glass and fibre					
Er	⁶⁸ erbium	optic cables (Ebendorff-Heidepriem et al., 2008)	n.f.	n.f.	15.5
Tm	⁶⁹ thulium	X-ray technologies (Bohr et al., 1990)	n.f.	n.f.	n.f.
Yb	⁷⁰ ytterbium	Laser (Ouslimani et al., 2013)	n.f.	n.f.	n.f.
Lu	⁷¹ lutetium	Catalyst (Benndorf et al., 2011)	n.f.	n.f.	n.f.

59

60 Legend:

	Light Rare Earth elements	*	USGS, 2015(Gambogi, 2016)
	Heavy Rare Earth elements	†	http://mineralprices.com/rare-earth-metals/ , 2019
Critical REE	according to (Seredin, 2010)	n.f.	Information not found

61

62 Analogously, these are the activities that constitute the main sources of REE in the soil environment:
 63 mining and mineral processing, industrial production of catalysts, electronics, fertilizers, etc., and e-
 64 waste (including landfill) (Gwenzi et al., 2018). Soil contamination occurs at any stage of the REE
 65 life-cycle: mineral processing, product manufacturing, distributing and lastly disposal. Arguably,
 66 contaminated soil contain REE that can be extracted and reused. Because REE are usually found in
 67 trace quantities in soils, extraction costs may be a challenge for REE recovery (Sholl and Lively,
 68 2016). At the same time, conventional technologies for REE extraction and purification are high
 69 energy demanding and chemical intensive (Maes et al., 2017). The question of quantity is then an
 70 issue: is supply and demand high enough that it is profitable to extract this resource from a more
 71 diffuse medium, with consequently larger extraction times and potentially larger extraction costs?
 72 Once we hit a given threshold, urban “mining” as the act of extracting secondary metal resources
 73 from obsolete – or end-of-life – reservoirs (Krook and Baas, 2013), becomes economically viable.

74

75 Up to date, no country has imposed limit-values of REE in soils. Besides having reduced knowledge
76 regarding REE toxicology and bioavailability (Ramos et al., 2016), contaminant limit-values in soils
77 lack a consistent (European) guideline. REE caused ecological problems studies point to the whole
78 REE spectrum accumulating in bones, liver, and lungs (Hirano and Suzuki, 1996) and damaging
79 disparate organs such as the heart, liver, kidneys and gastrointestinal tract (Rim et al., 2013). In
80 grasshoppers, the ingestion of vegetation with elevated levels of cerium develop muscle paralysis
81 only four-days after consumption (Allison et al., 2015). Despite evidence, REE data was found to be
82 inconclusive and temporary (Ramos et al., 2016; USEPA, 2012). Overall, there is no consensus on
83 REE risk to human health – some report negative effect on humans, some positive – but mainly
84 dosage is key (ionic or chelated forms of REE proved to be less toxic as far as the median lethal dose
85 (LD50) is concerned) (Hirano and Suzuki, 1996). REE influence in soil microbiota is also very
86 limited. A recent study shows that mining considerably reduced microbial soil colonies but not
87 diversity (Liu et al., 2021). Because of this, it is crucial knowing soil REE content and chemical
88 complexation, so that ecological and human risk assessments can be defined.

89

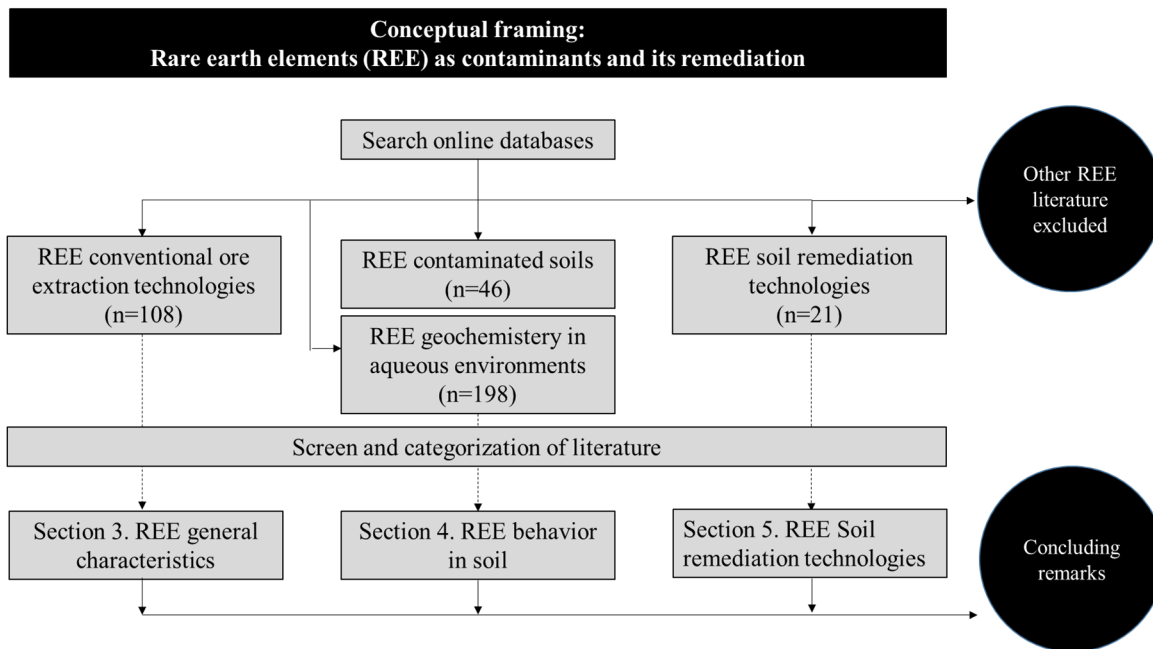
90 In the present study, we look into REE literature retrieved from various databases, published
91 manuscripts and repositories and critically review the latest research on (i) documented pollution of
92 soils by REE; (ii) conventional mining technologies used to extract REE from ore to discuss the
93 option for using these to treat soil; and (iii) verified technologies used to remove/remediate REE from
94 soils. Ultimately, we look at techniques that can simultaneously treat the soil and extract REE, so that
95 resource recovery can ensue and close the cycle. Given the wide awareness about resource extraction
96 and recovery, and the particular rise in REE economic interest during the last decade, this review
97 aims to critically look at REE contaminated soil as a potential resource for REE extraction and define

98 which known remediation techniques are better suited for this extraction. In this respect, the soil
99 remediation industry can play a big role in preventing polluted soils legacy for future generations and
100 creating an opportunity for resource recovery.

101

102 **2. Methodology**

103 This manuscript follows a literature review and research approach. We used “rare earth elements”,
104 “rare earth metals”, “soil contamination”, “extraction technologies” and “soil remediation” as the
105 search keywords and Scopus and DTUfindit as the main search databases. The used books were
106 retrieved from DTU library. The main criteria to include studies in this review consisted in data
107 availability/exposition. Scopus lists only 208 results when using the following keywords
108 simultaneously: “rare earth elements”, “soil contamination” and “soil remediation”. Since the
109 available research articles are within reasonable amount, all were initially considered to be included
110 in this review. Some did not show individual REE concentrations but instead presented the sum of
111 all REE, defeating the purpose of highlighting the priority REE and therefore were not included in
112 this review. As for conventional REE extraction techniques, we selected a few books and scientific
113 articles that guided us through the definitions. Lastly, REE soil remediation techniques were limited
114 – most pertaining to the remediation of water or other effluents. To the best of the authors’ knowledge,
115 all REE soil remediation publications are included in this review.



116
 117 Figure 1 - Research methodology for the current review article and the number of articles found in
 118 each keyword assembly (n=...)

119

120 **3. REE general characteristics**

121 The discovery of REE started at the end of the 18th century. The first element to be discovered was
 122 yttrium and the last one, after 150 years, was promethium (Krishnamurthy and Gupta, 2015;
 123 Voncken, 2016). Promethium, however, can only be artificially synthesized and does not occur in
 124 nature (Voncken, 2016). Most REE were first extracted as oxides, and in French (a major scientific
 125 language in the 19th century), an oxide of an element was known as the “terre” of that element, and
 126 “terre” translates into “earth”, explaining the earth in REE (Voncken, 2016). REE are usually found
 127 in higher amounts in uranium-rich ores, or rocks that present some type of natural radioactivity
 128 (Kraemer et al., 2015; Krishnamurthy and Gupta, 2015; Sahoo et al., 2001). Interestingly, in these
 129 environments, REE can be used as tracers in ground and surface water (Johannesson et al., 1996;
 130 Merten et al., 2005). REE-rich ore exploitation is concentrated mainly in Bayan Obo, China (Izatt et

131 al., 2010)) but REE reserves are evenly distributed between China (36%), Brazil (18%), Vietnam
132 (18%), Russia (10%) (USGS, 2020) and to a lesser extent through the East African rift zones, northern
133 Scandinavia-Kola peninsula and eastern Canada (Chen, 2011; Kanazawa and Kamitani, 2006;
134 Voncken, 2016).

135
136 Chemically, REE are very similar, and most exist solely in a 3^+ oxidation state – a state
137 characterized by an incompletely filled 4f shell (Hasegawa and Kitagawa, 2019). REE versatile
138 nature is due to the lanthanide contraction (Kronholm et al., 2013) – a result of poor shielding of
139 the nucleus by *f*-orbital electrons that translates into an increasingly effective nuclear charge as this
140 orbital is filled (Bünzli, 2010). The lanthanide contraction occurs because the ionic radius of
141 lanthanides decreases as the atomic number increases. Because of these chemical similarities, REE
142 are difficult to isolate from each other. This separation has been described as one of the biggest
143 chemical challenges of the 21st century (Sholl and Lively, 2016).

144
145 REE is still significantly mined from the natural environment and, like most mining operations,
146 generates large quantities of tailings or un-used materials – greater than 90 percent (USEPA, 2012).
147 The conventional techniques used to separate REE from the remaining ore can be found in Table 2.
148 In conventional processes, minerals containing large quantities of REE are first dissolved in
149 concentrated alkalis or acids – after this step, REE separation is difficult to achieve (Fray, 2000) and
150 undergoes a series of physical-chemical procedures at the industrial scale (Kronholm et al., 2013;
151 Voncken, 2016). The lanthanide contraction allows for four modes of separation: selective
152 oxidation/reduction, fractional crystallization, ion exchange, and solvent extraction (Kronholm et

153 al., 2013). Other methods such as mechanical (e.g. magnetic and electrostatic separation) and
 154 chemical processing (e.g froth flotation) are considered inefficient: they must contend with the
 155 complex compositions of mined ores, use large volumes of chemicals, and produce radioactive by-
 156 products as well as large amounts of waste (Sholl and Lively, 2016). Fractional crystallization (or
 157 precipitation), ion exchange and solvent extraction are commercially successful techniques (Fray,
 158 2000). But the largest quantity of REE commercially produced around the world is obtained by
 159 electrolytic methods (Krishnamurthy and Gupta, 2015). *Electrodeposition*, or *Electrowinning*, is a
 160 seasoned technology for the separation of metals at high temperatures (~1050°C) in the
 161 hydrometallurgy industry. The high temperatures are achieved by high voltages or thermal energy
 162 (furnace) and used to maintain the salt bath molten. An applied electric current is passed from anode
 163 to cathode to reduce the salt and deposit the metal at the cathode (Krishnamurthy and Gupta, 2015).
 164 The anode usually consists of graphite, while the cathode is made of a nobler metal such as iron,
 165 molybdenum or tungsten. This is successful for REE recovery at high temperatures and has also been
 166 attempted at “lower temperatures” (>700°C) using cadmium, magnesium, zinc, manganese,
 167 chromium and cobalt as cathode (Krishnamurthy and Gupta, 2015).

168

169 Table 2 – Summary of some large scale techniques used to recover REE from ore, soil and wastes

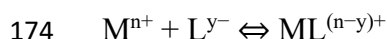
Extraction technique	How it works	REE	Original material with REE	Reference
Cloud point extraction	REE extraction at a cold temperature followed by an equilibrium phase	Nd, Sm, Eu	Standard soils (NIST) and soil from radioactive contaminated areas	(Labrecque et al., 2016; Labrecque and

					Larivière, 2014)
Solvent extraction – Pyrophosphate extraction	Use a mixture of $K_4P_2O_7$ and KOH at pH 11 to leach metals bound to organic complexes, after the step for Mn oxides extraction	La, Ce, pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu	Background, aerially and hydrogenically contaminated soil samples		(Fedotov et al., 2019)
Solvent extraction - sodium hydroxide	Hydrolytic precipitation of REE from sulfate solutions with sodium hydroxide	Y, La, Ce, Pr, Nd, Sm, Eu	High-silica ore		(Karshigina et al., 2018)
Oxidation	Use of $KMnO_4$ and Na_2CO_3 to oxidize Ce(III) to Ce(IV) and then precipitate it	Ce	Monazite ore		(Abreu and Morais, 2010)
Chromatographic separation of REE	SiO_2 nanopowder with a monolayer of DTPA-derived ligands used in chromatography	La, Ce, Pr, Nd, and Dy	Waste liquids		(Ashour et al., 2018)
Antisolvent precipitation	Adding ethanol or 2-propanol (antisolvent) to sulfuric acid REE extractions drive them to precipitate as sulfates	La, Y, Ce, Pr, Nd	NiMH (Recycling) battery		(Korkmaz et al., 2020)
Acid and/or alkaline digestions	Microwave digestion or alkaline fusion	All	Phosphate products		(Dinali et al., 2019)
Electrodeposition of rare earth elements from ionic liquids	Use of organic solvents for the electrodeposition of REE at room temperature	Particularly La, Ce, Sm, Eu, Yb Y, Gd	Ore		(Bourbos et al., 2014)

171 4. REE behavior in soil

172 4.1. Dissolved REE

173 The general reaction between a metal ion and a ligand can be described as



175 And the equilibrium constant may be defined as

$$176 \beta_1 = [ML^{(n-y)+}] / [M^{n+}][L^{y-}]$$

177 where M stands for metal ion, L for ligand, n the oxidation number of M and y the oxidation number of L .

178 Aqueous conditions prevail in soil – surfactants and oils are foreign substances to soil, introduced

179 either by pollution or by an attempt to remediate that pollution. Aqueous REE can be described in

180 the general formula $[Ln(H_2O)_n]^{3+}$ (Cotton, 2006). Ce(IV) is the only tetrapositive REE that is stable

181 in aqueous solutions (Krishnamurthy and Gupta, 2015). Besides the typically trivalent rare earths,

182 only Ce(IV) and Eu/Sm/Yb(II) have low enough redox potential to exist in aqueous solution

183 without decomposing water (Kronholm et al., 2013). If we take REE rich-ore, it is constituted mainly

184 of carbonatite/alkaline rocks, such as monazite ((Ce,La,Nd,Th)(PO₄,SiO₄)), bastnaesite ((La,

185 Ce)CO₃F), and xenotime (YPO₄) (Gambogi, 2016; Kanazawa and Kamitani, 2006). Lanthanides

186 complex with chloride, bromide, bromate, perchlorate, nitrate, acetate, and iodide salts are all soluble

187 in water; sulfates are sparingly soluble; and fluorides, carbonates, oxalates, and phosphates are

188 insoluble (Cotton, 2006). Table 3 summarizes the equilibrium constants of aqueous REE explained

189 in (Cotton, 2006; Giffaut et al., 2014; Luo and Byrne, 2004; Schijf and Byrne, 2004). We can observe

190 that specifically La, Lu, Y and Sc will be in aqueous form as chlorides and bromides, and Lu³⁺ as

191 nitrites (Table 3). Since most REE containing minerals are complexed with phosphates, fluorides and

192 carbonates in the ore, we can assume that most REE will be insoluble in aqueous conditions at neutral

193 pH. Indeed, REE complexation is generally dominated by the formation of carbonate complexes in
 194 alkaline waters with CO₂ partial pressures (PCO₂) greater than or equal to the PCO₂ of the
 195 atmosphere (Luo and Byrne, 2004). This can change however with decreasing pH. Bau (Bau, 1999)
 196 established pH 5 as the value at which most REE are scavenged by Fe oxyhydroxides – pH 4 for
 197 Scandium (Lozano et al., 2019). Below this pH, REE dissolve in solution (Bau, 1999; Lozano et al.,
 198 2020). Specifically, iron and sulfates can form secondary minerals that co-precipitate REE and other
 199 heavy metals (Merten et al., 2005). Indeed, sulfates play a major role in REE scavenging, since at
 200 any given pH values, REE sorption increases with sulfate concentration (Lozano et al., 2019). This
 201 is dependent, of course, of our natural water composition and the abundance of sulfates in our
 202 environment. Relatively more frequent are iron and aluminum, also playing important roles in REE
 203 dissolution, since REE are scavenged by these phases at near-neutral pH conditions (Lozano et al.,
 204 2020).

205
 206 Table 3 – Aqueous equilibrium constants (log β₁) for (a) La, Lu, Y and Sc (Cotton, 2006), (b) Eu
 207 from the Thermochemie database (Giffaut et al., 2014), (c) for SO₄²⁻ in monosulfate complexes from
 208 (Schijf and Byrne, 2004) and (d) for CO₃²⁻ (Luo and Byrne, 2004). Bold numbers explain when the
 209 reaction is favorable.

	I (mol/dm ³)	La ³⁺ (a)	Lu ³⁺ (a)	Y ³⁺ (a)	Sc ³⁺ (a)	Eu ³⁺ (b)
F-	1.0	2.67	3.61	3.60	6.20	6.55
Cl-	1.0	-0.1	-0.4	-0.1	0	-0.05
Br-	1.0	-0.2		-0.15	-0.07	-0.09
NO ₃ -	1.0	0.1	-0.2		0.3	1.21
OH-	0.5	4.7	5.8	5.4	9.0	

SO_4^{2-} (c)		3.21-3.65	3.34-3.47	2.13-3.72
CO_3^{2-} (d)	0	6.98-8.33	7.73-8.88	7.10-8.86

210

211 **4.2. REE complexed with soil minerals**

212 REE, similar to heavy metals, tend to concentrate on the upper layers of a soil profile (Chao et al.,
213 2016; Mleczek et al., 2018). Also similar to most of heavy metals in soils (Alloway, 2013; Bourg,
214 1995), REE are highly dependent of pH (Bau, 1999). In general, alkaline soils have higher REE
215 concentrations (Sadeghi et al., 2013). REE complexation in natural soils is rather independent of
216 redox conditions. Y, La, Ce and Pr show no differences between the oxic and the anoxic phases,
217 being mainly associated with organic matter, or Fe (hydr)oxides and colloids (Brioschi et al., 2013;
218 Fedotov et al., 2019; Marsac et al., 2012; Riedel et al., 2015). Generally, the light REE (La to Sm)
219 reside in trace minerals such as apatite (calcium phosphate mineral), epidote (calcium aluminium iron
220 sorosilicate mineral) and allanite (aluminium-iron silicate mineral), whereas the heavy REE (Gd to
221 Lu) are associated with minerals such as zircon (also silicate mineral) (Aide and Aide, 2012). REE
222 in soils can be mainly bound to organic matter and Fe, Al and Mn oxides (Rabung et al., 2000), but
223 soils with low pH and low Fe oxide content, have REE mainly bound to Mn oxides (Hu et al., 2006)
224 and Al and Fe sulfato-hydroxides (Lozano et al., 2019). REE replace and compete with Ca for binding
225 sites on proteins and compromise membrane stability (Brown et al., 1990). In plants, 40% of the REE
226 are bound to high molecular proteins (Guo et al., 1996). REE isomorphous replacement of Ca happens
227 because of their similar ionic radius, making carbonates a preferable form of REE in soils (this study
228 established a total of 8% of carbonates as a result of the sequential extraction procedure) (Hu et al.,
229 2006). Some studies report that the main mechanism behind incipient REE weathering from rock and
230 soil minerals is carbonation, enhanced by microbial biotic respiration (Zaharescu et al., 2017).

231
232 REE can also be found adsorbed to the surfaces of clay minerals and Fe-Mn oxides (29% of the total
233 REE content) (Hu et al., 2006). Al-oxides were not quantified in this study, but previous studies have
234 determined this phase as an important REE scavenger in natural environments (Lozano et al., 2019).
235 In fact, REE are fractionated due to preferential co-precipitation of heavy REE with amorphous Fe-
236 hydroxides in aqueous solutions (Merten et al., 2005), especially with Fe(oxy)hydroxides. This form
237 of REE is not plant-available, needing to go through reducing conditions to be able to be absorbed
238 by plants (Zhu et al., 1998). And indeed there are reports that list REE to be adsorbed by plants by
239 the same mechanisms as for Fe adsorption, suggesting that REE plant assimilation is intimately
240 coupled with the Fe cycle (Brioschi et al., 2013).

241
242 Aqueous REE form minerals as oxides, halides, carbonates, phosphates (like phosphorite nodules),
243 silicates, borates or arsenates, but not sulfides (Fischer, 2002; Henderson, 1984; Voncken, 2016;
244 Wood, 1990). In the presence of reduced conditions, where sulfide phases occur, REE are found to
245 be bound as carbonates (Merten et al., 2005). In oxidized conditions however, heavy REE are mostly
246 complexed with organic complexes, Mn-oxides or Fe-oxyhydroxides while light REE are more easily
247 desorbed (Brioschi et al., 2013; Fedotov et al., 2019; Marsac et al., 2012). REE complexation with
248 organic matter can form chelates or organic sulfides. The type and composition of organic matter
249 serving as sink of REE seem to have shale origin, and its release controlled by H₂O₂ (Censi et al.,
250 2010; Freslon et al., 2014). REE complexation with organic matter in soils are reportedly low, making
251 up to 4.5% of REE in soil (Hu et al., 2006). The greater fraction of REE, up to 89%, are in a residual
252 form in soils, the strongest bound in a sequential extraction (Hu et al., 2006; Zhu et al., 1998). Results
253 from sequential extraction studies seem to depend highly of soil initial characteristics: soils with

254 higher carbonate content may expect higher REE scavenged in this fraction. While equilibrium
255 constants may give us an idea of the complexes stability in water, sequential extraction studies will
256 clearly define which phases scavenge REE in our soil of study.

257

258 **4.3. REE as soil pollutants**

259

260 Table 4 shows the global average values of REE in soils and compares them to soils where content
261 has been stated as elevated, either because of contamination suspicions or because of their
262 placement close to potential REE sources. Exception to this are the high REE contents found in
263 topsoil resulting from the weathering of carbonatites, an extremely rare magmatic rock found
264 mainly in the islands of Cape Verde and Canary (Marques et al., 2017, 2016). REE concentrations
265 in bulk rock or ore are up to 4 orders of magnitude higher (Britannica, n.d.) to those observed in soils
266 (Table 4). Despite of this, contaminated soils may present values that are in the same order of
267 magnitude as ore, particularly if they are located in mine vicinities. Li et al. (Li et al., 2010) reported
268 soils surrounding REE mines of China with up to 12 000 mg Ce/kg, and Guo et al. (Guo et al., 2013)
269 soils around tailing ponds of Mongolia with circa 7000 mg Nd/kg. Other potential REE enriched soils
270 are farmland or agricultural soils, mainly Ce and La but all REE are found in high amounts in these
271 soils (Dolenec et al., 2007; Jin et al., 2019; Liu et al., 2019). Since the 1970s, REE have been widely
272 used in agriculture as microfertilizers, which improve the quality and yield of agricultural products
273 (Gueroult et al., 2018; Swain and Mishra, 2019). Because phosphate rock is used in the manufacture
274 of detergents, animal feed, and phosphate (PO_4^{3-}) fertilizers, its application in agriculture have
275 enriched soils with hazardous elements including REE (Fayiga and Nwoke, 2016). Road side soils

276 have also been listed as enriched in REE compared to surrounding areas, especially Nd and Ce
277 (Mleczek et al., 2018). Lastly, soils in the vicinity of coal mines of Brazil are also high in REE,
278 particularly Ce (Fdez-Ortiz de Vallejuelo et al., 2017). This reflection is supported by existing
279 literature. As alerted by Hogland et al. (Hogland et al., 2018), landfills are enriched with contaminants
280 and pose environmental risks to soil, water and air, existing 75–100 thousand old landfills and dumps
281 in the Baltic Sea Region alone (Hogland et al., 2018). However, we might want to focus on the critical
282 REE. According to Seredin, the ideal ore contains as much as possible critical REE and as little as
283 possible excess REE (Seredin, 2010). Being Y, Nd, Eu, Tb, Dy and Er the critical REE, we can verify
284 that the highest concentrations are found in tailing soils (Guo et al., 2013) and paddy soils (Liu et al.,
285 2019), with the latter being even comparable to numbers in bulk rock (Kraemer et al., 2015) (Table
286 4).

287

288

289 Table 4 - REE concentrations mg/kg in polluted soil around the world, from different sources. Comparison with the earth's average.
 290 Legend: (Critical REE); n.d. – not determined, not mentioned; ICP-MS – Inductive coupled plasma-mass spectrometer; Spectrometry –
 291 Inductive coupled plasma optical emission spectrometer or atomic emission spectrometer; INAA – Instrumental neutron activation
 292 analysis

Type of soils	Location	Description	Quantification technique	Ce	Eu	Gd	La	Nd	Pr	Sm	Dy	Er	Ho	Lu	Sc	Tb	Tm	Y	Yb	Reference
Road side soils	Poland	Luvisols loamy sand texture	Spectrometry, HNO ₃	9-18	0.08-0.15	1.11-1.54	2.66-3.86	8-13	0.67-0.86	0.01-0.03	0.41-0.56	37-47	0.01-0.04	0.09-0.15	0.58-0.71	0.13-0.23	0.88-1.05	1.79-2.24	0.18-0.34	(Mieczek et al., 2018)
Mine vicinity soils	China	Calcareous, saline, alluvial plain	ICP-MS, n.d.	93-12169			40-6905	42-5726	10.6-1614											(Li et al., 2010)
Paddy field soil	China	Paddy field soil	Spectrometry, HNO ₃ /HCl/HClO ₄	295	4.4	9.4	275	167	74.7	30.4	3.7	6.5	2.9			2.9		37		(Jin et al., 2019)
Red yellow earth, abandoned	China	Clay bottom	ICP-MS, HNO ₃ /HClO ₄ /HF/HCl	157-195	1.7-7.3	11-47	60-247	38-187	11-54	7.9-38	11-46	7.0-28	2.4-9.7	0.8-3.9		1.7-7.7	0.98-4.3	139-282	5.5-27	(Chao et al., 2016)

mine area																				
Marine discharge zone	Tunisia	Marine, sandy, high salinity	Spectrometry, HNO ₃ /HCl	11.5-30.3			9.7-22.5	7.11-20.5									8.6-18.6			(Jalali et al., 2019)
Artificial soil	Brazil	10% organic, 20% clay, 70% sand	Spectrometry, HNO ₃	(up to) 329.2																(Moreira et al., 2019)
Farm land and tailings soils	China	Sites monitored for 6, 9 and 14 years	ICP-MS, lithium borate melting	43.8-363	0.38-6.74	2.50-37.8	14.7-258	11.5-223	2.95-60.5	2.50-39.4	1.80-25.7	1.15-19.3	0.36-6.71	0.23-2.15		0.31-5.45	0.19-2.59	8.23-327	1.47-15.0	(Liu et al., 2019)
Soils around coal mining areas	Brazil	Fe-rich tropical soils	ICP-MS, HNO ₃ /HClO ₄ /HF	70.7-123	0.5-1.2	4.9-8.1	28.9-48	29.3-51.4	7.00-11.9	6.6-9.8	3.6-5.5	1.8-2.6	0.5-1.0	0.5-10.3-20.3	0.5-1.0		0.5-17.5-26.5	2.1-3.1		(Fdez-Ortiz de Vallejuelo et al., 2017)
Paddy soil	Macedonia	Paddy Kočani field soil	ICP-MS, lithium metaborate/tetraborate	41-103	1.0-1.91	4.04-7.84	18-50	21-45.9	4.75-11.5	4.30-8.70	3.97-7.86	2.25-4.3	0.76-1.52	0.32-0.65	12-21	0.67-1.47	0.37-0.69	24.1-46.6	2.22-4.2	(Dolenec et al., 2007)

			and dissolution in nitric acid																	
Forest, pasture , field garden soils	Russia	Several locations	ICP-MS, HNO ₃ -HF- HClO ₄	67.2- 72.0	1.6- 1.64	4.5- 5.0	37.7- 40.0	31.7- 33.1	8.65- 9.2	5.86- 6.16	3.6- 3.85	1.79- 1.91	0.68- 0.77	0.27- 0.30		0.85- 0.91	0.31- 0.36		1.78- 1.85	(Sahoo et al., 2001)
Bulk rock	Ocean Island, Mid- Ocean Ridge Basalts	Bulk rock trace element compositi on	ICP-MS, HNO ₃ -HF- HClO ₄	8.87- 147. 4	0.38- 3.23	2.26- 18.8	2.92- 67.3 6	8.1- 60.1 6	1.48- 17.6 1	1.87- 16.4 4	2.46- 26.2 5	1.56- 16.4 3	0.54- 5.41	0.22- 2.22		0.37- 3.73	0.22- 2.15	18.0- 160. 7	1.56- 15.6 4	(Kraemer et al., 2015)
Tailing s soil	Mongo lia	Soil around tailing reservoir	n.d.	7.14- 23.6	36.2- 94.2		3112 - 1114 5	2552 - 6855	1467 - 4568	210. 8- 582. 2								63.2- 136. 2		(Guo et al., 2013)
Arable topsoil	Germa ny	Floodplai n, river Oker	Spectromet ry,HNO ₃ - HF-HClO ₄	65.1	1.28	5.2	30.1	27	7.43	5.8	4.5	2.3	0.8	0.3		0.7	0.3	20.9	2.3	(Riedel et al., 2015)
Topsoi l from	Côte d'Ivoir e	Non specified	ICP-MS, HNO ₃ - HClO ₄	25.9- 158. 5	0.63- 1.51	2.68- 5.62	11.6- 36.7	14.4- 30.6	3.51- 7.51	2.87- 5.8	2.59- 4.30	1.60- 2.53	0.51- 0.87	0.22- 0.4		0.45- 0.81	0.2- 0.38		1.39- 2.55	(Sako et al., 2018)

Tongo n mine																				
Volcanic soils	Panya m, Central Nigeria	Study used 7 soil samples from the volcanic province	ICP-MS, HNO3-HClO4	89.1-141	1.3-2.9	4.7-8.7	36.5-55.7	30.4-52	8.7-14.3	5.8-10.2	3.6-6.6	1.7-2.7	0.7-1.1	0.1-0.2		0.6-1.2	0.2-0.4		1.3-1.8	(Lar and Gusikit, 2015)
Topsoil	Cape Verde	Study based on 43 topsoil (0-20 cm)	INAA	265	6.13		149	122		20.3	11.3			0.53	16.6	2.29			4.6	(Marques et al., 2017, 2016)
Ash tuff and spatter	Cape Verde	Concentrations in the parent material	INAA	1827	32.1		1070	707		80.2	64.0			3.33	2	11.9			26.7	(Marques et al., 2017, 2016)
Average of European soils	Europe	Agricultural and grazing soils	ICP-MS, HNO3-HClO4				13.6-14.3													(Sadeghi et al., 2013)
Luna soil	Moon	Luna soil	INAA, Gamma-ray spectrometry	20.3-38.5	0.96-2.3		6.4-12	11.1-25		3.2-7.3				0.37-0.78		0.66-1.5	0.35-0.78		2.3-5.4	(Jérôme and Philippot, 1973)

Earth's crust	Global average	All	Estimates	60-66	1.2-2.1	5.4-6.1	30-35	28-40	0.08-9.1	6.0-7.0	3.0-4.5	2.8-3.5	1.2-1.3	0.5-0.8		0-9-1.2	0.48-0.5	31-33	3.1	(Greenwood and Earnshaw, 2006; Taylor, 1964)
---------------	----------------	-----	-----------	-------	---------	---------	-------	-------	----------	---------	---------	---------	---------	---------	--	---------	----------	-------	-----	--

293

294 **5. REE Soil remediation technologies**

295 Remediation strategies for metal contaminated soil are, in general, limited. Most hazardous metal
296 contaminated soil is deposited in landfill (Hogland et al., 2018), being even mandatory in countries
297 like Denmark (Council of Ministers, 2019). In section 3.3, we saw that REE enriched soil is mostly
298 located in the vicinities of mining sites and in farming areas due to the use of phosphate-base
299 fertilizers (Table 4). Because agricultural soils are continually under scrutiny, they have been tested
300 for many amendment strategies to reduce or eliminate the mobile fraction of several metals. Table
301 5 summarizes the few soil remediation techniques that can be found in literature. To complement
302 this list, some techniques used to treat acid mine drainage and pure ore were added to the list in
303 order to flag potential techniques for REE treatment. Generally, we can divide soil remediation
304 strategies in two categories: *stabilization* techniques and *extraction* techniques. We explore both in
305 soil REE remediation.

306

307 Several *stabilization* techniques are renowned and widely used remediation technologies: soil
308 amendments, bioremediation, and ecosystem-based solutions such as constructed wetlands.
309 Amendments consist in adding foreign chemicals or amendments to the soil environment (Adriano
310 et al., 2004; Dresel et al., 2011). The principles of such strategy consist in adding the amendment
311 to the soil so that contaminants are adsorbed onto it or precipitated as new solids. Several
312 amendments have been used REE remediation: phosphate (Jin et al., 2019), biochar (Riedel et al.,
313 2015), alkalinity (Ayora et al., 2016) including hydroxide complexation (Wu et al., 2019), nano-iron
314 (Su, 2017), and coal fly ash (He et al., 2019). Ecological engineering strategies have also been used
315 to polish mining or agricultural runoff, such as constructed wetlands (Prudêncio et al., 2015).

316 Constructed wetlands are designed to retain runoff aqueous contaminants in the wetlands' organic-
317 rich, highly reactive soil (Tromp et al., 2012). Bioremediation uses bacteria *B. licheniformis* to
318 form a La(III) amorphous phosphate that over time converts into a scorpion-like nanomineral
319 monazite (Cheng et al., 2018). All of these technologies have one thing in common: they trap and
320 keep REE (and other metals) bound to the soil. This approach can be efficient and cost effective,
321 but should be seen as a short to mid-term solution. In spite of being good sinks for contaminants
322 and reporting high remediation efficiencies, the *stabilization* techniques only immobilize them and
323 keep them in the soil environment until environmental conditions become prone for the
324 contaminant to leach once again – such as changing soil pH or redox conditions due to spills,
325 fluctuating water table, soil sealing, etc. In fact, soils keep being “enriched” with them to the point
326 that soils themselves become sources of contaminants.

327

328 Stabilization techniques make good use of knowledge about REE complexation in natural
329 environments. Most mined REE deposits are located in carbonatites and other alkaline magmatic
330 intrusive rocks (Ayora et al., 2016). This leads to the conclusion that REE would be mainly found as
331 carbonates in soils, but also oxides, phosphates, silicates and fluorides (Chakhmouradian and Zaitsev,
332 2012). This assumption contradicts other studies that performed sequential extraction analysis in soils
333 and found that up to 89% of REE are found in a residual form in soils, namely silicates (Hu et al.,
334 2006; Zhu et al., 1998). Indeed, soils are heterogeneous in nature, with reported REE bound as
335 carbonates (Merten et al., 2005), adsorbed by organic matter, Fe (hydr)oxides and colloids (Brioschi
336 et al., 2013; Fedotov et al., 2019; Marsac et al., 2012; Riedel et al., 2015). Aide & Aide (2012)
337 explained the difference: the light REE (La-Sm) have greater affinity to calcium phosphate minerals
338 (apatite), calcium aluminium iron sorosilicate minerals (epidote) and alluminium-iron silicate mineral

339 (allanite), while the heavy REE (Gd-Lu) associate with silicate minerals (zircon). Since REE are
340 notorious for its difficult separation (Sholl and Lively, 2016), it is understandable why authors resort
341 to stabilization strategies. Furthermore, investigations regarding geochemical REE sorption on to the
342 surfaces of minerals explain that calcite, apatite, and iron oxyhydroxides (goethite and magnetite)
343 can significantly reduce the mobility of REE (Edahbi et al., 2018). Conversely, aqueous complexation
344 would increase the mobility.

345
346 However, *extraction* remediation techniques do the opposite of stabilization ones: they remove and
347 isolate the contaminants from the soil environment. We found three main types used in REE
348 remediation in the literature: leaching techniques, phytoremediation and electrokinetic based
349 remediation.

- 350 • Leaching techniques aim at desorbing, dissolving and leaching the target contaminant with
351 biosurfactants saponin and rhamnolipid (Li et al., 2017; Zhou et al., 2017) but also with nitric
352 acid, citric acid, and EDTA (Tang et al., 2017). REE removal efficiencies are high, namely
353 with EDTA (approx. 70%, Table 5) (Tang et al., 2017). Because leaching is usually carried
354 out via acid solutions, damage of soil physical-chemical structure ensues and soil integrity
355 may be compromised (Zhou et al., 2017). Leaching should then be seen as an option as ex-
356 site treatment when excavation and soil landfilling is seen as the only site remediation option.
- 357 • Phytoremediation, another extraction remediation technique, extracts and transfers REE to
358 the plants (Chour et al., 2018; Liu et al., 2018; Qin et al., 2019; Wu et al., 2013). This
359 technique is already being used at large-scale in the vicinities of mining areas in China,
360 reporting high removal yields (up to 96% of REE) (Chour et al., 2018; Liu et al., 2018; Qin
361 et al., 2019; Wu et al., 2013). Besides *P. americana* exhibiting a slight preference for

362 accumulating light REE (Liu et al., 2018), no further information is given about plant uptake
363 of individual REE. Plants are known to be incinerated after adsorbing REE in these mining
364 areas (Qin et al., 2019), but no further information is given regarding the fate of REE or its
365 recovery from the resulting waste ashes.

366 • Electroremediation has been widely used in the treatment of soils, wastes, sediments and
367 others for metals and organic contaminants (Lima et al., 2017; Ottosen et al., 2008, 1997).
368 The few found studies on REE extraction are from contaminated soil (Pires et al., 2019) and
369 pure ore (Maes et al., 2017). When compared to the aforementioned techniques,
370 electroremediation does provide lower removal yields (max 42%, Table 5).

371 Both techniques transfer REE to another form: phytoremediation to the plants themselves and
372 electroremediation to the electrolytes. This implies also waste to handle. While plants are normally
373 incinerated, generating further hazardous wastes, electroremediation already provides REE in a
374 dissolved or precipitated form. This option could be further explored to achieve circular recovery of
375 REE back into production.

376
377 All *extraction* techniques use acid to dissolve soil minerals and mobilize REE: nitric acid, citric acid,
378 and EDTA in leaching (Tang et al., 2017); nitric acid in the case of phytoremediation (Chour et al.,
379 2018); citric acid (Maes et al., 2017) and acetic acid (Pires et al., 2019) in electroremediation. REE
380 are only soluble in the form of chlorides, bromides, nitrates, bromates, perchlorate, acetate, sulfates,
381 and iodide salts (Cotton, 2006; Krishnamurthy and Gupta, 2015). REE speciation is not explored in
382 the studies mentioned in Table 5, but REE are expected to be complexed as citrates when in contact
383 with citric acid, nitrates when in contact with nitric acid, acetates when in contact with acetic acid,
384 and a metallic complex when in contact with EDTA (C₁₀H₁₆N₂O₈). EDTA complexation showed the

385 highest desorption rates (Tang et al., 2017). Although REE sulfate complexation has been ambiguous
 386 regarding solubility potential (Cotton, 2006), recent studies showed that sulfide oxidation increase
 387 the leaching of REE due to low pH (poor adsorption) and aqueous sulfate complexes (Edahbi et al.,
 388 2018; Lozano et al., 2019). From conventional ore separation technologies, we know that REE can
 389 be separated by controlling their oxidation state and taking advantage of the difference in volatility
 390 between the di- and trihalides (Uda et al., 2000). More research is needed in this direction.

391
 392 Table 5 – Summary of the main lab and vase scale techniques found in literature that are used in the
 393 remediation of soil contaminated with REE.

Type of remediation	Description	Efficiency	Reference
Phosphate amendments	Pot experiment with field collected paddy soil was tested with phosphate rock, superphosphate, bone char, and calcium magnesium phosphate	Reports 30-96% decrease of mobile REE in soil solution	(Jin et al., 2019)
Coal fly ash amendments	Pot experiments tested different rates of coal fly ash on plant growth	The presence of coal fly ash in soil negatively affected plant growth	(He et al., 2019)
Passive remediation technique (from acid mine drainage)*	Uses alkalinity basaluminite residue (up to pH 9) to precipitate divalent metals	All dissolved REE were retained in basaluminite and calcite passive remediation systems	(Ayora et al., 2016)
Engineered magnetite (Fe ₃ O ₄) nanoparticles	Fe ₃ O ₄ and oleic acid are used for adsorbing REE	100% performance for europium stabilization	(Su, 2017)
Bioremediation*	Bacillus licheniformis to adsorb lanthanum and subsequent	The amorphous material was converted into scorpion-like	(Cheng et al., 2018)

	mineralization process in contaminated water	monazite (LaPO ₄ nanoparticles) in a month	
Phytoremediation	With either <i>Dicranopteris dichotoma</i> , <i>Phytolacca Americana</i> from contaminated soil	240 to 300 kg REE ha ⁻¹ (Liu et al., 2018); extraction of REE from plants with 70–96 % yield (Chour et al., 2018)	(Chour et al., 2018; Liu et al., 2018; Qin et al., 2019; Wu et al., 2013)
Bio-surfactant leaching	(Artificially contaminated) Soil column-leaching using saponin and rhamnolipid	35-58% La, 26-57% Ce, 31-64% Y and 30% Eu	(Li et al., 2017; Zhou et al., 2017)
Soil washing to extract REE	Contaminated cropland soil batch experiments using nitric acid, citric acid, EDTA	REE total recovery of 70.96%, 64.38%, and 62.12% by EDTA, nitric acid, and citric acid, respectively	(Tang et al., 2017)
Electrokinetic remediation	Application of an electric field to artificially contaminated soil (or waste)	42.6% Y recovery in the cathode with acetic acid (Pires et al., 2019)	(Pires et al., 2019)
Microbial leaching+electrochemical extraction [†]	Recycled citric acid from microbial digestion was used to pre-treat monazite (ore), followed by electrochemical treatment	28-33% Neodymium was extracted by the combination of citric acid leaching and electrochemical extraction.	(Maes et al., 2017)

394

395 Legend:



Stabilization of REE in soils

Extraction of REE from soils

* Example used in the remediation of acid mine drainage

† Example used in the treatment of ore

396

397 **6. Considerations for future Rare Earth Elements remediation**

398 REE are deemed to rise in demand up to 2600% over the next 25 years (Alonso et al., 2012).
399 Seredin (Seredin, 2010) considers Y, Nd, Eu, Tb, Dy, and Er the most critical for industrial
400 production, with Tb and Dy extraction being the most costly (Table 1). Currently pristine ore
401 mining continues to be used as the main source of REE while only 2,8% of all disposed REE is
402 currently being recycled (Jowitt et al., 2018). To achieve circular economy and a truly sustainable
403 technological society, REE extraction and recovery need to be addressed. Soil contaminated with
404 REE can be a potential source, so that valuable mineral resources are not lost in its production
405 series. We believe that *soil remediation strategies are needed that simultaneously cleanup soil and*
406 *harvest REE, contributing to a circular economy.* Electroremediation and phytoremediation seem
407 to fit this goal.

408

409 Soil remediation technologies that explore resource recovery should be underscored in the current
410 pursuit for a sustainable and circular economy. Previous reviews singled out electroremediation
411 and phytoremediation as remediation technologies able to remove in-situ metals from soil
412 (Cameselle et al., 2013; Lima et al., 2017). All other remediation technologies are either difficult
413 for field applications and/or environmentally costly (Lima et al., 2017). Electroremediation has
414 also side-effects, namely pH fluctuations near the electrodes and consequent alteration of soil
415 mineralogy, hydrogen and chlorine gas generation at the electrodes and/or other unpredicted redox
416 reactions (Acar and Alshawabkeh, 1993). But these side-effects have been used combined with
417 other remediation technologies such as in-situ chemical oxidation (USEPA, 2006), bioremediation
418 (Secord et al., 2016) and phytoremediation (Cameselle et al., 2013; Kubiak et al., 2012), with

419 successful results. Section 4 reflected on two types of REE remediation strategies: stabilization
420 and extraction. Even though some of the stabilization techniques present high removal efficiencies
421 at the laboratory scale (Table 5), REE remain in the soil. With time, soils will become saturated
422 and contamination of adjacent media will ensue – namely groundwater and surface water.
423 Remediation techniques with extraction capability are then key for achieving resource recovery
424 for both soils and REE: we can concomitantly (i) extract and isolate REE and reintroduce it back
425 to production and (ii) clean-up the soil.

426 Techniques with extraction capability are then key for achieving resource recovery for both soils
427 and REE: we can concomitantly (i) extract and isolate REE and reintroduce it back to production
428 and (ii) clean-up the soil.

429

430

431 Electroremediation has great potential to achieve this ambitious goal. If we consider the industrial
432 scale equivalent – electrowinning or REE electrodeposition at the electrodes at high temperatures
433 (Table 2)– we can observe the great impact the technique has made in the metallurgic industry
434 (Krishnamurthy and Gupta, 2015). At room temperature, however, electrowinning has shown
435 complications because of water electrolysis (McCoy, 1941) that has been recently been bypassed
436 by the introduction of membrane separation technologies (Scarazzato et al., 2017). Scarazzato et
437 al. (2017) introduced membrane technologies as a cleaner alternative solution for metal
438 electrodeposition in aqueous, room temperature, solutions and discuss in detail the evolution of
439 cleaner production strategies in the electroplating industry, including electrodialysis (electric
440 current applied to aqueous media using membranes to separate ions). Indeed, electrodialysis has

441 also been used by Maes et al. (2017) in REE extraction from ore with good results. We recommend
442 prior to start remediation efforts to identify REE speciation in soil – through a simple method of
443 sequential extraction – to understand how strongly REE are bound to soil minerals. Establishing
444 how mobile REE are will predetermine extraction amounts and soil integrity after remediation. As
445 an estimation, the larger the REE fraction bound to the residual phase is, the more soil mineral
446 dissolution remediation has to achieve to extract REE. With more research and applications,
447 electroremediation can present a sustainable (Lima et al., 2017; S. Zhang et al., 2015) and holistic
448 technology for the recovery of critical metals such as REE.

449

450 REE ore extraction costs in USD per kg of extracted REE vary significantly, depending on the
451 impending REE (Table 1). However, these are the only numbers that we can compare to resource
452 recovery achieved through unconventional, remediation methods. The only study found that lists
453 costs pertains to the combination of bioleaching with electroremediation (Maes et al., 2017). The
454 study points 314 USD/kg as Nd extraction costs (Maes et al., 2017). If we compare this number to
455 the 55 USD/kg Nd conventional extraction costs (Table 1), we conclude that this number is about
456 6 times higher. We can also argue that the mining extraction Nd costs are highly underestimated,
457 since they do not reflect the true environmental costs of the extraction. In addition, 314 USD/kg
458 of Nd is definitely in the ballpark of REE costs, ranging from 5.11 USD/kg for Ce to 542 USD/kg
459 for Tb (Table 1). So, the combination bioleaching and electroremediation (Maes et al., 2017) can
460 definitely be seen as a promising cost-effective alternative to REE extraction.

461

462

463 **Conclusion**

464 This paper reviews technologies that simultaneously remediate REE polluted soils and extract and
465 recover REE. While they are contaminants in the soil environment, REE and metals are in high
466 demand in industrial and technological applications. In fact, we urge that remediation strategies
467 should aim to recover and recycle REE (or other valuable metal contaminant).
468 Electroremediation and phytoremediation seem to fulfil this criteria and fit the sustainable and
469 circular economy future ambitions.

470 REE polluted soils are found in connection with REE mines, coal mines, agricultural soils,
471 landfills, and roads. Agricultural soils enrichment are a legacy of phosphate fertilizers, since REE
472 are high in these. Knowledge of REE toxicology and bioavailability is limited, and subsequently
473 limiting values in soils are missing. The reported concentrations in the polluted soils are lower
474 than in the ores from where the REE are extracted, and the binding mechanisms to the soil differs.
475 But in spite of the reported concentrations being lower than in the ores from where the REE are
476 extracted, soils can present a good albeit dilute source of REE. Consequently technologies other
477 than the conventional REE separation methods used in the mining industry are needed. Thus, the
478 major future research focus should be on REE extraction from the polluted soil, targeting mainly
479 the critical REE (Y, Nd, Eu, Tb, Dy and Er). Literature on REE remediation polluted soil is very
480 scarce. The strategies are extraction or immobilization. The latter involves bioremediation and soil
481 amendment with chemicals or particulate materials, which adsorb REE, but these strategies
482 counteracts the aim of recovery. Phytoremediation, soil washing and electrokinetic treatment have
483 all shown ability to simultaneously mobilize and extract REE and remediate soils, and this can
484 form the base for future development of (combined) methods for REE recovery from polluted sites.

485

486 **Acknowledgements**

487 The research leading to these results has received funding from the European Union's Horizon
488 2020 research and innovation program under the Marie Skłodowska-Curie grant agreement no.
489 713683 (COFUNDfellowsDTU).

490

491 **References**

492 Abreu, R.D., Morais, C.A., 2010. Purification of rare earth elements from monazite sulphuric
493 acid leach liquor and the production of high-purity ceric oxide. *Miner. Eng.* 23, 536–540.
494 <https://doi.org/10.1016/j.mineng.2010.03.010>

495 Acar, Y.B., Alshawabkeh, A.N., 1993. The principles of Electrokinetics. *Env. Sci Technol* 27, 1–
496 5.

497 Adriano, D.C., Wenzel, W.W., Vangronsveld, J., Bolan, N.S., 2004. Role of assisted natural
498 remediation in environmental cleanup. *Geoderma* 122, 121–142.

499 Aide, M.T., Aide, C., 2012. Rare Earth Elements: Their Importance in Understanding Soil
500 Genesis. *Int. Sch. Res. Netw. ISRN Soil Sci.* 2012, 11. <https://doi.org/10.5402/2012/783876>

501 Allison, J.E., Boutin, C., Carpenter, D., Ellis, D.M., Parsons, J.L., 2015. Cerium chloride
502 heptahydrate (CeCl₃·7H₂O) induces muscle paralysis in the generalist herbivore,
503 *Melanoplus sanguinipes* (Fabricius) (Orthoptera: Acrididae), fed contaminated plant tissues.
504 *Chemosphere* 120, 674–679. <https://doi.org/10.1016/j.chemosphere.2014.09.058>

505 Alloway, B.J., 2013. Heavy Metals in Soils, Heavy Metals in Soils. Springer, Dordrecht.
506 https://doi.org/10.1007/978-94-007-4470-7_10

507 Alonso, E., Sherman, A.M., Wallington, T.J., Everson, M.P., Field, F.R., Roth, R., Kirchain,
508 R.E., 2012. Evaluating rare earth element availability: A case with revolutionary demand
509 from clean technologies. *Environ. Sci. Technol.* 46, 3406–3414.
510 <https://doi.org/10.1021/es203518d>

511 Ashour, R.M., Samouhos, M., Legaria, E.P., Svä, M., Hö, J., Forsberg, K., Palmlö, M., Kessler,
512 V.G., Seisenbaeva, G.A., Rasmuson, Å.C., 2018. DTPA-Functionalized Silica Nano-and
513 Microparticles for Adsorption and Chromatographic Separation of Rare Earth Elements.
514 <https://doi.org/10.1021/acssuschemeng.8b00725>

515 Ayora, C., Macías, F., Torres, E., Lozano, A., Carrero, S., Nieto, J.M., Pérez-López, R.,
516 Fernández-Martínez, A., Castillo-Michel, H., 2016. Recovery of Rare Earth Elements and
517 Yttrium from Passive-Remediation Systems of Acid Mine Drainage. *Environ. Sci. Technol.*
518 50, 8255–8262. <https://doi.org/10.1021/acs.est.6b02084>

519 Bau, M., 1999. Scavenging of dissolved yttrium and rare earths by precipitating iron
520 oxyhydroxide: Experimental evidence for Ce oxidation, Y-Ho fractionation, and lanthanide
521 tetrad effect. *Geochim. Cosmochim. Acta* 63, 67–77. [https://doi.org/10.1016/S0016-7037\(99\)00014-9](https://doi.org/10.1016/S0016-7037(99)00014-9)

523 Benndorf, P., Jenter, J., Zielke, L., Roesky, P.W., 2011. Chiral lutetium benzamidinate
524 complexes. *Chem. Commun.* 47, 2574–2576. <https://doi.org/10.1039/c0cc04962j>

525 Binnemans, K., Jones, P.T., Blanpain, B., Van Gerven, T., Yang, Y., Walton, A., Buchert, M.,
526 2013. Recycling of rare earths: a critical review. *J. Clean. Prod.* 51, 1–22.

527 <https://doi.org/10.1016/j.jclepro.2012.12.037>

528 Bohr, J., Gibbs, D., Huang, K., 1990. X-ray-diffraction studies of the magnetic state of thulium.
529 Phys. Rev. B 42, 4322–4328. <https://doi.org/10.1103/PhysRevB.42.4322>

530 Bourbos, E., Giannopoulou, I., Karantonis, A., Panias, D., Paspaliaris, I., 2014. Electrodeposition
531 of Rare Earth metals in ionic liquids, in: ERES2014: 1st European Rare Earth Resources
532 Conference. Milos, p. 7.

533 Bourg, A.C.M., 1995. Speciation of heavy metals in soils and groundwater and implications for
534 their natural and provoked mobility. Heavy Met. Probl. Solut. 19–31.

535 Brioschi, L., Steinmann, M., Lucot, E., Pierret, M.C., Stille, P., Prunier, J., Badot, P.M., 2013.
536 Transfer of rare earth elements (REE) from natural soil to plant systems: Implications for
537 the environmental availability of anthropogenic REE. Plant Soil 366, 143–163.
538 <https://doi.org/10.1007/s11104-012-1407-0>

539 Britannica, n.d. Rare-earth element - Electronic structure and ionic radius | Britannica [WWW
540 Document]. URL [https://www.britannica.com/science/rare-earth-element/Electronic-](https://www.britannica.com/science/rare-earth-element/Electronic-structure-and-ionic-radius)
541 [structure-and-ionic-radius](https://www.britannica.com/science/rare-earth-element/Electronic-structure-and-ionic-radius) (accessed 3.26.20).

542 Brown, P.H., Rathjen, A.H., Graham, R.D., Tribe, D.E., 1990. Rare earth elements in biological
543 systems, in: Gschneider, K.A., Eyring, L. (Eds.), Physics and Chemistry of Rare Earths.
544 Elsevier, Amsterdam, pp. 423–450.

545 Bünzli, J.C.G., 2010. Lanthanide luminescence for biomedical analyses and imaging. Chem.
546 Rev. 110, 2729–2755. <https://doi.org/10.1021/cr900362e>

547 Comeselle, C., Chirakkara, R.A., Reddy, K.R., 2013. Electrokinetic-enhanced phytoremediation

548 of soils: status and opportunities. *Chemosphere* 93, 626–36.
549 <https://doi.org/10.1016/j.chemosphere.2013.06.029>

550 Censi, P., Zuddas, P., Randazzo, L.A., Saiano, F., Mazzola, S., Aricò, P., Cuttitta, A., Punturo,
551 R., 2010. Influence of dissolved organic matter on rare earth elements and yttrium
552 distributions in coastal waters. *Chem. Ecol.* 26, 123–135.
553 <https://doi.org/10.1080/02757541003627720>

554 Chakhmouradian, A.R., Zaitsev, A.N., 2012. Rare earth mineralization in igneous rocks: Sources
555 and processes. *Elements* 8, 347–353. <https://doi.org/10.2113/gselements.8.5.347>

556 Chao, Y., Liu, W., Chen, Y., Chen, W., Zhao, L., Ding, Q., Wang, S., Tang, Y.T., Zhang, T.,
557 Qiu, R.L., 2016. Structure, Variation, and Co-occurrence of Soil Microbial Communities in
558 Abandoned Sites of a Rare Earth Elements Mine. *Environ. Sci. Technol.* 50, 11481–11490.
559 <https://doi.org/10.1021/acs.est.6b02284>

560 Chen, Z., 2011. Global rare earth resources and scenarios of future rare earth industry. *J. Rare*
561 *Earths* 29, 1–6. [https://doi.org/10.1016/S1002-0721\(10\)60401-2](https://doi.org/10.1016/S1002-0721(10)60401-2)

562 Cheng, Y., Zhang, L., Bian, X., Zuo, H., Dong, H., 2018. Adsorption and mineralization of
563 REE—lanthanum onto bacterial cell surface. *Environ. Sci. Pollut. Res.* 25, 22334–22339.
564 <https://doi.org/10.1007/s11356-017-9691-0>

565 Chour, Z., Laubie, B., Morel, J.L., Tang, Y., Qiu, R., Simonnot, M.O., Muhr, L., 2018. Recovery
566 of rare earth elements from *Dicranopteris dichotoma* by an enhanced ion exchange leaching
567 process. *Chem. Eng. Process. - Process Intensif.* 130, 208–213.
568 <https://doi.org/10.1016/j.cep.2018.06.007>

569 Cotton, S., 2006. Lanthanide and Actinide Chemistry, Lanthanide and Actinide Chemistry.
570 wiley. <https://doi.org/10.1002/0470010088>

571 Council of Ministers, N., 2019. Contaminated Sediments: Review of solutions for protecting
572 aquatic environments. <https://doi.org/10.6027/TN2019-514>

573 Čuček, L., Klemeš, J.J., Kravanja, Z., 2012. A review of footprint analysis tools for monitoring
574 impacts on sustainability. *J. Clean. Prod.* 34, 9–20.
575 <https://doi.org/10.1016/j.jclepro.2012.02.036>

576 Dinali, G.S., Ramos, S.J., de Carvalho, T.S., Carvalho, G.S., de Oliveira, C., Siqueira, J.O.,
577 Guilherme, L.R.G., 2019. Dissolution techniques for determination of rare earth elements in
578 phosphate products: Acid digestion or alkaline fusion? *J. Geochemical Explor.* 197, 114–
579 121. <https://doi.org/10.1016/j.gexplo.2018.11.016>

580 Dodson, J.R., Hunt, A.J., Parker, H.L., Yang, Y., Clark, J.H., 2012. Elemental sustainability:
581 Towards the total recovery of scarce metals. *Chem. Eng. Process. Process Intensif.* 51, 69–
582 78. <https://doi.org/10.1016/j.cep.2011.09.008>

583 Dolenc, T., Serafimovski, T., Tasev, G., Dobnikar, M., Dolenc, M., Rogan, N., 2007. Major
584 and trace elements in paddy soil contaminated by Pb-Zn mining: A case study of Kočani
585 Field, Macedonia. *Environ. Geochem. Health* 29, 21–32. [https://doi.org/10.1007/s10653-](https://doi.org/10.1007/s10653-006-9057-x)
586 [006-9057-x](https://doi.org/10.1007/s10653-006-9057-x)

587 Dresel, P.E., Wellman, D.M., Cantrell, K.J., Truex, M.J., 2011. Review: Technical and policy
588 challenges in deep vadose zone remediation of metals and radionuclides. *Environ. Sci.*
589 *Technol.* 45, 4207–16. <https://doi.org/10.1021/es101211t>

590 Du, X., Graedel, T.E., 2013. Uncovering the end uses of the rare earth elements. *Sci. Total*
591 *Environ.* 461–462, 781–784. <https://doi.org/10.1016/j.scitotenv.2013.02.099>

592 Ebendorff-Heidepriem, H., Foo, T.C., Li, Y., Oermann, M.R., Monro, T.M., 2008. New tellurite
593 glasses for erbium fibre lasers, in: 2008 Joint Conference of the Opto-Electronics and
594 Communications Conference and the Australian Conference On Optical Fibre Technology,
595 OECC/ACOFT 2008. <https://doi.org/10.1109/OECCACOFT.2008.4610515>

596 Edahbi, M., Plante, B., Benzaazoua, M., Ward, M., Pelletier, M., 2018. Mobility of rare earth
597 elements in mine drainage: Influence of iron oxides, carbonates, and phosphates.
598 *Chemosphere* 199, 647–654. <https://doi.org/10.1016/j.chemosphere.2018.02.054>

599 El-Hagar, S.M., 2007. Sustainable Industrial Design and Waste Management: Cradle-to-Cradle
600 for ... - Salah El Haggag - Google Books. Elsevier, London.

601 Elkina, V., Kurushkin, M., 2020. Promethium: To Strive, to Seek, to Find and Not to Yield.
602 *Front. Chem.* <https://doi.org/10.3389/fchem.2020.00588>

603 Fayiga, A.O., Nwoke, O.C., 2016. Phosphate rock: Origin, importance, environmental impacts,
604 and future roles. *Environ. Rev.* <https://doi.org/10.1139/er-2016-0003>

605 Fdez-Ortiz de Vallejuelo, S., Gredilla, A., da Boit, K., Teixeira, E.C., Sampaio, C.H., Madariaga,
606 J.M., Silva, L.F.O., 2017. Nanominerals and potentially hazardous elements from coal
607 cleaning rejects of abandoned mines: Environmental impact and risk assessment.
608 *Chemosphere* 169, 725–733. <https://doi.org/10.1016/j.chemosphere.2016.09.125>

609 Fedotov, P.S., Rogova, O.B., Dzhendloda, R.K., Karandashev, V.K., 2019. Metal-organic
610 complexes as a major sink for rare earth elements in soils. *Environ. Chem.*

611 <https://doi.org/10.1071/EN18275>

612 Fischer, C., 2002. Oberflächenquantifizierung an Schwarzpeliten unterschiedlicher
613 Verwitterungsgrade. University of Jena.

614 Fray, D.J., 2000. Separating rare earth elements. *Science* (80-.).
615 <https://doi.org/10.1126/science.289.5488.2295>

616 Freslon, N., Bayon, G., Toucanne, S., Bermell, S., Bollinger, C., Chéron, S., Etoubleau, J.,
617 Germain, Y., Khripounoff, A., Ponzevera, E., Rouget, M.L., 2014. Rare earth elements and
618 neodymium isotopes in sedimentary organic matter. *Geochim. Cosmochim. Acta* 140, 177–
619 198. <https://doi.org/10.1016/j.gca.2014.05.016>

620 Gambogi, J., 2016. USGS 2015 Minerals Yearbook - Rare Earths.

621 Giffaut, E., Grivé, M., Blanc, P., Vieillard, P., Colàs, E., Gailhanou, H., Gaboreau, S., Marty, N.,
622 Madé, B., Duro, L., 2014. Andra thermodynamic database for performance assessment:
623 ThermoChimie. *Appl. Geochemistry* 49, 225–236.
624 <https://doi.org/10.1016/j.apgeochem.2014.05.007>

625 Goonan, T.G., 2011. Rare Earth Elements-End Use and Recyclability USGS Scientific
626 Investigations Report 2011-5094.

627 Greenwood, N.N., Earnshaw, A., 2006. 01 - Chemistry of the Elements, Second. ed, Elsevier.
628 Pergamon Press.

629 Gueroult, R., Rax, J.M., Fisch, N.J., 2018. Opportunities for plasma separation techniques in rare
630 earth elements recycling. *J. Clean. Prod.* 182, 1060–1069.
631 <https://doi.org/10.1016/j.jclepro.2018.02.066>

632 Guo, F.Q., Wang, Y.Q., Sun, J.X., Chen, H.M., 1996. Ree bound proteins in natural plant fern
633 *Dicranopteris dichitoma* by MAA. *J. Radioanal. Nucl. Chem.* 209, 91–99.
634 <https://doi.org/10.1007/BF02063534>

635 Guo, W., Fu, R.Y., Zhao, R.X., Zhao, W.J., Guo, J.Y., Zhang, J., 2013. Distribution
636 characteristic and current situation of soil rare earth contamination in the Bayan Obo mining
637 area and Baotou tailing reservoir in Inner Mongolia. *Huanjing Kexue/Environmental Sci.*
638 34, 1895–1900.

639 Gwenzi, W., Mangori, L., Danha, C., Chaukura, N., Dunjana, N., Sanganyado, E., 2018.
640 Sources, behaviour, and environmental and human health risks of high-technology rare
641 earth elements as emerging contaminants. *Sci. Total Environ.*
642 <https://doi.org/10.1016/j.scitotenv.2018.04.235>

643 Haque, N., Hughes, A., Lim, S., Vernon, C., 2014. Rare earth elements: Overview of mining,
644 mineralogy, uses, sustainability and environmental impact. *Resources.*
645 <https://doi.org/10.3390/resources3040614>

646 Hasegawa, Y., Kitagawa, Y., 2019. Thermo-sensitive luminescence of lanthanide complexes,
647 clusters, coordination polymers and metal-organic frameworks with organic
648 photosensitizers. *J. Mater. Chem. C.* <https://doi.org/10.1039/c9tc00607a>

649 He, H., Fan, C., Peng, Q., Wu, M., Zheng, J., Wu, G.-L., 2019. Bioaccumulation and
650 translocation of rare earth elements in two forage legumes grown in soils treated with coal
651 fly ash. *Chem. Geol.* 528, 119284. <https://doi.org/10.1016/j.chemgeo.2019.119284>

652 Hemmilä, I., Dakubu, S., Mukkala, V.M., Siitari, H., Lövgren, T., 1984. Europium as a label in
653 time-resolved immunofluorometric assays. *Anal. Biochem.* 137, 335–343.

654 [https://doi.org/10.1016/0003-2697\(84\)90095-2](https://doi.org/10.1016/0003-2697(84)90095-2)

655 Henderson, P., 1984. Rare earth element geochemistry. Elsevier.

656 Hirano, S., Suzuki, K.T., 1996. Exposure, metabolism, and toxicity of rare earths and related
657 compounds. *Environ. Health Perspect.* 104, 85–95. <https://doi.org/10.1289/ehp.96104s185>

658 Hogland, M., Āriņa, D., Kriipsalu, M., Jani, Y., Kaczala, F., de Sá Salomão, A.L., Orupõld, K.,
659 Pehme, K.M., Rudoviča, V., Denafas, G., Burlakovs, J., Vincēviča-Gaile, Z., Hogland, W.,
660 2018. Remarks on four novel landfill mining case studies in Estonia and Sweden. *J. Mater.*
661 *Cycles Waste Manag.* 20, 1355–1363. <https://doi.org/10.1007/s10163-017-0683-4>

662 Hu, Z., Haneklaus, S., Sparovek, G., Schnug, E., 2006. Rare Earth Elements in Soils. *Commun.*
663 *Soil Sci. Plant Anal.* 37, 1381–1420. <https://doi.org/10.1080/00103620600628680>

664 Izatt, S.R., Izatt, N.E., Bruening, R.L., 2010. Metal separations of interest to the Chinese
665 metallurgical industry. *J. Rare Earths* 28, 22–29. <https://doi.org/10.1016/S1002->
666 [0721\(10\)60370-5](https://doi.org/10.1016/S1002-0721(10)60370-5)

667 Jalali, J., Gaudin, P., Capiiaux, H., Ammar, E., Lebeau, T., 2019. Fate and transport of metal trace
668 elements from phosphogypsum piles in Tunisia and their impact on soil bacteria and wild
669 plants. *Ecotoxicol. Environ. Saf.* 174, 12–25. <https://doi.org/10.1016/j.ecoenv.2019.02.051>

670 Jérôme, D.Y., Philippot, J.C., 1973. Chemical composition of Luna 20 soil and rock fragments.
671 *Geochim. Cosmochim. Acta* 37, 909–914. [https://doi.org/10.1016/0016-7037\(73\)90188-9](https://doi.org/10.1016/0016-7037(73)90188-9)

672 Jin, S., Hu, Z., Huang, Y., Hu, Y., Pan, H., 2019. Evaluation of several phosphate amendments
673 on rare earth element concentrations in rice plant and soil solution by X-ray diffraction.
674 *Chemosphere* 236, 124322. <https://doi.org/10.1016/j.chemosphere.2019.07.053>

675 Johannesson, K.H., Lyons, W.B., Yelken, M.A., Gaudette, H.E., Stetzenbach, K.J., 1996.
676 Geochemistry of the rare-earth elements in hypersaline and dilute acidic natural terrestrial
677 waters: Complexation behavior and middle rare-earth element enrichments. *Chem. Geol.*
678 133, 125–144. [https://doi.org/10.1016/S0009-2541\(96\)00072-1](https://doi.org/10.1016/S0009-2541(96)00072-1)

679 Jowitt, S.M., Werner, T.T., Weng, Z., Mudd, G.M., 2018. Recycling of the rare earth elements.
680 *Curr. Opin. Green Sustain. Chem.* <https://doi.org/10.1016/j.cogsc.2018.02.008>

681 Kanazawa, Y., Kamitani, M., 2006. Rare earth minerals and resources in the world, in: *Journal of*
682 *Alloys and Compounds*. pp. 1339–1343. <https://doi.org/10.1016/j.jallcom.2005.04.033>

683 Karshigina, Z., Abisheva, Z., Bochevskaya, Y., Akcil, A., Sargelova, E., Sukurov, B., Silachyov,
684 I., 2018. Recovery of rare earth metals (REMs) from primary raw material: sulphatization-
685 leaching-precipitation-extraction. *Miner. Process. Extr. Metall. Rev.* 39, 319–338.
686 <https://doi.org/10.1080/08827508.2018.1434778>

687 Korkmaz, K., Alemrajabi, M., Rasmuson, Å.C., Forsberg, K.M., 2020. Separation of valuable
688 elements from NiMH battery leach liquor via antisolvent precipitation. *Sep. Purif. Technol.*
689 234, 115812. <https://doi.org/10.1016/j.seppur.2019.115812>

690 Kraemer, D., Kopf, S., Bau, M., 2015. Oxidative mobilization of cerium and uranium and
691 enhanced release of “immobile” high field strength elements from igneous rocks in the
692 presence of the biogenic siderophore desferrioxamine B. *Geochim. Cosmochim. Acta* 165,
693 263–279. <https://doi.org/10.1016/j.gca.2015.05.046>

694 Krishnamurthy, N. (Nagaiyar), Gupta, C.K., 2015. *Extractive metallurgy of rare earths*, Second.
695 ed. CRC Press, Boca Raton.

696 Kronholm, B., Anderson, C.G., Taylor, P.R., 2013. A primer on hydrometallurgical rare earth
697 separations. *JOM* 65, 1321–1326. <https://doi.org/10.1007/s11837-013-0718-9>

698 Krook, J., Baas, L., 2013. Getting serious about mining the technosphere: A review of recent
699 landfill mining and urban mining research. *J. Clean. Prod.* 55, 1–9.
700 <https://doi.org/10.1016/j.jclepro.2013.04.043>

701 Kubiak, J.J., Khankhane, P.J., Kleingeld, P.J., Lima, A.T., 2012. An attempt to electrically
702 enhance phytoremediation of arsenic contaminated water. *Chemosphere* 87, 259–264.
703 <https://doi.org/10.1016/j.chemosphere.2011.12.048>

704 Labrecque, C., Larivière, D., 2014. Quantification of rare earth elements using cloud point
705 extraction with diglycolamide and ICP-MS for environmental analysis. *Anal. Methods* 6,
706 9291–9298. <https://doi.org/10.1039/c4ay01911c>

707 Labrecque, C., Lebed, P.J., Larivière, D., 2016. Isotopic signature of selected lanthanides for
708 nuclear activities profiling using cloud point extraction and ICP-MS/MS. *J. Environ.*
709 *Radioact.* 155–156, 15–22. <https://doi.org/10.1016/j.jenvrad.2016.02.002>

710 Lar, U.A., Gusikit, R.B., 2015. Environmental and health impact of potentially harmful elements
711 distribution in the Panyam (Sura) volcanic province, Jos Plateau, Central Nigeria. *Environ.*
712 *Earth Sci.* 74, 1699–1710. <https://doi.org/10.1007/s12665-015-4178-0>

713 Li, J., Hong, M., Yin, X., Liu, J., 2010. Effects of the accumulation of the rare earth elements on
714 soil macrofauna community. *J. Rare Earths* 28, 957–964. [https://doi.org/10.1016/S1002-](https://doi.org/10.1016/S1002-0721(09)60233-7)
715 [0721\(09\)60233-7](https://doi.org/10.1016/S1002-0721(09)60233-7)

716 Li, Z., Zhou, D., Su, J., Luo, X., 2017. Leaching Remediation of Lanthanum, Cerium and

717 Yttrium from Contaminated Soil by Saponin. *Zhongguo Xitu Xuebao/Journal Chinese Rare*
718 *Earth Soc.* 35, 672–679. <https://doi.org/10.11785/S1000-4343.20170514>

719 Lima, A.T., Hofmann, A., Reynolds, D., Ptacek, C.J.J., Van Cappellen, P., Ottosen, L.M.M.,
720 Pamukcu, S., Alshawabekh, A., O’Carroll, D.M.M., Riis, C., Cox, E., Gent, D.B.B., Landis,
721 R., Wang, J., Chowdhury, A.I.A.I.A., Secord, E.L.L., Sanchez-Hachair, A., 2017.
722 *Environmental Electrokinetics for a sustainable subsurface. Chemosphere* 181, 122–133.
723 <https://doi.org/10.1016/j.chemosphere.2017.03.143>

724 Liu, C., Yuan, M., Liu, W.-S., Guo, M.-N., Huot, H., Tang, Y.-T., Laubie, B., Simonnot, M.-O.,
725 Morel, J.L., Qiu, R.-L., 2018. Element Case Studies: Rare Earth Elements, in: Ent, A. van
726 der, Echevarria, G., Baker, A.J.M., Morel, J.L. (Eds.), *Agromining: Farming for Metals -*
727 *Extracting Unconventional Resources Using Plants.* Springer, Nancy, pp. 297–308.
728 https://doi.org/10.1007/978-3-319-61899-9_19

729 Liu, J., Liu, W., Zhang, Y., Chen, C., Wu, W., Zhang, T.C., 2021. Microbial communities in rare
730 earth mining soil after in-situ leaching mining. *Sci. Total Environ.* 755.
731 <https://doi.org/10.1016/j.scitotenv.2020.142521>

732 Liu, W.S., Guo, M.N., Liu, C., Yuan, M., Chen, X.T., Huot, H., Zhao, C.M., Tang, Y.T., Morel,
733 J.L., Qiu, R.L., 2019. Water, sediment and agricultural soil contamination from an ion-
734 adsorption rare earth mining area. *Chemosphere* 75–83.
735 <https://doi.org/10.1016/j.chemosphere.2018.10.109>

736 Lozano, A., Ayora, C., Fernández-Martínez, A., 2019. Sorption of rare earth elements onto
737 basaluminite: The role of sulfate and pH. *Geochim. Cosmochim. Acta* 258, 50–62.
738 <https://doi.org/10.1016/j.gca.2019.05.016>

739 Lozano, A., Ayora, C., Macías, F., León, R., Gimeno, M.J., Auqué, L., 2020. Geochemical
740 behavior of rare earth elements in acid drainages: Modeling achievements and limitations. *J.*
741 *Geochemical Explor.* 216, 106577. <https://doi.org/10.1016/j.gexplo.2020.106577>

742 Luo, Y.R., Byrne, R.H., 2004. Carbonate complexation of yttrium and the rare earth elements in
743 natural waters. *Geochim. Cosmochim. Acta* 68, 691–699. [https://doi.org/10.1016/S0016-](https://doi.org/10.1016/S0016-7037(03)00495-2)
744 [7037\(03\)00495-2](https://doi.org/10.1016/S0016-7037(03)00495-2)

745 Maes, S., Zhuang, W.Q., Rabaey, K., Alvarez-Cohen, L., Hennebel, T., 2017. Concomitant
746 Leaching and Electrochemical Extraction of Rare Earth Elements from Monazite. *Environ.*
747 *Sci. Technol.* 51, 1654–1661. <https://doi.org/10.1021/acs.est.6b03675>

748 Markard, J., Raven, R., Truffer, B., 2012. Sustainability transitions: An emerging field of
749 research and its prospects. *Res. Policy* 41, 955–967.
750 <https://doi.org/10.1016/j.respol.2012.02.013>

751 Marques, R., Prudêncio, M.I., Waerenborgh, J.C., Rocha, F., Ferreira da Silva, E., Dias, M.I.,
752 Madeira, J., Vieira, B.J.C., Marques, J.G., 2016. Geochemical fingerprints in topsoils of the
753 volcanic Brava Island, Cape Verde. *Catena* 147, 522–535.
754 <https://doi.org/10.1016/j.catena.2016.08.008>

755 Marques, R., Prudêncio, M.I., Waerenborgh, J.C., Vieira, B.J.C., Rocha, F., Dias, M.I., Madeira,
756 J., Mata, J., 2017. Extrusive carbonatite outcrops – A source of chemical elements
757 imbalance in topsoils of oceanic volcanic islands. *Catena* 157, 333–343.
758 <https://doi.org/10.1016/j.catena.2017.05.035>

759 Marsac, R., Davranche, M., Gruau, G., Dia, A., Bouhnik-Le Coz, M., 2012. Aluminium
760 competitive effect on rare earth elements binding to humic acid. *Geochim. Cosmochim.*

761 Acta 89, 1–9. <https://doi.org/10.1016/j.gca.2012.04.028>

762 Massari, S., Ruberti, M., 2013. Rare earth elements as critical raw materials: Focus on
763 international markets and future strategies. *Resour. Policy* 38, 36–43.
764 <https://doi.org/10.1016/j.resourpol.2012.07.001>

765 McCoy, H.N., 1941. Europium and Ytterbium Amalgams. *J Am Chem Soc* 63, 1622–1624.

766 Merten, D., Geletneky, J., Bergmann, H., Haferburg, G., Kothe, E., Büchel, G., 2005. Rare earth
767 element patterns: A tool for understanding processes in remediation of acid mine drainage.
768 *Chemie der Erde* 65, 97–114. <https://doi.org/10.1016/j.chemer.2005.06.002>

769 Mleczek, P., Borowiak, K., Budka, A., Niedzielski, P., 2018. Relationship between concentration
770 of rare earth elements in soil and their distribution in plants growing near a frequented road.
771 *Environ. Sci. Pollut. Res.* 25, 23695–23711. <https://doi.org/10.1007/s11356-018-2428-x>

772 Moreira, C.G., Carvalho, T.S. de, de Oliveira, C., Abreu, L.B. de, Castro, A.C.S. de, Ribeiro,
773 P.G., Bispo, F.H.A., Boutin, C., Guilherme, L.R.G., 2019. Ecological risk assessment of
774 cerium for tropical agroecosystems. *Chemosphere* 124–131.
775 <https://doi.org/10.1016/j.chemosphere.2018.12.195>

776 Ottosen, L.M., Christensen, I. V., Rørig-Dalgård, I., Jensen, P.E., Hansen, H.K., 2008.
777 Utilization of electromigration in civil and environmental engineering—Processes, transport
778 rates and matrix changes. *J. Environ. Sci. Heal. Part A* 43, 795–809.
779 <https://doi.org/10.1080/10934520801973949>

780 Ottosen, L.M., Hansen, H.K., Laursen, S., Villumsen, A., 1997. Electrodialytic remediation of
781 soil polluted with copper from wood preservation industry. *Environ. Sci. Technol.* 31,

782 1711–1715.

783 Ouslimani, H., Bastard, L., Broquin, J.-E., 2013. Q-switched distributed-Bragg-reflector
784 ytterbium laser, in: *Integrated Optics: Devices, Materials, and Technologies XVII*. SPIE, p.
785 86270F. <https://doi.org/10.1117/12.2004361>

786 Pires, C.M.G., Ponte, H. de A., Pereira, J.T., Ponte, M.J.J. de S., 2019. Yttrium extraction from
787 soils by electric field assisted mining applying the evolutionary operation technique. *J.*
788 *Clean. Prod.* 227, 272–279. <https://doi.org/10.1016/j.jclepro.2019.04.077>

789 Prudêncio, M.I., Valente, T., Marques, R., Sequeira Braga, M.A., Pamplona, J., 2015.
790 Geochemistry of rare earth elements in a passive treatment system built for acid mine
791 drainage remediation. *Chemosphere* 138, 691–700.
792 <https://doi.org/10.1016/j.chemosphere.2015.07.064>

793 Qin, B., Liu, W., He, E., Li, Y., Liu, C., Ruan, J., Qiu, R., Tang, Y., 2019. Vacuum pyrolysis
794 method for reclamation of rare earth elements from hyperaccumulator *Dicranopteris*
795 *dichotoma* grown in contaminated soil. *J. Clean. Prod.* 229, 480–488.
796 <https://doi.org/10.1016/j.jclepro.2019.05.031>

797 Rabung, T., Stumpf, T., Geckeis, H., Klenze, R., Kim, J.I., 2000. Sorption of AM(III) and Eu(III)
798 onto γ -alumina: Experiment and modelling, in: *Radiochimica Acta*. R. Oldenbourg Verlag
799 GmbH, pp. 711–716. <https://doi.org/10.1524/ract.2000.88.9-11.711>

800 Ramos, S.J., Dinali, G.S., Oliveira, C., Martins, G.C., Moreira, C.G., Siqueira, J.O., Guilherme,
801 L.R.G., 2016. Rare Earth Elements in the Soil Environment. *Curr. Pollut. Reports*.
802 <https://doi.org/10.1007/s40726-016-0026-4>

803 Riedel, T., Hennessy, P., Iden, S.C., Koschinsky, A., 2015. Leaching of soil-derived major and
804 trace elements in an arable topsoil after the addition of biochar. *Eur. J. Soil Sci.* 66, 823–
805 834. <https://doi.org/10.1111/ejss.12256>

806 Rim, K.T., Koo, K.H., Park, J.S., 2013. Toxicological evaluations of rare earths and their health
807 impacts to workers: A literature review. *Saf. Health Work.*
808 <https://doi.org/10.5491/SHAW.2013.4.1.12>

809 Sadeghi, M., Petrosino, P., Ladenberger, A., Albanese, S., Andersson, M., Morris, G., Lima, A.,
810 De Vivo, B., 2013. Ce, La and Y concentrations in agricultural and grazing-land soils of
811 Europe. *J. Geochemical Explor.* 133, 202–213. <https://doi.org/10.1016/j.gexplo.2012.12.007>

812 Sahoo, S.K., Yonehara, H., Kurotaki, K., Shiraishi, K., Ramzaev, V., Barkovski, A., 2001.
813 Determination of rare earth elements, thorium and uranium by inductively coupled plasma
814 mass spectrometry and strontium isotopes by thermal ionization mass spectrometry in soil
815 samples of Bryansk region contaminated due to Chernobyl accident. *J. Radioanal. Nucl.*
816 *Chem.* 247, 341–345. <https://doi.org/10.1023/A:1006757718985>

817 Sako, A., Semdé, S., Wenmenga, U., 2018. Geochemical evaluation of soil, surface water and
818 groundwater around the Tongon gold mining area, northern Côte d'Ivoire, West Africa. *J.*
819 *African Earth Sci.* 145, 297–316. <https://doi.org/10.1016/j.jafrearsci.2018.05.016>

820 Scaff, W.L., Dyer, D.L., Mori, K., 1969. Fluorescent europium chelate sta. *J. Bacteriol.* 98, 246–
821 248.

822 Scarazzato, T., Panossian, Z., Tenório, J.A.S., Pérez-Herranz, V., Espinosa, D.C.R., 2017. A
823 review of cleaner production in electroplating industries using electrodialysis. *J. Clean.*
824 *Prod.* 168, 1590–1602. <https://doi.org/10.1016/j.jclepro.2017.03.152>

825 Schijf, J., Byrne, R.H., 2004. Determination of SO₄²⁻ for yttrium and the rare earth elements at I
826 = 0.66 m and t = 25°C-implications for YREE solution speciation in sulfate-rich waters.
827 *Geochim. Cosmochim. Acta* 68, 2825–2837. <https://doi.org/10.1016/j.gca.2003.12.003>

828 Secord, E.L., Kottara, A., Cappellen, P. V., Lima, A.T., 2016. Inoculating Bacteria into
829 Polycyclic Aromatic Hydrocarbon-Contaminated Oil Sands Soil by Means of
830 Electrokinetics. *Water, Air, Soil Pollut.* <https://doi.org/10.1007/s11270-016-2991-z>

831 Seredin, V. V., 2010. A new method for primary evaluation of the outlook for rare earth element
832 ores. *Geol. Ore Depos.* 52, 428–433. <https://doi.org/10.1134/S1075701510050077>

833 Sholl, D.S., Lively, R.P., 2016. Seven chemical separations to change the world. *Nature*.
834 <https://doi.org/10.1038/532435a>

835 Smith, A.B., 2001. Fast-neutrons incident on holmium. *Ann. Nucl. Energy* 28, 1745–1758.
836 [https://doi.org/10.1016/S0306-4549\(01\)00015-9](https://doi.org/10.1016/S0306-4549(01)00015-9)

837 Su, C., 2017. Environmental implications and applications of engineered nanoscale magnetite
838 and its hybrid nanocomposites: A review of recent literature. *J. Hazard. Mater.* 322, 48–84.
839 <https://doi.org/10.1016/j.jhazmat.2016.06.060>

840 Swain, N., Mishra, S., 2019. A review on the recovery and separation of rare earths and
841 transition metals from secondary resources. *J. Clean. Prod.*
842 <https://doi.org/10.1016/j.jclepro.2019.02.094>

843 Tang, H., Shuai, W., Wang, X., Liu, Y., 2017. Extraction of rare earth elements from a
844 contaminated cropland soil using nitric acid, citric acid, and EDTA. *Environ. Technol.*
845 (United Kingdom) 38, 1980–1986. <https://doi.org/10.1080/09593330.2016.1244563>

846 Taylor, S.R., 1964. Abundance of chemical elements in the continental crust: a new table.
847 *Geochim. Cosmochim. Acta* 28, 1273–1285. [https://doi.org/10.1016/0016-7037\(64\)90129-2](https://doi.org/10.1016/0016-7037(64)90129-2)

848 Tromp, K., Lima, A.T., Barendregt, A., Verhoeven, J.T.A., 2012. Retention of heavy metals and
849 poly-aromatic hydrocarbons from road water in a constructed wetland and the effect of de-
850 icing. *J. Hazard. Mater.* 203–204, 290–298. <https://doi.org/10.1016/j.jhazmat.2011.12.024>

851 Uda, T., Jacob, K.T., Hirasawa, M., 2000. Technique for enhanced rare earth separation. *Science*
852 (80-.). 289, 2326–2329. <https://doi.org/10.1126/science.289.5488.2326>

853 USEPA, 2012. Rare earth elements : a review of production , processing , recycling , and
854 associated environmental issues. <https://doi.org/EPA/600/R-12/572>

855 USEPA, 2006. In-Situ Chemical Oxidation. Cincinnati, OH.

856 USGS, 2020. USGS Mineral Commodity Summaries 2020. Reston, Virginia.
857 <https://doi.org/https://doi.org/10.3133/mcs2020>

858 Voncken, J.H.L., 2016. The Rare Earth Elements An Introduction. SpringerBriefs in Earth
859 Science, Delft. <https://doi.org/10.1007/978-3-319-26809-5>

860 Wang, W., Pranolo, Y., Cheng, C.Y., 2011. Metallurgical processes for scandium recovery from
861 various resources: A review. *Hydrometallurgy* 108, 100–108.
862 <https://doi.org/10.1016/j.hydromet.2011.03.001>

863 Wood, S.A., 1990. The aqueous geochemistry of the rare-earth elements and yttrium. 1. Review
864 of available low-temperature data for inorganic complexes and the inorganic REE
865 speciation of natural waters. *Chem. Geol.* 82, 159–186. [https://doi.org/10.1016/0009-](https://doi.org/10.1016/0009-2541(90)90080-Q)
866 [2541\(90\)90080-Q](https://doi.org/10.1016/0009-2541(90)90080-Q)

867 Wu, J., Chen, A., Peng, S., Wei, Z., Liu, G., 2013. Identification and application of amino acids
868 as chelators in phytoremediation of rare earth elements lanthanum and yttrium. *Plant Soil*
869 373, 329–338. <https://doi.org/10.1007/s11104-013-1811-0>

870 Wu, P., Cui, P., Alves, M.E., Peijnenburg, W.J.G.M., Liu, C., Zhou, D., Wang, H., Ok, Y.S.,
871 Wang, Y., 2019. Interactive effects of rice straw biochar and Γ -Al₂O₃ on immobilization
872 of Zn. *J. Hazard. Mater.* 250–257. <https://doi.org/10.1016/j.jhazmat.2019.03.076>

873 Wu, Y., Yin, X., Zhang, Q., Wang, W., Mu, X., 2014. The recycling of rare earths from waste
874 tricolor phosphors in fluorescent lamps: A review of processes and technologies. *Resour.*
875 *Conserv. Recycl.* <https://doi.org/10.1016/j.resconrec.2014.04.007>

876 Zaharescu, D.G., Burghilea, C.I., Dontsova, K., Presler, J.K., Maier, R.M., Huxman, T.,
877 Domanik, K.J., Hunt, E.A., Amistadi, M.K., Gaddis, E.E., Palacios-Menendez, M.A.,
878 Vaquera-Ibarra, M.O., Chorover, J., 2017. Ecosystem Composition Controls the Fate of
879 Rare Earth Elements during Incipient Soil Genesis. *Sci. Rep.* 7, 1–15.
880 <https://doi.org/10.1038/srep43208>

881 Zhang, S., Zhang, J., Cheng, X., Mei, Y., Hu, C., Wang, M., Li, J., 2015. Electrokinetic
882 remediation of soil containing Cr(VI) by photovoltaic solar panels and a DC-DC converter.
883 *J. Chem. Technol. Biotechnol.* 90, 693–700. <https://doi.org/10.1002/jctb.4359>

884 Zhang, W., Rezaee, M., Bhagavatula, A., Li, Y., Groppo, J., Honaker, R., 2015. A Review of the
885 Occurrence and Promising Recovery Methods of Rare Earth Elements from Coal and Coal
886 By-Products. *Int. J. Coal Prep. Util.* 35, 295–330.
887 <https://doi.org/10.1080/19392699.2015.1033097>

888 Zhou, D., Li, Z., Luo, X., Su, J., 2017. Leaching of rare earth elements from contaminated soils

889 using saponin and rhamnolipid bio-surfactant. *J. Rare Earths* 35, 911–919.
890 [https://doi.org/10.1016/S1002-0721\(17\)60994-3](https://doi.org/10.1016/S1002-0721(17)60994-3)

891 Zhu, J.G., Zhang, Y.L., Sun, X.M., Yamaski, S., Tsumura, A., 1998. Water-Soluble Rare Earth
892 Elements in Some Top-Soils of China, in: *Chemistry for the Protection of the Environment*
893 3. Springer US, Boston, MA, pp. 313–319. https://doi.org/10.1007/978-1-4757-9664-3_33

894