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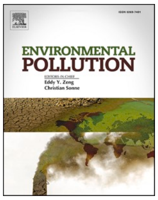

From mine to mind and mobiles – Lithium contamination and its risk management[☆]

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ABSTRACT

With the ever-increasing demand for lithium (Li) for portable energy storage devices, there is a global concern associated with environmental contamination of Li, via the production, use, and disposal of Li-containing products, including mobile phones and mood-stabilizing drugs. While geogenic Li is sparingly soluble, Li added to soil is one of the most mobile cations in soil, which can leach to groundwater and reach surface water through runoff. Lithium is readily taken up by plants and has relatively high plant accumulation coefficient, albeit the underlying mechanisms have not been well described. Therefore, soil contamination with Li could reach the food chain due to its mobility in surface- and ground-waters and uptake into plants. High environmental Li levels adversely affect the health of humans, animals, and plants. Lithium toxicity can be considerably managed through various remediation approaches such as immobilization using clay-like amendments and/or chelate-enhanced phytoremediation. This review integrates fundamental aspects of Li distribution and behaviour in terrestrial and aquatic environments in an effort to efficiently remediate Li-contaminated ecosystems. As research to date has not provided a clear picture of how the increased production and disposal of Li-based products adversely impact human and ecosystem health, there is an urgent need for further studies on this field.

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1. Introduction

Humans and animals exposed to increasing environmental lithium (Li) concentrations through the use and improper disposal of Li-containing products has been an emerging concern globally. Global Li production is currently 77,000 tons year^{-1} (USGS, 2020), a three-fold increase since 2000 (Kelly et al., 2010). The growing use of rechargeable Li-ion batteries in electronic products and electric vehicles drives global lithium demand (Mohr et al., 2012). Soil contamination with the degradation products of electronic waste could add Li to soil as Li^+ ions or Li_2O nanoparticles (Avila-Arias et al., 2019; Li and Achal, 2020). Vehicular grease contains 1% Li, which enters the environment through runoff from roads (Zeng and Li, 2014). Similarly, ceramics and mood-stabilizing drugs contain Li, which can enter soil and water from landfill leachate, stormwater, and sewage (Aral and Vecchio-Sadus, 2008; Hao et al., 2017). Lithium is also used in the illicit manufacture of methamphetamine and could enter the environment through inappropriate disposal (Person et al., 2005).

Bibienne et al. (2020), in their article entitled '*From mine to mind and mobiles: Society's increasing dependence on Li*' delineated the journey of Li from mining through to a suite of applications, including safe, clean, and portable energy storage devices and pharmaceuticals. We have deliberately retained part of their article in our review's title to highlight the importance of this emerging, potentially toxic element (PTE) in industrial and medicinal applications and its eventual release to the environment. Highly variable concentrations in soil and water in different agro-ecological regions of the world have led to variations in Li concentrations in plants and food products, posing various levels of risks to human populations (Shahzad et al., 2016, 2017; Sobolev et al., 2019).

Lithium is not one of the essential nutrients for vital functions as its deficiency causes no obvious symptoms in humans. However, low Li intake has been associated with neurotransmission regulation, indicating that low Li levels have beneficial effects for living organisms (Jakobsson et al., 2017). Low Li levels have also been linked to slowing cell growth and impairment of reproductive function and life expectancy in humans (Sobolev et al., 2019). However, at high doses (e.g., 15–20 mg L^{-1} blood concentrations), Li is toxic to humans causing nausea, visual impairment, and kidney problems, or even medical emergencies such as coma and cardiac arrest. Excretion of Li from animals, including humans, occurs predominantly via the kidney; the proximal renal tubule reabsorbs approximately 80% of Li, with 20% excreted in urine. However, apart from a medium-specific concentration of 0.005 mg L^{-1} for Li, recently proposed by the Pennsylvania Department of Environmental Protection, it is largely unregulated in ground or drinking water.

There are few reports on Li in soil, sediments, and aquatic environments (Bradford, 1973; Robinson et al., 2018). However, awareness of Li in the environment has increased recently due to its extensive use in the energy sector (Bibienne et al., 2020). Lithium has been recently drawing attention of the general public and scientific community, and it is considered as an emerging environmental contaminant. Recent review articles have concentrated on the dynamics and mobility of Li in the soil–plant system (Robinson et al., 2018), including toxicity in plants (Shahzad et al., 2016), aquatic systems (Kszos and Stewart, 2003), and humans (Aral and Vecchio-Sadus, 2008). Of these, only two review articles have critically analysed soil remediation with Li in terrestrial and aquatic ecosystems (Shahzad et al., 2017; Kszos and Stewart, 2003). More research is warranted to establish the scientific and technological soundness for remediation of Li-contaminated environments, especially in soils, sediments, and aquatic systems.

The goal of this review is to critically analyse literature on the sources of Li inputs to soil and aquatic environments, potential risks posed by increased emissions of Li into the environment, and strategies to manage these risks. Specifically, we determine the (1) origins of Li contamination in soil and water, (2) likely Li fluxes in the water–soil–-plant continuum, (3) potential effects of elevated Li on human health and wellbeing, and (4) strategies to manage the risks posed by Li-contaminated environments with a view to their eventual remediation. Literature was analysed through the databases of Google Scholar, Scopus, Web of Science and other web sources using the key words: 'Lithium', 'Lithium Battery', 'Lithium medicine', 'Lithium Characteristics', 'Lithium Dynamics', 'Lithium Bioavailability', 'Lithium toxicity', "Lithium Reactions', 'Lithium Remediation', 'Lithium uptake' and 'Lithium Leaching'. Fig. 1 depicts the article decision-making process and the scope of this review. The review aims to fill knowledge gaps about the increasing emissions of Li to soil and aquatic environments and assist in developing sustainable strategies for managing Li contamination in the environment. The review will outline areas in which more research needs to be conducted to address knowledge gaps effectively. Moreover, increased knowledge about Li dynamics in soil and aquatic environments will benefit the mining industries by changing public perception of Li contamination.

2. Origin and sources of lithium contamination

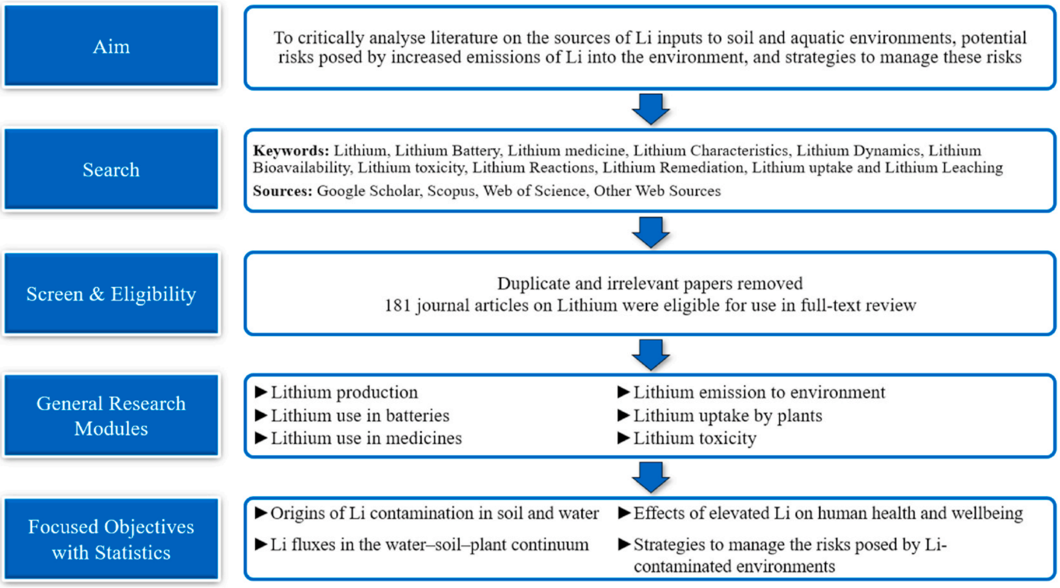
Environmental Li contamination originates mainly from geogenic and anthropogenic origin (Aral and Vecchio-Sadus, 2011), which can be further divided into point and nonpoint sources based on the exact location Li enters the environmental compartments (i.e., air, water, and soil) (Fig. 2). Point sources are when pollution occurs from a defined area, and nonpoint sources are when pollution comes from diffused places. As a reactive element, Li does not occur in its free form in nature but is predominantly associated with mineral components such as apatite or aluminium silicide (Dessemond et al., 2019; Gourcerol et al., 2019; Kesler et al., 2012; Kushnir, 2013), and various salts such as lithium carbonate (Li_2CO_3), lithium chloride (LiCl), and lithium hydroxide (LiOH) (Bleiwas and Coffman, 1986; Speirs et al., 2014). Furthermore, Li in aquatic marine environments can be terrigenous and authigenic (or hydrogenous) in origin (Qin et al., 2015).

2.1. Geogenic origin

Geogenic sources provide baseline or background levels of Li in the environment. Lithium occurs naturally, and it is mainly deposited in rocks, minerals, or mines at different concentrations (Aral and Vecchio-Sadus, 2008, 2011). Lithium is enriched in the Earth's crust, ranging from of 20–60 mg kg^{-1} (0.002–0.006%) of the Earth's crust (Danielik and Fellner, 1998), with 22, 15, 66, 15, and 5 mg kg^{-1} in granite, diabase, shales, sandstones, and carbonates, respectively (Mason and Moore, 1985). In soils, the highest Li concentrations occur in arid and saline soils, where Li and other salts are concentrated through surface evaporation (Merian and Clarkson, 1991).

Natural weathering processes in geological deposits, hot springs arising from geothermal activities, and volcanic eruptions are the major geogenic sources releasing Li into the environment, and play an important role in balancing the global Li cycle (Robinson et al., 2018). Lithium also occurs naturally in trace amounts in freshwater, groundwater, oceans, soil, and the atmosphere and rarely occurs in elevated concentrations in water, soil, or bottom sediments. The average background Li concentration in soils is ~30 mg kg^{-1} (Schrauzer, 2002); however, its concentrations ranged from 0.08 up to 92 mg kg^{-1} in New Zealand soils (Yalamanchali, 2012). Topsoil has lower Li contents than underlying layers (Merian and Clarkson, 1991) and clay fractions have significantly higher Li contents than organic fractions (Schrauzer, 2002). Volcanic fumaroles, thermal springs, material from weathering processes, and water sourcing from rocks rich in Li or Li deposits can have higher Li concentrations than background levels (Aral and Vecchio-Sadus, 2011). For example, in Kuril Island (Russia), Li concentration up to ~927 μg L^{-1} was reported in the waters (rich in sulfate) of the Mendeleev volcano, while it ranged from 264 to 398 μg L^{-1} in the Stolbovskie springs (Aral and Vecchio-Sadus, 2011).

The weathering of rocks (igneous and sedimentary) releases Li into soil (Aral and Vecchio-Sadus, 2008, 2011) and aquatic systems (Millot



To critically analyse literature on the sources of Li inputs to soil and aquatic environments, potential risks posed by increased emissions of Li into the environment, and strategies to manage these risks

Keywords: Lithium, Lithium Battery, Lithium medicine, Lithium Characteristics, Lithium Dynamics, Lithium Bioavailability, Lithium toxicity, Lithium Reactions, Lithium Remediation, Lithium uptake and Lithium Leaching
Sources: Google Scholar, Scopus, Web of Science, Other Web Sources

Duplicate and irrelevant papers removed
181 journal articles on Lithium were eligible for use in full-text review

▶ Lithium production
▶ Lithium use in batteries
▶ Lithium use in medicines
▶ Lithium emission to environment
▶ Lithium uptake by plants
▶ Lithium toxicity

▶ Origins of Li contamination in soil and water
▶ Li fluxes in the water–soil–plant continuum
▶ Effects of elevated Li on human health and wellbeing
▶ Strategies to manage the risks posed by Li-contaminated environments

Fig. 1. Flowchart of the journal article decision-making process and scope of the review.

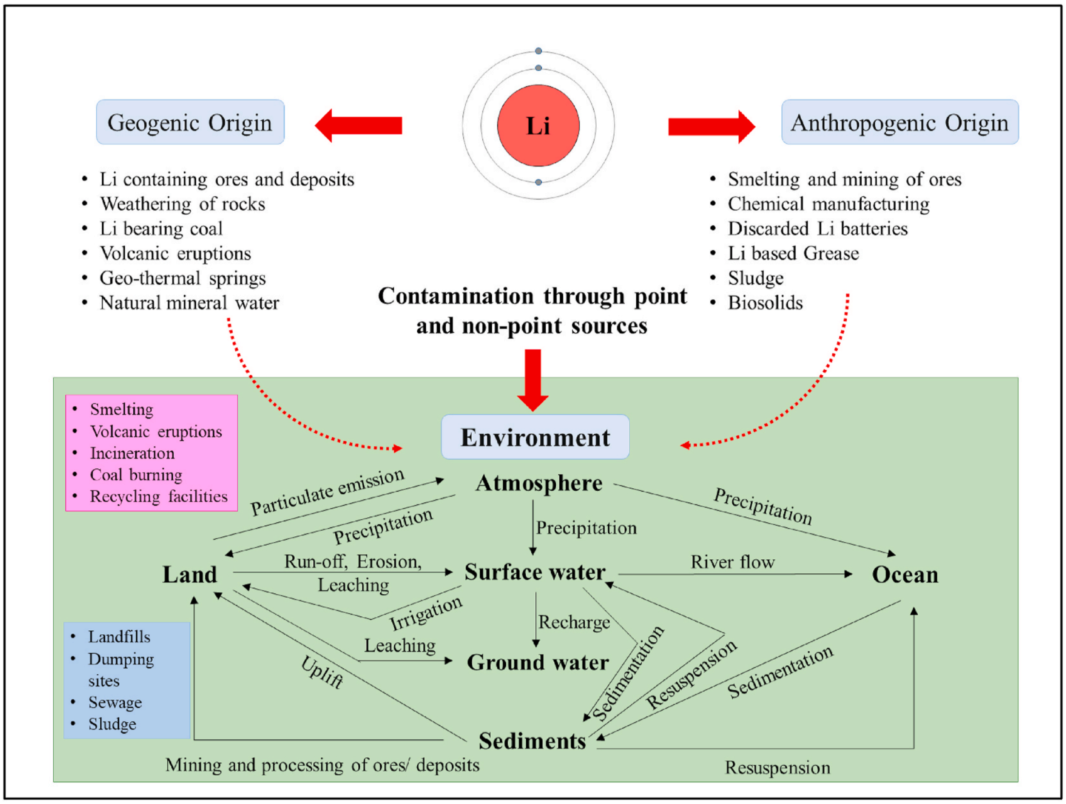


Fig. 2. Sources and pathways of lithium contamination in the environment.

et al., 2010). Coarse-grained, intrusive igneous rocks, pegmatites (such as spodumene, petalite, lepidolite, amblygonite and eucryptite), and sedimentary rocks (mainly clays as hectorite and lacustrine evaporates) contain Li-bearing minerals (Gruber et al., 2011). Aral and Vecchio-Sadus (2011) reported much higher Li contents (200–500 mg kg^{-1}) in authigenic clays than igneous rocks (~30 mg kg^{-1}) and detrital clays (70–80 mg kg^{-1}). Lithium and Li-containing compounds are highly mobile. They can easily leach into surface- and ground-water sources (Yalamanchali, 2012), becoming highly bioavailable, and tend to bioaccumulate in certain microbiota, which could explain the high Li content in authigenic clays.

Millot et al. (2010) reported that the dissolved Li content in the Mackenzie River Basin in Northwestern Canada is mainly from the silicate and carbonate weathering. Compared to carbonates, high Li concentrations (>50 mg kg^{-1}) are associated with silicates, because Li can strongly bind to silicate matrices. Carbonates have low Li (<1 mg kg^{-1}), but the high dissolution rate makes Li available in carbonate-rich regions (Millot et al., 2010). Further, high Li concentrations have been reported in carbonates precipitated from evaporated lake water; for example, 19 mg kg^{-1} of Li in aragonite from the Dead Sea (Aral and Vecchio-Sadus, 2011).

In coal deposits, Li is enriched in extractable quantities (Qin et al., 2015). For the first time globally, Li associated ore deposits were found in coals at the Jungar Coalfield, China (Sun et al., 2010, 2012). Lithium distribution in fly and bottom ashes were similar during and after coal combustion (Qin et al., 2015). In another study, however, Bhangare et al. (2011) found that fly ash contained more Li than bottom ash. In this light, Pougnet et al. (1985) reported that fly ash samples in South Africa had Li concentrations ranging from 65 to 287 mg kg^{-1}.

In freshwater systems, natural sources of Li in water include rock weathering (e.g., bedrock solubilization, desorption from secondary minerals containing Li), precipitation and dry deposition from the atmosphere, and groundwater input (Millot et al., 2010). However, several studies have highlighted low Li contents from natural sources. For example, the major rivers of the United States have low Li concentrations (2.0 μg L^{-1}) (Kszos and Stewart, 2003). Typical background concentrations of Li ranged from 1.0 to 10 μg L^{-1} in surface water and up to 0.5 mg L^{-1} in ground water (Schrauzer, 2002). However, Li levels in ground water of the South East of Ireland reached 500 mg L^{-1} (Kavanagh et al., 2017). In the Massif Central in France, Négrel et al. (2010) reported fluctuating Li concentrations in ground water ranging from 0.07 μg L^{-1} in springs feeding a peat bog to 196 μg L^{-1} in a peat bog of a maar depression. Therefore, fluctuations in Li concentrations in ground water are likely affected by geological factors and hydrological regimes. Worldwide, Li concentrations in mineral water generally range from 0.05 to 1.0 mg L^{-1} but can reach 100 mg L^{-1} in some places (Schrauzer, 2002).

River inputs, weathering of oceanic silicates, and high-temperature hydrothermal fluxes in mid-ocean ridges are the major sources of Li enrichment in oceans (Chan et al., 2006; Misra and Froelich, 2012). Lithium is incorporated into authigenic aluminosilicate clays formed in/on the seafloor (Chan et al., 2006). Apart from the deposited fraction, the dissolved fraction of Li (0.17–0.18 mg L^{-1}) is homogeneously distributed throughout the oceanic water column regardless of depth and latitude (Aral and Vecchio-Sadus, 2011; Misra and Froelich, 2012; Riley and Tongudai, 1964; Thibon et al., 2021). Circulation of water from the tropics to polar regions, and vice versa, and mixing might explain the homogenous distribution of Li in oceanic water. Even though oceans have a large Li reservoir (231.4 trillion tons), recovering Li from seawater is not economically viable due to its homogeneous distribution with low Li concentration in the water (Diallo et al., 2015). Moreover, Li accumulates to high concentrations in calcareous shales and carbonates, precipitated from evaporated seawater or lagoon water (Aral and Vecchio-Sadus, 2011). Besides, brine deposits found in dry lakes (e.g., the Salar de Atacama in Chile) contain high Li concentrations (Yaksic and Tilton, 2009).

Sediments act as a contamination source because they can adsorb and accumulate pollutants. Further, the contamination remains for long periods, even after the pollution has declined (USEPA, 2004). Slukovskii and Svetov (2016) indicated that bottom sediments in small rivers can be used as an indicator for determining the anthropogenic status of alkali metals (including Li) in an urban, because higher concentrations of the pollutants accumulated in sediments than overlying water, with the smallest concentrations in surface water. Kjølholt et al. (2003) reported that the amount of Li accumulated in sediment from road runoff ranged from 15.5 to 16.3 mg kg^{-1}. Notably, the adsorption of pollutants onto suspended sediments and resuspension of adsorbed sediments in the sediment–water interface enrich the pollutants in sediments and their associated water column. A study indicated that approximately 10 mg kg^{-1} of Li was adsorbed onto riverbed sediments and clays (Aral and Vecchio-Sadus, 2011). Elevated levels of Li were reported in the sediments in sabkhas and evaporite deposits of lagoons (Aral and Vecchio-Sadus, 2011). Generally, the concentrations of Li, and rubidium (Rb) and caesium (Cs) in bottom sediments correlate with their contents in parent material (Kabata-Pendias and Mukherjee, 2007).

Major potential natural sources of Li entering the atmosphere include dust and fumes released during volcanic eruptions and airborne soil particles (Kavanagh et al., 2018), but the subsequent wet and dry deposition and associated Li concentrations in terrestrial and aquatic ecosystems have not been studied.

2.2. *Anthropogenic origin*

The world's annual consumption and demand for Li have increased significantly in recent decades and continue to rise with new technologies and growing industries (Choi et al., 2019). Worldwide Li production increased from 28,000 to 95,000 tons in 2018 (USGS, 2017, 2020). Lithium content exceeds its natural threshold level in environmental compartments mainly as a consequence of various anthropogenic activities (e.g., high consumption and improper disposal of Li based products) (Mohr et al., 2012; Winslow et al., 2018).

The anthropogenic use of Li-based compounds occurs in aluminium (Al) processing, chemical treatments (e.g., nano-Li concrete floor treatments, air purification, organic compounds, and as drying agents), pharmaceuticals, lubricants, Li-ion batteries (LIBs) for electrical appliances and electric vehicles, and glass and ceramic production (Hao et al., 2017; Kokkotis et al., 2017; Tanveer et al., 2019; Winslow et al., 2018). By 2025, >80% of the total Li market will be used to produce LIBs (Harper et al., 2019). Apart from Li, spent LIBs also contain other PTEs, including Co, Mn, Ni, Fe, Cu, Al, and organic compounds (Lv et al., 2018; Siqi et al., 2019; Sun et al., 2017). The PTE concentrations in LIBs are often higher than those in naturally occurring ores (Meshram et al., 2015). Furthermore, LIBs need to be replaced after 300 to 500 charge cycles or two to three years. Moreover, Li has the worst recycling recovery rate of all PTEs used in LIBs (Harper et al., 2019). Consequently, spent LIBs are a potential source of contaminants in the environment. The ultimate destinations of spent LIBs are landfills (municipal solid waste dumping sites), specialized recycling facilities, and waste-to-energy facilities (Bernardes et al., 2004). However, consumers dispose of spent LIBs in municipal solid waste due to the lack of awareness about the toxicity of spent LIBs (Aral and Vecchio-Sadus, 2008). If Li-containing wastes are disposed of in landfills or open dumps or buried in soil, the chemical content in the wastes, together with other substances, can leach into soil and contaminate surface- and ground-water sources. During the compaction process of landfills, the outer casing of batteries and other devices can be broken, or the casing can degrade chemically, releasing toxic substances to the natural environment (Li et al., 2009b; Winslow et al., 2018). Richa (2016) found that a lithium nickel manganese cobalt (NMC) oxide ($LiNiMnCoO_2$ or Li-NMC) battery disposed into landfill could leach 42.50% Li, 11.45% Mn, and <4% of the total Co, Ni, Al, Cu, and Fe into the leachate solution. In the United States, Li-contaminated ground water at toxic levels for freshwater organisms (50–170 μg L^{-1}) due to improper waste disposal practices had been noticed (Kszos and Stewart, 2003).

Apart from landfill leachates and dumping sites, anthropogenic sources of Li that can contaminate soil, and surface- and ground-water sources include chemical manufacturing facilities, spills from manufacturing and recycling facilities, and industrial effluents (Kszos and Stewart, 2003; Winslow et al., 2018). The lubricating greases used in vehicles manufactured from $LiOH{\cdot}H_2O$ can release Li into the environment through surface-water runoff from roads (Aral and Vecchio-Sadus, 2008; Martin et al., 2017; Yalamanchali, 2012). Agricultural and soil amendments are another potential sources of Li contamination, because biosolids (sludge) from wastewater treatment facilities are used as soil amendments (Aral and Vecchio-Sadus, 2008; Kjølholt et al., 2003). Lithium concentration in sludge from a municipal wastewater treatment facility ranged from 5.0 to 6.0 mg kg^{-1} (Kjølholt et al., 2003). Furthermore, composts from household and garden wastes contained nearly 4.6 mg kg^{-1} of Li (Kjølholt et al., 2003). However, there are no comprehensive data on the amount of Li present in composts and biosolids in various countries.

In a riverine system, Li contamination can occur through geogenic sources, such as leaching from natural weathering of silicate rocks and minerals (Millot et al., 2010). Anthropogenic activities from urban areas and industries are also responsible for increased Li content in rivers and other water bodies (Choi et al., 2019). Urban runoff is a significant source of Li pollution. Many substances deposited on surface materials are washed off and transported to nearby drainage systems and water bodies. Choi et al. (2019) showed that the downstream area of the Han River in Seoul (South Korea) contained a high Li concentration (1.57 mg L^{-1}) compared to the upstream area (0.28 mg L^{-1}), due to pollution from LIBs, therapeutic drugs in hospital waste, and food waste from households and industries. Tap water for human consumption was also contaminated with Li because it came from the Han River. The Li contamination was aggravated due to the inefficiency of wastewater treatment processes for Li removal. Indeed, Li concentration was not significantly different between influent and effluent wastewaters (Choi et al., 2019). In the USA, the Environmental Protection Agency (EPA) has regulated Li regional screening level of 40 μg L^{-1} for residential tap waters (Winslow et al., 2018).

Annually, more than 600,000 tons of Li are extracted from ore deposits (e.g., spodumene, lepidolite, petalite, amblygonite, and zinnwaldite) and mineralized zones (USGS, 2017), which are circulated in the environment, with around 9510 tons of Li assumed to end up as wastes (Peiró et al., 2013). Therefore, mining, smelting, and foundries are point sources for major Li contamination during the exploitation and processing of ore deposits (Tanveer et al., 2019). Processing ore results in the disposal of tailings and discharge of tailing effluents containing Li, contaminating all three compartments of the environment, i.e., air, water, and soil. Moreover, tailing water is repeatedly used without any additional treatment intensifying the dissolved Li content in water systems (Shah et al., 2016). Finely ground Li minerals (e.g., Li-containing phosphate ores) are more readily dissolved in water due to their increased surface area (Aral and Vecchio-Sadus, 2008). Aral and Vecchio-Sadus (2011) reported that Li mineral beneficiation plants had high concentrations of Li (>15 mg L^{-1}) dissolved in tailing water. Figueroa et al. (2012) mentioned that Li contents in surface-water sources of northern Chile were remarkably high due to the presence of ores from actively mined areas. Lithium is also present as a trace metal impurity in minerals such as feldspars, micas, and illites (Scott and Smith, 1987). Therefore, Li can also enter the environment through waste-rock dumps and mine drainage (Kavanagh et al., 2018).

Anthropogenic sources of Li emission into the atmosphere include the burning of coal enriched with Li, dust, and minute particles released during ore-processing activities, domestic e-waste from recycling centres, and during waste incineration (Bernardes et al., 2004; Li et al., 2010; Winslow et al., 2018). Dai et al. (2010) found that the volatile loss of Li during the combustion of coal was less than 5%. Nevertheless, once in the atmosphere, Li can be condensed on fine particles and transported by wind over long distances, and may contribute to the deposition of Li in terrestrial and aquatic environments.

3. Distribution and speciation of lithium in the environment

The environmental chemistry of Li has received much attention due to its numerous and important applications for green energy and the automotive and health sectors (Négrel et al., 2017). Additionally, Li's stable isotope structure plays a vital role in gathering quantitative geochemical information about various Earth processes (i.e., sediment recycling, global chemical weathering, carbon cycling, hydrothermal alteration, and groundwater evolution) (Négrel et al., 2010).

3.1. Distribution in soils and sediments

Lithium is often found in traces in soils (Table 1). Lithium is widely distributed in sedimentary rocks and released into soil through the

Table 1

Selected references on lithium concentrations in soil, water, and sediment.

A. Li concentrations in different soils

Locations and soil depth	Soil type	Total Li concentration (mean) (mg kg^{-1})	Reference
Lincoln University dairy farm, New Zealand 0–20 cm	Templeton silt loam	31.8	Robinson et al. (2018)
Jiajika rare metal mining area, China 10–25 cm	Not available	169.5	Xu et al. (2019)
Weinan, China 0–14 m	Not available	28.1	Tsai et al. (2014)
Nearby desert areas 0–20 cm	Mud crust	24.3	
Transbaikal region, The Republic of Buryatia 0–20 cm	Gray forest soil	25.1	Kashin (2019)
Cecil,	Clayey, kaolinitic	11.49	Anderson et al. (1988)
Iredell,	Fine, montmorillonotic	25.38	
Madison,	Clayey, kaolinitic	11.93	
Louisa,	Fine, montmorillonotic	33.29	
Wakulla; and	Sandy, siliceous	5.82	
Bonifay, USA Soil depth: not available	Loamy, siliceous	3.74	
Jordan Valley (JV); Northern JV; Middle JV; and South JV, Jordan 0–20 cm (topsoil) 20–40 cm (subsoil)	Ustochreptic and ustollic camborthids and calciorthids, ustic torriorthents	Mean soluble Li concentration ranged from 1.04 to 2.68 mg L^{-1}; Soluble Li concentration in subsoil layer was relatively higher than that of topsoil layer.	Ammari et al. (2011)

B. Li concentrations in different water bodies

Location of water sample	Water body	Li concentration (Mean)	Reference
Tibetan Plateau, China	Lake Donggi Cona	4.6–5.7 μmol L^{-1}	Weynell et al. (2017)
	Dongqu River	1.2–1.8 μmol L^{-1}	
	Yellow River	8.3–8.6 μmol L^{-1}	
	Stream	2.8–5.2 μmol L^{-1}	
	Spring	2.9 μmol L^{-1}	
	Pond	5.7–9.3 μmol L^{-1}	
	Hot spring Wenquan	103.1–135.2 μmol L^{-1}	
Stillwater Wildlife Management Area in Nevada, USA	Wetland	>1000 μg L^{-1}	Hallock (1993)
Abia and Imo States, Southeast Nigeria	Springs	2.49 μg L^{-1}	Ewuzie et al. (2020)
	Streams	1.58 μg L^{-1}	
Public supply wells	Groundwater	<1–396 μg L^{-1}	Lindsey et al. (2021)
Domestic supply wells Across USA		<1–1700 μg L^{-1}	
Northeast Iceland	Groundwater	130–10,000 nmol L^{-1}	von Strandmann et al. (2016)
	River	116–237 nmol L^{-1}	
Changjiang, China	River	1260 nmol L^{-1}	Wang et al. (2015)
North Atlantic	Sea water	0.22 ppm	

(*continued on next page*)

Table 1 (*continued*)

A. Li concentrations in different soils			
Locations and soil depth	Soil type	Total Li concentration (mean) (mg kg^{-1})	Reference
Japan	Coastal Water	1.173 ppm	Choubey et al. (2017)
India Ocean	Coastal Water	0.160 ppm	
North Sea	Sea water	0.1 ppm	
C. Li concentrations in sediment			
Location of sediment sample	Sediment	Li concentration (range/mean) (mg kg^{-1})	Reference
China	Top catchment sediment	5.37–400	Liu et al. (2020)
	Deep catchment sediment	5.27–400	
Patos Lagoon, Brazil	Lagoon sediments	10.05–61.61	Niencheski et al. (2002)
Aegean Sea, Greece	Coastal sediments	9.74–37.1	Aloupi and Angelidis (2001)
Costa Rica	Subduction zone sediments	0.50–78.09	Chan and Kastner (2000)
Mariana Island Arc	Subduction zone sediments	7.3–50.8	Bouman et al. (2004)
South Sandwich Island Arc		6.2–57.3	
East Sunda Island Arc		2.4–41.9	
Lesser Antilles Island Arc		35.2–74.3	
Mackenzie tributary	River sediments	57.8	Millot et al. (2010)
Red Arctic tributary		56.8	
Liard tributary		46.1	
Slave tributary Canada		41.2	
Loire River Basin, France	River sediments	41–73	Millot and Négrel (2021)
Dongqu River	River sediments	14.7–44.9	Weynell et al. (2017)
Lake Donggi Cona Tibetan Plateau, China	Lake sediments	52.2	

weathering process (Aral and Vecchio-Sadus, 2008; Chan et al., 1997). Topsoil usually contains less Li due to the interaction of plant roots, i.e., plant uptake (Yalamanchali, 2012). Arid and saline soils generally contain higher background soil Li concentrations than other soils, due to the selective concentration of alkaline metals, including Li, Na, and K (Yalamanchali, 2012; Hayyat et al., 2021). Lithium concentration in the clay fraction was reported ranging from 7 to 200 mg kg^{-1} (Schrauzer, 2002). In soils with high salt content, such as sodic and saline soils, Li concentrations can exceed 200 mg kg^{-1} (Yalamanchali, 2012). Lithium can replace Al in montmorillonite, and Mg in several minerals due to the similar size of their atomic radii (90 and 86 pm (pm), respectively) (Mason and Moore, 1985). Clay minerals characteristically concentrate Li (Anderson et al., 1988), retaining significantly more than other inorganic soil fractions (Schrauzer, 2002). Anderson et al. (1988) suggested that Li may be present in ditrigonal cavities of clay minerals.

Anderson et al. (1988) studied Li distribution in different layers of Bonifay soils. The authors determined that the concentrations (both total and exchangeable forms) of Li are directly proportional to depth, with the top 60 cm of soil profile containing the lowest Li content. Increasing Li concentrations with depth are likely due to the increase in clay minerals. Clay minerals can include Li through isomorphous substitution, where the structural cations present in the tetrahedral and octahedral sheets in clay minerals are likely substituted by cations with a similar charge and ionic radius (Anderson et al., 1988; Bolan et al., 1999).

Lithium is taken up by all plants, although the element appears not to be essential for their growth and development (Shahzad et al., 2016). Schrauzer (2002) and Lenntech (2007) reported that Li stimulates plant growth. Plants generally have 0.2–30 mg kg^{-1} Li depending on their preferential uptake or exclusion of available Li in soils. For instance, in soil spiked with 5 mg kg^{-1} Li, the amount of Li taken up by plants and stored in leaves was up several hundred mg kg^{-1}, without any loss in the plant biomass (Robinson et al., 2018).

Robinson et al. (2018) observed a strong positive relationship between Li and Al, B, Fe, K, Mg, Mn, and Zn in soil. In contrast to other cations in soil, Li is relatively mobile; it can therefore be taken up by plants or leached into receiving waters (Robinson et al., 2018). Much like other PTE cations, the soil adsorption coefficient (K_d) increased exponentially with increasing pH and decreased with increasing Li concentration (Robinson et al., 2018). For example, Li and Liu (2020) found that the sorption of Li onto kaolinite is pH-dependent, and the adsorbed Li proportion increased with an increase in pH from 3 to 10.

Négrel et al. (2017) investigated Li distribution in agricultural and grazing-land soils from 33 European countries spanning more than 5.6 million km^2 in different climate zones and landscapes. The authors reported a mean Li concentration in these soils of 11.4 mg kg^{-1}, about one-fourth of that in the Earth's upper continental crust (41 mg kg^{-1}). In addition, northern Europe soils had predominantly low Li concentrations (median 6.4 mg kg^{-1}), and southern Europe soils had significantly higher values (median 15 mg kg^{-1}) (Négrel et al., 2017). In southern Europe, climate (e.g., temperature and precipitation) increased weathering, accounting for the secondary Li enrichment in the region, with high Li concentrations observed in limestone areas. The spatial variance in Li concentration across Europe was also associated with parent materials and their weathering products (Négrel et al., 2017).

Hydrated cations tend to sorb to surfaces of soil- and sediment- (in aquatic systems) colloids with a binding strength proportional to the quotient of their charge or hydrated ionic radius (Sposito, 2008). Table 2 shows that Li^+ has weaker binding strength than all other metals present in soil solution. There are considerable energy differences between the bonding orbitals on Li^+ and the corresponding orbitals on various functional groups of soil colloids or aquatic sediments (Cotton and Wilkinson, 1980). It is, therefore, unlikely that any significant covalent inner-sphere bonding of Li^+ by the functional groups of soil colloids. Robinson et al. (2018) reported that Li^+, when added to soil, is more mobile than other trace elements, with typical K_d values ranging from <1 to 10. As with other soluble cations primarily sorbed via outer-sphere processes, the K_d value for Li^+ was found to increase with enhancing pH and decrease with increasing ionic strength.

In contrast to ionic Li^+ introduced through Li compounds, geogenic Li present in clay minerals is sparingly soluble. The K_d of geogenic Li ranges from 200 to 500 (Milićević et al., 2018; Robinson et al., 2018), indicating that Li is essentially immobile in aluminosilicate lattices and enters soil solution gradually from the weathering of sedimentary minerals and phyllosilicate clays (Aral and Vecchio-Sadus, 2008; Chan

Table 2

Charge and hydrated ionic radii of lithium and other significant cations in soil, ranked in order of selectivity for outer sphere binding.

Elements	Ionic radius hydrated (pm)	Charge/hydrated radius (e pm^{-1})	References
Al^{3+}	480	6.25×10^{-3}	(Israelachvili, 2011; Volkov et al., 1997; Wiberg, 2001)
Pb^{2+}	401	4.99×10^{-3}	
Ca^{2+}	412	4.85×10^{-3}	
Cd^{2+}	426	4.69×10^{-3}	
Mg^{2+}	428	4.67×10^{-3}	
Zn^{2+}	430	4.65×10^{-3}	
K^+	331	3.02×10^{-3}	
Na^+	360	2.78×10^{-3}	
Li^+	382	2.62×10^{-3}	

et al., 1997).

Lithium is also found in sediments of water bodies such as lakes, rivers and marine (Chan et al., 2006; Weynell et al., 2017). Sedimentary sources of Li originate when Li is slowly washed out of volcanic minerals into basins, where the element reacts with other minerals in water bodies. In addition, water-rock interactions within water bodies, soil erosion and wet and dry depositions are also sources of Li in sediments. Lithium in sediments is mainly bound to Fe–Mn oxide and residual fractions (Li et al., 2021). Generally, Li concentrations in sediments are higher than those in the continental crust, indicating that Li is enriched during sediment formation (Liu et al., 2020). Various factors such as water flow, ionic strength, and rock types affect level and composition of Li in sediments (Li et al., 2021; Liu et al., 2020).

Several Li fractions, which differ in their mobility and interaction, are incorporated in soil and sediment components. Lithium existing in soil and sediment matrices include exchangeable (mainly carbonate phases), oxidizable (sulfides and organic matter), reducible (Fe/Mn (hydro)-oxides), and residual (silicate phases) fractions. The Community Bureau of Reference (BCR) sequential extraction protocols are commonly used to study speciation of metals, including Li, in soils and sediments (Kumkrong et al., 2021; Tong et al., 2020). For instance, Li et al. (2020) studied Li distribution in a shallow arid regolith using this sequential extraction. The residual fractions ranged from 59 to 84% of total Li while the exchangeable fractions contained the lowest Li content (<2%). The oxidizable fractions and reducible fractions accounted for 1–5% and 3–28%, respectively.

3.2. *Distribution in aquatic environments*

In natural waters, Li concentrations are correlated with other alkali metals, particularly Na. In freshwater systems, the average Li concentration is relatively low (<3 μg L^{-1}) in comparison to other alkali metals (e.g., Na, K) (Mason and Moore, 1985). Higher concentrations occur in streams and lakes surrounded by Li-rich rocks and soils (Kavanagh et al., 2017) (Table 1). As with Na, Li concentrations are orders of magnitude higher in sea water (170–190 μg L^{-1}) than in fresh water (0.07–40 μg L^{-1}) (Mason and Moore, 1985). Lithium is often found in trace concentrations in surface- and ground-water sources (Aral and Vecchio-Sadus, 2008). As a highly soluble ion, Li in water systems is primarily present as ions. Due to its low concentration in the natural environment, Li does not usually cause any detrimental effects to the environment. In hydrological studies, LiCl is often used to trace streams due to its highly soluble and chemically inert characteristics. Unlike Na and K that account for >2% of the Earth's crust, Li only accounts for 0.0065% (Hou et al., 2014; Tarascon, 2010).

Whereas the naturally occurring Li concentration in surface waters is generally less than 0.04 mg L^{-1}, its concentration is elevated in Li-contaminated water bodies (Emery et al., 1981; Hill and Gilliom, 1993; Mathis and Cummings, 1973; Tanner, 1995). Only few studies reported Li concentration in drinking water, with values ranging from 1.0 to 10 μg L^{-1} (Anderson et al., 1988; Choi et al., 2019; Durfor and Becker, 1964; Sievers and Cannon, 1975).

4. Bioavailability and toxicity of lithium to biota, including humans

4.1. *Toxicity to microorganisms and plants*

Lithium is considered toxic to organisms at high concentrations. However, application of Li_2O nanoparticles and Li^+ ions to soil at 500 mg kg^{-1} increased methane, nitrous oxide, and carbon dioxide emissions (Avila-Arias et al., 2019), paradoxically indicating some stimulatory effect of the compound on soil respiration, possibly due to an increase in soil pH. These results also indicate that Li is relatively non-toxic to soil microorganisms (Avila-Arias et al., 2019). In contrast, high Li concentrations impede plant growth; Li phytotoxicity greatly depends on plant species and type of Li salt. Plants can take up Li in soil; Li contents in plants grown in uncontaminated soils can be as low as a few mg kg^{-1} dry weight. For example, Li contents in plants grown in uncontaminated pasture soils in Western Transbaikalia (Russia) and New Zealand were 2–4 mg kg^{-1} and 1.5 mg kg^{-1} (dry weight), respectively (Kashin, 2019; Robinson et al., 2018). Leaves of pepper, cucumber, potato, and citrus from the Jordan Valley contained foliar Li concentrations of 5–30 mg kg^{-1} (Ammari et al., 2011). Given the relatively low solubility of geogenic Li in uncontaminated soil (<0.1 mg L^{-1}), these results indicate that Li in soil solution is readily taken up by plants.

Lithium can enter plant roots via the apoplastic and symplastic pathways (Tanveer et al., 2019) (Fig. 3), and its entry can be facilitated by the non-specificity of uptake pathways for similarly sized ions such as Mg^{2+} (Reid and Hayes, 2003). In soils where Li^+ has been added, bioaccumulation coefficients of >10 have been reported for perennial ryegrass, sunflower (Robinson et al., 2018), lettuce, buckwheat, and maize (Franzaring et al., 2016). Plant uptake of Li could be limited only by the tolerance of plants to Li. In perennial ryegrass and lettuce, Li contents (up to 1000 mg kg^{-1}) in the plant tissue were reported with little effect on biomass (Kalinowska et al., 2013; Robinson et al., 2018). Sword-leaf dogbane, used in Chinese medicine, can accumulate >1800 mg kg^{-1} Li (Jiang et al., 2014). The low plant toxicity of Li combined with high bioaccumulation coefficients indicates that plants will afford little food-chain protection from Li contamination compared to other soil contaminants (Chaney, 1980; Henschel et al., 2020; Hayyat et al., 2021).

Although Li is not considered to be essential for plants, some studies have indicated that deficiency of this element affects plant growth and development (Shahzad et al., 2016). Lithium reduces plant morphological and physiological growth by altering plant metabolism, such as developing necrotic spots due to ethylene accumulation (Naranjo et al., 2003), reducing enzyme activities during pollen development (Gumber et al., 1984), and altering gravi-trophism in plant roots (Mulkey, 2007). Lithium also affects plant metabolism in different organelles (e.g., mitochondria, peroxisomes, and chloroplasts) (Qiao et al., 2018), and its impact is considered non-specific because Li is able to substitute other important monovalent cations (e.g., Na^+, K^+) in plant cells (Kabata--Pendias and Mukherjee, 2007). Lithium inhibits inositol monophosphatases and calcium signalling at high concentrations by interfering with the activity of inositol monophosphatases and dephosphorylation of inositol-1-phosphate (Baran, 2019; Murry et al., 2019).

Because Li complexes more readily with organic and inorganic anions than other alkali metals, it can replace these metals (e.g., Na, K) at the uptake sites of biota, including higher plants (Aral and Vecchio-Sadus, 2008). This complexing characteristic may explain the lower K concentrations at higher Li exposures in spinach (antagonistic effect) (Bakhat et al., 2019). A similar effect was also observed for Ca uptake in the plant species (i.e., Ca concentration in the spinach shoots was decreased by increasing Li concentration in soil) (Bakhat et al., 2019) (Table 3). Therefore, Li can induce various Ca-dependent responses inside plant bodies by involving in glutamate-receptor and acting via the inositol signalling pathway, as suggested by Stevenson et al. (2000). Jacobson et al. (1960), however, found that Ca regulated K acquisition in plants by reducing Li uptake. The authors proposed that Li and Ca compete for absorption sites in roots, while Li hinders K uptake in plants.

Lithium toxicity reduces photosynthesis by reducing chlorophyll content (Table 3). For instance, in maize, Li toxicity reduced chlorophyll *a* content by 45% and carotenoid content by 67% (Hawrylak-Nowak et al., 2012). Likewise, Li toxicity reduced chlorophyll content and caused necrotic spots in lettuce (Kalinowska et al., 2013), Ethiopian mustard (Li et al., 2009a), avocado, sour orange, and soybean (Bingham et al., 1964). A Li-induced decrease in photosynthesis might be linked to changes in content and stability of pigment-protein complexes as well as in metabolite composition, deprivation of chlorophyll contents, and

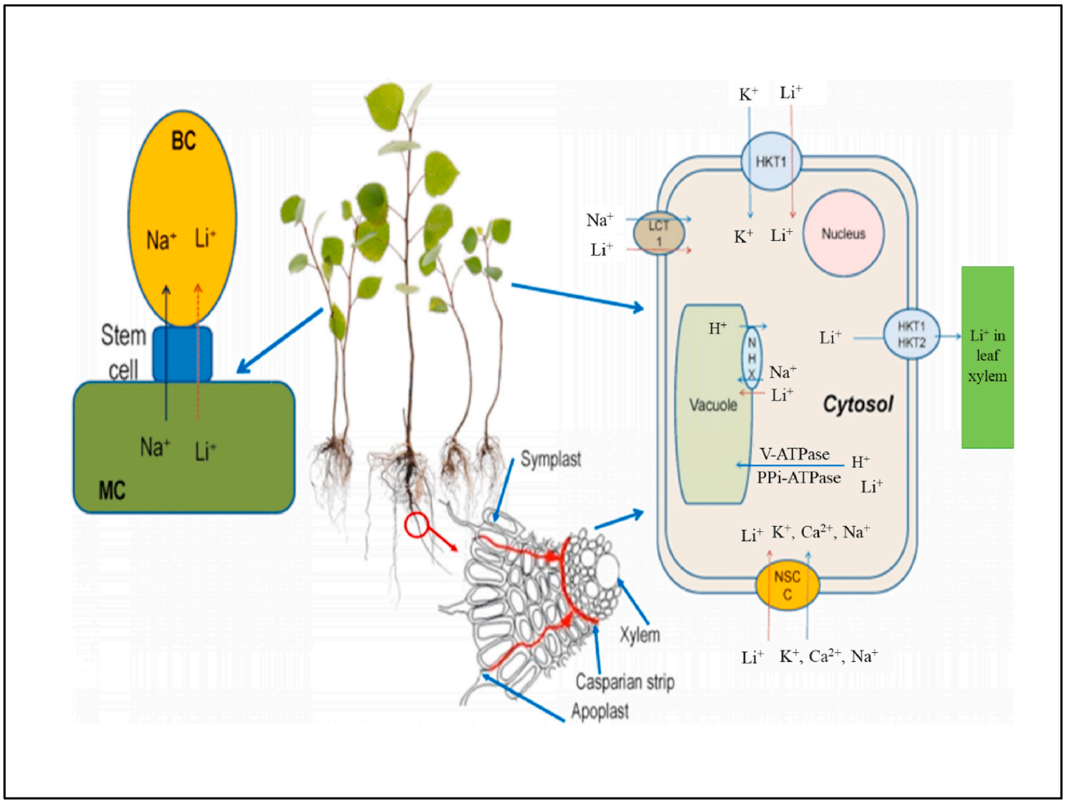


Fig. 3. Apoplastic and symplastic pathways of lithium movement into plant roots.

Table 3
Selected examples of the response of different plant species to lithium toxicity.

Plant species	Growth media	Lithium concentration	Plant species responses	References
Apocynum venetum	Soil	<200 mg kg^{-1}	Reduced biomass production, accompanied by reduced chlorophyll contents and leaf gas exchange	Jiang et al. (2014)
Helianthus annuus	Nutrient solution	<60 mM	Reduced hypocotyl length (34–55%, relative to control), and in circumnutation (30–70%).	Stolarz et al. (2015)
Lactuca sativa	Nutrient solution	50 and 100 mg dm^{-3}	Reduced shoot biomass (58–69% and 91–95% at 50 and 100 mg dm^{-3}, respectively, relative to control) and formation of necrotic spots. Reduced of root biomass ($\sim$40 and $\sim$80% at 50 and 100 mg dm^{-3}, respectively, relative to control)	Kalinowska et al. (2013)
Zea mays and *Helianthus annuus*	Nutrient solution	50 mg dm^{-3}	Reduced shoot biomass (27 and 32% in *H. annuus* and *Z. mays*, respectively) and formation of necrotic spots in *H. annuus.* Reduced chlorophyll *a and b* by 47 and 43%, respectively in *Z. mays.*	Hawrylak-Nowak et al. (2012)
Spinacia oleracea	Soil	$>$20–80 mg kg^{-1}	Reduced dry weight with increasing Li concentration. Higher activities of antioxidant enzymes in shoots of *S. oleracea* with increasing Li concentration.	Bakhat et al. (2019)

reduction in Mg^{2+} uptake (Kabata-Pendias and Mukherjee, 2007; Shahzad et al., 2017).

Most importantly, Li toxicity induces oxidative damage by promoting the generation of reactive oxygen species (ROS) (Kiełczykowska et al., 2004; Oktem et al., 2005) (Fig. 4). High levels of malondialdehyde (a marker for oxidative stress) in plants in response to Li exposure have been reported (Hawrylak-Nowak et al., 2012; Naranjo et al., 2003). Lithium can also induce inhibitory effects on nucleic acids (important components of overall plant metabolism) and protein biosynthesis, leading to the alteration of translation and transcription process (Aral and Vecchio-Sadus, 2008; Allagui et al., 2007; Dichtl et al., 1997). Moreover, Li-induced ROS might be associated with the inactivation or physiologically zero activation of enzymatic antioxidant systems, or reduced transcription of genes involved in antioxidant defence. In plants, Li toxicity can increase intracellular levels of O^{2-} or OH^- radicals through a Fenton-type reaction known to initiate lipid peroxidation (Shahzad et al., 2016). Plants' Li sensitivity and their tolerance to this element may vary with species. For instance, plant species from Asteraceae and Solanaceae families showed Li tolerance (Kabata-Pendias and Mukherjee, 2007; Schrauzer, 2002), while citrus plants showed Li sensitivity (Aral and Vecchio-Sadus, 2008; Bradford, 1973); the species differences might be due to the activation of different antioxidant enzymes (Tanveer et al., 2019). Production of various antioxidants is not in a similar manner, or in direct proportion to stress severity; it can depend on the study sample and sampling time (Tanveer and Shabala, 2018). For instance, glutathione peroxidase (GPX) activity was inhibited at 1 mM Li (7.0 mg L^{-1}), whereas superoxide dismutase activity was promoted, indicating the dissimilar response of various antioxidants to Li stress (Nciri et al., 2012). Hence, it is warranted to examine antioxidant activity in response to Li exposure in plants under different experimental conditions (both in vitro and in vivo study).

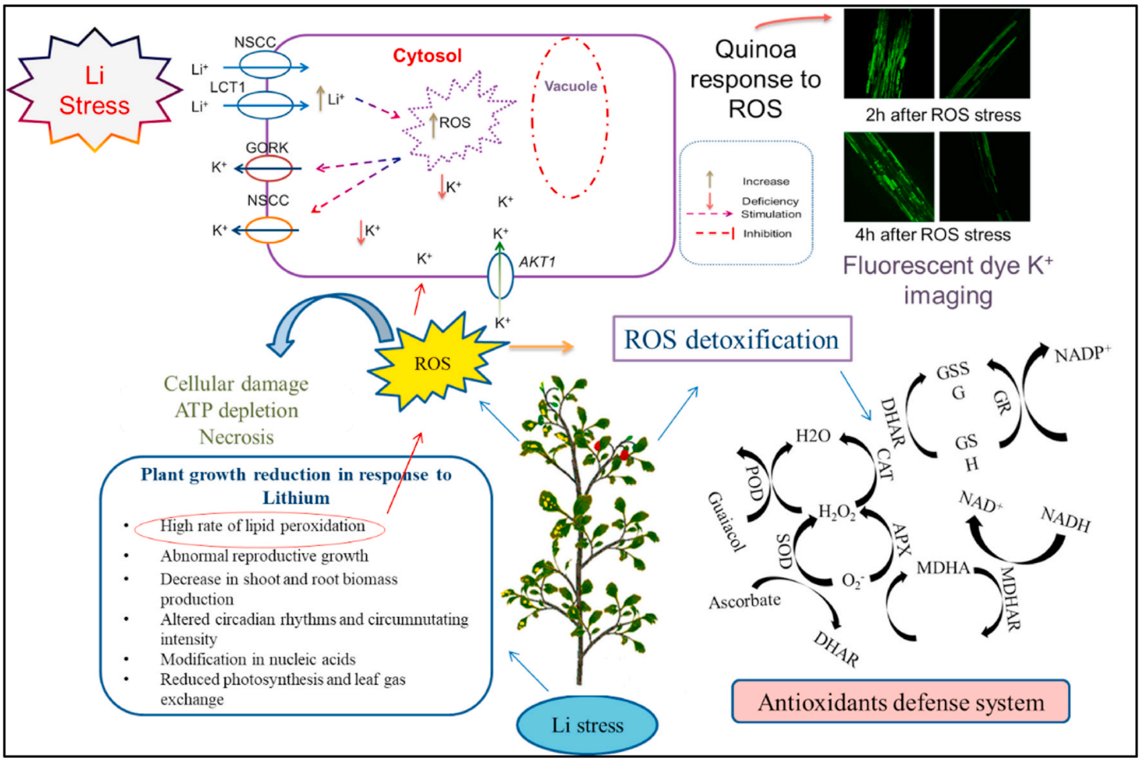


Fig. 4. A typical model, showing the response of plants under Li stress. Upon Li stress, Li reduced plant growth by reducing and altering numerous physiological mechanisms, and ROS production is major one, which concomitantly induces oxidative stress. Increase in cytosolic Li concentration (via non-selective cation channels (NSCC) and low cation channels 1(LCT1)) results in high ROS production (possibly via the Fenton reaction), which further results in the activation of ROS activated K^+-efflux channels, K^+-outward rectifying channel (GORK), or non-selective cation channels with the concomitant development of K^+ deficiency in the cytosol. Such a decline in cytosolic K^+ deficiency further induces ROS production. Thus, Li induced nutrient deficiency might be plausibly and highly linked with ROS production. A typical example of the response of quinoa to ROS stress showed that, the longer the timing of ROS exposure, the higher was the decline in K^+ fluorescent in quinoa roots (unpublished results). In response to oxidative stress, activation of the antioxidant system is a key Li tolerance mechanism in plants, which also plays an important role in reducing K^+ deficiency under Li stress.

4.2. Toxicity to aquatic life

Elevated Li concentrations in various waters have been reported, which negatively affects aquatic life. For instance, in northern Chile, Li concentrations in ground water is up to 500 mg L^{-1}, while in surface waters it only reaches 5.2 mg L^{-1} (Schrauzer, 2002; Zaldívar, 1980). In the USA, Li was detected at 0.002 mg L^{-1} in some major rivers (Kszos and Steward, 2003). Thus, it is important to examine the sources of Li contamination in water and the Li-induced detrimental effects on aquatic life.

Kszos et al. (2003) investigated Li toxicity to three aquatic organisms [*Pimephales promelas* (fathead minnow), *Ceriodaphnia dubia*, and a freshwater snail (*Elimia clavaeformis*)] by adding elevated Li levels to induce toxic effects. Under laboratory, hydroponic-solution-culture conditions, Li suppressed the growth of *P. promelas* and reproductive response of *C. dubia* as measured by 25% inhibitory concentration (IC_{25}–Li concentration which caused 25% inhibition in growth), but the results varied depending on the composition of the solution culture. In the hydroponic solution containing ~2.8 mg Na L^{-1}, IC_{25} values for *P. promelas* and *C. dubia* were 0.38 and 0.32 mg Li L^{-1}, respectively. These values increased to 1.99 and 3.33 mg Li L^{-1}, respectively, in ambient stream water containing ~17 mg Na L^{-1}, indicating lower toxicity at higher Na concentration (Kszos et al., 2003). In addition, the feeding patterns of *E. clavaeformis* were disrupted when Li concentrations reached 0.15 mg L^{-1}. However, adequate Na content in the solution enabled *P. promelas* and *C. dubia* to tolerate Li concentrations >6 mg L^{-1}. The authors stated that the reproduction of *C. dubia* was not affected when exposed to a mixture of Li and Na with a logarithmic ratio of mmol Na: mmol Li > 1.63. The concentrations of Na in most natural waters are sufficient to inhibit Li toxicity (Kszos et al., 2003). However, a comprehensive water quality assessment is warranted, especially in areas with historical Li use, disposal or recycling processes.

Based on the Li^+ lethal concentration (LC_{50}) in the growth medium, the sensitivity of different fish species was in the range of 13 mg L^{-1} to >100 mg L^{-1} (Hamilton, 1995; Long et al., 1998). Likewise, the lowest observed effect concentration (LOEC) of 200 μg L^{-1} Li was observed in the hepatopancreas, while it was 1000 μg L^{-1} in the gills in the epidermis (Sawasdee et al., 2011). Moreover, embryonic development of *M. cornuarietis* was highly sensitive to Li, with an LC_{50} value of 2500 μg L^{-1} Li (Sawasdee and Köhler, 2010). LC_{50} values from 1.2 to 8.7 mg L^{-1} Li were observed for *P. promelas* after 26 days of Li stress, and 9.2–62 mg L^{-1} in white cloud mountain minnow, *Tanichthys albonubes*, after 48 h of Li stress (Lenntech, 2007).

Several case studies on Li toxicity in *O. mykiss* have underpinned some key mechanisms involved in the Li-induced negative effects on trout; in particular, the interference of Li with Na^+ and K^+ transport. Fish gills uptake Li in water most likely via a putative Na^+ channel, and ionic disturbance in the pavement cells of fish gills alters the ionic homeostasis (Bury et al., 2003; Wood, 2001). Lithium decreases branchial citrate synthase activity and alters the regulation of Na^+ and K^+-ATPases, thus reducing trout growth (Tkatcheva et al., 2007a). Moreover, Li disrupts the ultrastructure and lipid composition of fish gills by altering membrane fluidity and increasing sphingomyelin (Tkatcheva et al., 2007b; Tkatcheva et al., 2004). Lithium also imposed negative effects on plasma due to high arachidonic acid production, inhibition of prostaglandin synthase, and reduced concentrations of Na^+, K^+, and Mg^{2+}. Although these studies revealed some impacts of Li on different metabolic mechanisms in trout, it remains unclear how Li interferes with them. Potentially, electrophysiological and gene-expression approaches can answer this question.

In addition, the presence of Li in sediments causes potential toxic impacts towards benthic communities (Viana et al., 2020). Benthic organisms have an important role to play in the continuity of the aquatic ecosystem; therefore, the protection of benthic habitats (i.e., sediments) is of great importance. Empirical sediment quality guidelines (SQGs) have been developed to predict the potential toxicities of contaminants in aquatic sediments posed to sediment-dwelling organisms (Kwok et al., 2014). Threshold effect level and probable effect level derived from SQGs were proved to reliably predict the toxicities of both inorganic and organic pollutants in sediments (Zhang et al., 2017). However, this information is still lacking in the case of Li and thus, further investigation is warranted for toxicity and risk assessment of Li in sediments.

4.3. *Risks to humans and animals*

Lithium can enter animal and human bodies through drinking water and plants (Franzaring et al., 2016). Whereas several studies have reported positive effects of Li in drinking water (at low concentrations) on human mental health and quality of life, some health groups are concerned about the detrimental effects of Li at high concentrations. For instance, high Li levels in waterbodies and tap water due to pollution from Li batteries, caused toxicity to human cardiomyocytes (Shen et al., 2020). Indeed, Li significantly repressed cell capability and proliferation of human cardiomyocytes and promoted cell apoptosis. These effects were attributed to the regulation of glycogen synthase kinase 3 beta (GSK3β), an enzyme in humans that is encoded by the GSK3β gene (Shen et al., 2020). Abnormal regulation and expression of GSK3β are associated with mood disorders, bipolar disorder and depression, and schizophrenia (Jope and Roh, 2006).

In addition, high Li concentrations can affect hematopoietic stem cell differentiation and glycogen synthesis during fetal development (Phiel and Klein, 2001). However, Li at suitable doses could be used to treat several mental illnesses, including depression, suicide, dementia, and bipolar effect (Kessing et al., 2017; Ohgami et al., 2009; Won and Kim, 2017), and, thus, a safe limit of Li is required.

Lithium toxicity results in abnormalities and dysfunctions in several metabolic pathways, causing serious problems to human health (Table 4). Cardiotoxicity and changes in benign electrocardiograms and the development of fatal arrhythmias due to Li toxicity have been reported (Maddala et al., 2017; Mehta and Vannozzi, 2017). High Li levels can also lead to dysfunction of sinus node dysfunction, prothrombin ratio (PR), QT prolongation (a measure between Q wave and T wave in the heart's electrical cycle), heart blocks, and ventricular tachyarrhythmias (Goldberger, 2007; Mehta and Vannozzi, 2017; Waring, 2007). Lithium overdose can induce several nephrological disorders in humans such as cognitive impairment, coma, autism, and stupor, and weight gain (McKnight et al., 2012; Munshi and Thampy, 2005).

Table 4
Selected examples of the effect of lithium intoxication in different body parts in humans.

Body part	Acute symptoms	Chronic symptoms	References
Neuron	Tremor, muscle twitching, cognitive impairment	Muscle seizures, coma, stupor	(Dunne, 2010; Gong et al., 2016; Malhi et al., 2020; Shahzad et al., 2017; Timmer and Sands, 1999)
Renal	Water and Na^+ diuresis	Hyperchloremic acidosis, renal acidosis, renal failure, insipidus	
Gastrointestinal	Mild nausea and vomiting	Severe vomiting, diarrhea, nausea	
Neuro-muscles	Neuropathy	Psychosis	
Cardiac	Prolonged PR, and QT intervals[a]	Myocarditis, development of arrhythmia, heart blocks	
Weight gain	Development of diabetes	–	
Thyroid	–	Development of hyperparathyroidism; high Ca^{2+} concentration in blood	

[a] PR (prothrombin ratio); QT (measure between Q wave and T wave in the heart's electrical cycle).

Renal toxicity is another consequence of Li overdose. Renal toxicity increases water and Na^+ diuresis, dehydration, hyperchloremic metabolic acidosis, renal tubular acidosis, and reduction in urinary concentrating ability (Gong et al., 2016; Grünfeld and Rossier, 2009). Lithium further alters renal functionality by dysregulating aquaporin activity and entering apical membranes via epithelial Na^+ channels, which inhibit signaling pathways involving glycogen synthase kinase type 3β (Davis et al., 2018; Grünfeld and Rossier, 2009). Hyperparathyroidism (overproduction of thyroid hormones) is another inevitable feature of Li toxicity (Palmér et al., 1987). Such a response is attributable to Li-induced inactivation of Ca^{2+} sensing receptors and interference with intracellular second messenger signalling, which concomitantly results in high parathyroid hormone production and Ca^{2+} concentrations in the blood (Szalat et al., 2009). Lithium, after oral ingestion, does not bind to serum proteins in the gastrointestinal tract (GIT), but completely dissolves in water in the human body and remains there for a long time, causing renal failure in response to Li toxicity.

A few studies have indicated the toxic effects of Li on different animals, with the toxic or fatal dose depending on species and Li compounds. For example, doses of 500 and 700 mg kg^{-1} body weight showed severe depression and ataxia in beef cattle (Johnson et al., 1980), with residual Li found in striated muscle, heart, liver, kidney, and brain tissues. Using lithium hypochlorite (LiClO) as the sole source of drinking water for two dogs, Davies (1991) reported clinical signs of polyuria, polydipsia, loss of body mass, dehydration, diarrhea, and general weakness in one dog, and, polyuria, polydipsia, loss of body mass, and seizure in the other dog. Similarly, Li exposure significantly increased lipid peroxidation, decreased renal functionality, and caused weight loss in rats (Schrauzer, 2002; Tandon et al., 1998). Overdose supplementation of Li to a pregnant mouse caused drastic malformations in offspring, indicating that Li-induced effects can be transferred to offspring (Smithberg and Dixit, 1982). Due to the similar physicochemical properties of Li^+ with Na^+ and K^+, Li interferes with the uptake of these essential elements and alters cellular ionic homeostasis affecting the mechanism associated with dose-dependent inhibition of myocyte voltage-gated Na^+ channels that decrease intracellular K^+ and causing electrical instability in atria and ventricles (Singer and Rotenberg, 1973).

5. Risk management of lithium in contaminated environments

The lack of a complete Li recycling system requires disposal of Li-containing materials, such that Li can leach into the environment and present a risk to humans and ecosystems (Henschel et al., 2020; Tanveer et al., 2019). Developing cost-effective and eco-friendly remediation technologies for the sustainable management of Li-contaminated environments is necessary.

5.1. *Remediation of lithium-contaminated soils and sediments*

Various technologies, including physical, chemical, and biological strategies, are currently accessible for controlling metal pollution in the soil environment, which can be theoretically applied to sediment (Bolan et al., 2014). Wuana and Okieimen (2011) categorized the general practices used in metal remediation: isolation, extraction, and reduction in toxicity and mobility. The operation of these strategies can either be *in situ* (takes place at the contaminated sites without moving or excavating soil) or *ex-situ* (soil is moved or excavated from the contaminated site).

As Li is highly mobile in soil and sediment, reducing the mobility and toxicity of Li by applying various amendments and followed by phytoremediation, which is considered a cost-effective and eco-friendly remediation process, is an effective approach. Shahzad et al. (2016) proposed the following Li-remediation technologies: (1) application of soil amendments to immobilize Li in contaminated soil (Table 5). For example, applying zeolite to Li-contaminated soil significantly reduced plant uptake over 14 days (Harbottle et al., 2007), and adding 33% sphagnum peat moss and 33% coal fly ash to Li-contaminated soil significantly reduced Li in the soil leachate (Bilski et al., 2013). Amorphous and nano-size materials provide a large surface area, which can contribute to the increased sorption of Li in soil (Abbas et al., 2021); (2) identification of plants and microorganisms that can accumulate and tolerate Li. For instance, *Apocynum venetum* (sword-leaf dogbane) and *Apocynum pictum* (Indian hemp or dogbane) show high translocation and bioaccumulation of Li (Jiang et al., 2014; Jiang et al., 2018) (Table 6). The underlying mechanism of plant uptake and translocation of Li in soil remains unclear; however, it is suggested that Li can be absorbed by plants in a similar way to other alkali metals (e.g., Na and K) due to functional analogues (Shkolnik, 1984). A few microbial strains, including *Arthrobacter nicotianae* and *Brevibacterium helovolum*, also have the potential to accumulate Li. These microbial strains can exhibit a strong negative charge on their cell surface owing to high level of ionized phosphate groups, to enhance chelate formation between the cell surface and Li ions (Tsuruta, 2005); (3) application of a chelating agent to Li-contaminated soil to alter Li toxicity and bioavailability (Table 5). For example, ethylenediaminetetraacetic acid (EDTA) and ethylenediamine-N, N′-di succinic acid (EDDS) were used as Li chelators, which increased bioavailable Li for plant uptake, facilitating the phytoremediation of Li- contaminated soils (Kavanagh et al., 2018). The addition of chelating agents can enhance Li-chelant complexes, which are directly absorbed by the plant roots. Moreover, the dissociation of the complexes releases freely available Li for plant uptake (Brian et al., 2018). Several factors affecting Li remediation efficiency include environmental conditions (e.g., temperature, precipitation), soil properties (e.g., texture, pH), and chemical composition of Li in soils and sediments (Shahzad et al., 2016).

Table 5
Selected references on the (im)mobilization of Li in soil.

Treatments	Experimental conditions	Main findings	References
Zeolite amendment	Soil: Zeolite = 4: 1 Temperature: 21 °C Humidity: 45% Spiked Li: 1.56 and 7.8 g	Inhibited Li uptake in poplar plants due to increased adsorption of the sorbent.	Harbottle et al. (2007)
Raw white eggshells and acetic acid-treated white eggshells	pH: 4 Incubation: 7 d Temperature: 45 °C Dosage: 3 g	40.73% Li adsorbed with the addition of raw white eggshells 85.38% Li adsorbed after adding crushed white eggshells pre-treated with acetic acid	Abbas et al. (2021)
Sphagnum peat moss and	Optimum composition: soil: fly ash: moss = 33: 33: 33%	Reduced Li concentration in leachate, relative to the control	Bilski et al. (2013)
Ethylenediaminetetraacetic acid (EDTA) and ethylenediamine-N,N′-disuccinic acid (EDDS)	Chelator concentration: 0.05 M, 100 mL Temperature: 24 °C Plant growth periods: 7 d	Increased Li availability in soil, which increased plant uptake More than 4 g kg^{-1} Li accumulated in some plants	Kavanagh et al. (2018)

Table 6
Selected references on the phyto-availability of Li in soil.

Plant species	Li speciation and concentrations	Main findings	References
Apocynum pictum (dogbane)	LiCl 50, 200, and 400 mg kg^{-1}	Li accumulated at higher concentration in leaves than stems and roots. Plants survived under highest Li concentration and accumulated >1800 mg kg^{-1} Li in leaves	Jiang et al. (2018)
Beta vulgaris L. (beetroot) *Lactuca sativa* L. (lettuce) *Brassica nigra* L. (black mustard) *Lolium perene* L. (perennial ryegrass) *Helianthus annuus* L. (sunflower)	LiCl 1, 3, 10, 30, and 100 mg kg^{-1}	Leaf Li all the tested plants exceeded 1000 mg kg^{-1} at low Li content in the soil, indicating high bioaccumulation.	Robinson et al. (2018)
Brassica juncea (mustard)	LiCl 380 mg kg^{-1}	23% Li accumulated in leaves; 70% Li stabilized in the rhizosphere	Elektorowicz and Keropian (2015)
Apocynum venetum (sword-leaf dogbane)	LiCl 50, 200, and 400 mg kg^{-1}	No obvious phytotoxicity symptoms at 50 mg kg^{-1} Li Plants accumulated >1800 mg kg^{-1} Li in leaves Plants survived at 400 mg kg^{-1} Li	Jiang et al. (2014)
Brassica napus (rape) *Brassica oleracea* var. *capitate* (cabbage) *Helianthus annuus* (sunflower) *Solanum lycopersicum* (tomato) *Cardamine hirsute* (hairy bittercress)	LiCl 20, 50, 100, 150, 200, 300, 400, 500 and 1000 mg kg^{-1}	All species could accumulate Li *B. oleracea* var. *capitate, H. annuus,* and *C. hirsute* had higher accumulation capacity than the other two plant species (>1496 mg kg^{-1} in *B. oleracea* var. *capitate* and *H. annuus*)	Kavanagh et al. (2018)
Spinacia oleracea (spinach)	LiCl 20, 40, 60, and 80 mg kg^{-1}	No visual toxicity symptoms at any Li level Li mainly accumulated mainly in leaves	Bakhat et al. (2019)

5.2. *Removal of lithium from aquatic environments*

Among the available Li removal techniques for aquatic environments, recent attention has been given to various precipitation, adsorption, electro-dialysis, and ion exchanging approaches (Çiçek et al., 2018; Parsa et al., 2015; Zhao et al., 2020). The precipitation of Li with suitable anions is effective for removing Li from Li-containing wastewaters. Zhao et al. (2020) used a precipitation method to recover Li^+ in the presence of common cations, such as Ca^{2+}, Mg^{2+},

Al^{3+}, and Fe^{3+}, from low concentration, Li-containing wastewater. Under optimized conditions, they recovered 84.2% of Li^+ as a Li_3PO_4, with simultaneous removal of 99–99.8% of other associated cations (Zhao et al., 2020). Similarly, an environmentally friendly Li precipitation method successfully recovered Li from wastewater (Jandova et al., 2012), where CO_2 was used to precipitate Li as Li_2CO_3, with an efficiency of about 90%.

Lithium-selective resins have been used to successfully remove Li from water. For instance, an aminomethyl phosphonic acid-containing chelating resin was used as a cation exchanger under different optimized conditions, removing 13.65 mg g^{-1} of Li within 15 min (Çiçek et al., 2018). A novel magnetic Li imprinted-polymer is selective for the Li-ion, removing 4.10 mg g^{-1}, the maximum adsorption affinity, within 10 min at pH 6 (Luo et al., 2015). Zandevakili et al. (2014) used MnO_2 nanorod as a Li sieve to remove 63 mg g^{-1} Li from lake water. Similarly, a three-dimensional MnO_2 ion cage was successfully fabricated by Luo et al. (2016) to treat Li-contaminated wastewater, which selectively targeted Li ions in wastewater with an adsorption affinity of 57 mg g^{-1} Li. Kim et al. (2018) used an electrochemical technique to recover about 98.6% of Li ions (containing 1900 mg L^{-1} Li) from wastewater using a Li recovery electrode of Li Mn-oxide. It was also estimated that about 24 mg Li was recovered in each hour of operation (Kim et al., 2018).

5.3. Integrated risk management

In integrated risk strategies, two or more individual remediation techniques are used to improve Li removal i.e., chemical–biological, chemical–physical, biological–physical, or chemical–biological–physical. Integrated risk management ensures an effective and efficient achievement than individual approaches. For example, the addition of EDTA and EDDS, as chelating agents, to Li-containing soil enhanced the accumulation of Li in leaves of *Brassica napus* (rapeseed), *Brassica oleracea* (wild cabbage), and *Helianthus annuus* (sunflower) (Kavanagh et al., 2018). Similarly, under optimum conditions, Abbas et al. (2021) investigated the effect of raw, untreated-eggshells and acetic acid-treated eggshells as soil amendments to remove Li. The acetic acid-treated eggshells increased Li removal from 45.7 to 85.3% in the Li-contaminated soil, which was attributed to the reduction in soil pH induced by acetic acid (Abbas et al., 2021). Thus, mobilization of Li using soil amendments, followed by phytoremediation, is an integrated and effective approach for the remediation of Li-contaminated soils and sediments.

6. Final remarks and future research needs

Lithium is a potentially toxic element that adversely affects human health and ecosystems. Terrestrial and aquatic environments contaminated with Li from various (geogenic and anthropogenic) origins have been drawing public concern since last several years in many countries. The biogeochemistry of Li in soil is largely controlled by its interactions with the clay mineral components. Lithium in most soils is readily released into ground water due to its high mobility. Although the accumulation of Li in biota (plants and organisms) has been reported in various studies, biochemical transformations of Li in living organisms remain largely unknown.

Risk management of lithium contamination of terrestrial and aquatic environments is a great challenge, but essential, to protect and alleviate adverse impacts on ecosystem and public health. Remediation of these contaminated environments has been carried out using various physical, chemical, and biological approaches (e.g., precipitation, biosorption). Despite some remarkable positive results, most of these remedial technologies have been conducted under controlling conditions and pilot scales. Further research in field scale application of such technologies is therefore warranted to attract attention from the remediation industry or policy makers. Nevertheless, these remedial treatment actions require trained personnel for the operation of equipment to treat the contaminated soils and waters. Phytoremediation, which is cost-effective and environmentally friendly, could be a suitable alternative for the remediation of Li-contaminated environments.

Some key knowledge gaps, which require future research, include:

- Understanding biogeochemical processes regulating dynamics of Li in different media using advanced techniques (e.g., spectroscopic based).
- Elucidation of various environmental factors (e.g., pH) affecting the chemical and biological transformations of Li in soil and aquatic systems.
- Investigation of Li speciation in both solid- and liquid-phase in soil and water environments.
- Examination of biochemical mechanisms involved in Li accumulation in specific tissues or organs in living organisms.
- Evaluation of plants' response and tolerance to Li toxicity under field conditions.
- Determination of the underlying mechanisms in the rhizosphere underpinning effective phytoremediation of Li contamination.
- Exploitation of the effectiveness of industrial by-products containing high metallic oxide content to immobilize Li in the contaminated sites; examination of the desorption of sorbed Li from the immobilized media.
- Development of bio-monitors for risk assessment of Li-contaminated environments.

Credit author statement

Nanthi Bolan: Research idea, initiated and coordinated the review, provided the skeleton and overall contents, Contributed to Section 1 and 6, writing and editing. **Mohsin Tanveer and Lei Wang:** Contributed to section 4.1 and in compiling the associated Figures/tables, writing and editing. Son A. **Hoang and Shiv Bolan:** Contributed to Section 2.1 and in compiling the associated Figures/tables, writing and editing. **Prasanthi Sooriyakumar, Brett Robinson:** Contributed to section 2.2 and in compiling the associated Figures/tables, writing and editing. **Hasintha Wijesekara and Madhuni Wijesooriya:** Contributed to section 3.1 and in compiling the associated Figures/tables, writing and editing. **S. Keerthanan and Meththika Vithanage:** Contributed to section 3.2 and in compiling the associated Figures/tables, writing and editing. **Bernd Markert, Stefan Fränzle, and Simone Wünschmann:** Have written Section 4.2 and in compiling the associated Figures/tables, writing and editing. **Binoy Sarkar, Ajayan Vinu and Kadambot H.M Siddique:** Contributed to Section 5 and in compiling the associated Figures/tables, writing and editing. **M.B. Kirkhamn, Jörg Rinklebe:** Contributed to research concept, writing and editing, proof reading.

Declaration of competing interest

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

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