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Lithium as an emerging environmental contaminant: Mobility in the soil-plant system

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HIGHLIGHTS

- Geogenic Li is sparingly soluble and mostly unavailable to plants.
- Ionic Li added to soil binds weakly and is more mobile under acidic conditions.
- Plants take up ionic Li with a bioaccumulation coefficient >5.
- Plants accumulate up to 500 mg/kg Li in the water sinks with few toxicity symptoms.
- In contaminated environments, plants are likely to be an important exposure pathway for Li to enter humans or ecosystems.

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ABSTRACT

Contamination of soil with lithium (Li) is likely to increase due to its wider dispersal in the environment, associated in particular, with the disposal of the now ubiquitous Li-ion batteries. There is, however, a paucity of information on the behaviour of Li in the soil-plant system. We measured the sorption of added Li to soil, and uptake of Li by food and fodder species. Around New Zealand, soil concentrations were shown to range from 0.08 mg/kg to 92 mg/kg, and to be positively correlated with clay content. Most geogenic Li in soil is insoluble and hence unavailable to plants but, when Li^+ is added to soil, there is only limited sorption of Li. We found that Li sorption increased with increasing soil pH, and decreased proportionately with increasing Li concentrations. Compared to other cations in soil, Li is mobile and may leach into receiving waters, be taken up by plants, or have other biological impacts. In a soil spiked with just 5 mg/kg, plants took up several hundred mg/kg Li into leaves with no reduction in biomass. Lithium appears to be a phloem immobile element, with the highest concentrations occurring in the older leaves and the lowest concentrations occurring in the seeds or fruits. These results may raise concerns and risks in situations where food and fodder crops are associated with waste disposal.

1. Introduction

Lithium (Li) is a non-essential, but occasionally beneficial element for organisms (Schafer, 2012). Lithium has a relatively low toxicity for animals, with a lethal dose of >500 mg/kg bodyweight for rats (Aral and Vecchio-Sadus, 2008a). Relatively low concentrations affect the central nervous system, with reports that drinking water concentrations >0.17 mg/L reduce the incidence of suicide (Schrauzer and Shrestha, 1990) and also that concentrations >0.015 mg/L are protective against dementia (Kessing et al., 2017). Of course, Li is used in medications for bipolar disorder and manic and depressive episodes, and Goldstein and Mascitelli (2016) hypothesised that increased Li intake would reduce levels of aggression and violence.

Humans may be exposed to increasing Li concentrations as it enters the environment through the use and disposal of Li-containing products. Global Li production has tripled since 2000, with current production >600,000 tonnes (Kelly and Matos, 2013). This increase is largely driven by the pervasion of Li-ion batteries in electronic goods and increasingly, in vehicles (Mohr et al., 2012). Lithium is also used in greases, ceramics (Hao et al., 2017), mood-stabilising drugs (Aral and Vecchio-Sadus, 2008a), and sometimes

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in the illicit manufacture of methamphetamine (Person et al., 2005).

Drinking water and plants are likely to provide the primary exposure pathways for Li into the food chain (Franzaring et al., 2016); both are dependent on the dynamics of Li in soil. Background concentrations in soil range from <1 to 200 mg/kg (Schrauzer, 2002a; Aral and Vecchio-Sadus, 2008b), with higher concentrations occurring in arid and saline soils (Merian, 1991). Lithium generally enters the soil solution primarily through weathering of sedimentary minerals (Chan et al., 1997; Aral and Vecchio-Sadus, 2008a). Topsoil usually contains less Li than underlying layers (Merian, 1991). The clay fraction of soil contains higher Li concentrations than the organic soil fraction (Schrauzer, 2002b), with the Li present in ditrigonal cavities within clay minerals (Anderson et al., 1988).

Dissolved Li ions have a charge/hydrated radius quotient just 2.9 e/nm which is low compared to other common ions in soil solution Na^+, K^+, Mg^{2+} and Ca^{2+}, which have charge/hydrated radius quotients of 3.6, 4.3, 4.3, and 6.2 e/nm respectively (Wiberg et al., 2001). This indicates that the binding of Li via outer sphere electrostatic attraction is likely to be weak (Sposito, 2016). Similarly, it is unlikely that there will be significant covalent inner-sphere binding due to the large energy difference between the bonding orbitals on Li^+ and those on the functional groups present on soil colloids. We therefore hypothesise that Li^+ that is added to soil will be immobilised less readily and will be relatively mobile compared to other trace elements, with a higher potential to leach into receiving waters.

Citing numerous sources, Gough et al. (1979) reported a wide variation in plant tolerance to Li; citrus was found to be particularly sensitive, whilst cotton was more tolerant. In hydroponic conditions, Kalinowska et al. (2013) demonstrated that lettuce was tolerant to 20 mg/kg and accumulated Li in leaves at concentrations tenfold higher than the nutrient solution. Franzaring et al. (2016) reported that soil concentrations of Li spiked to 118 and 47 mg/kg were toxic to maize and buckwheat respectively and that leaf concentrations in maize were tenfold higher than soil concentrations. Jiang et al. (2014) reported that a traditional Chinese medicine herb, *Apocynum venetum*, accumulated >1800 mg/kg in leaf tissue, and postulated that such high Li concentrations may confer some of the known antidepressant and anxiolytic effects of this plant.

We hypothesised that ionic Li added to soil would be poorly adsorbed by the soil and readily accumulated by plants. We aimed to determine the sorption of Li by an agricultural silt loam, the tolerance and accumulation of Li in selected food and fodder crops, comparing these with Li concentrations in soil and ryegrass from a survey of 69 sites throughout New Zealand. We used sunflower grown in soils spiked with Li, to evaluate the partitioning of Li in plants.

2. Methods and materials

Batch and greenhouse experiments used the Templeton Silt Loam (TSL), a typic immature pallic soil, collected from the Lincoln University dairy farm (43°38′11.39″S, 172°26′19.09″E). Properties of the TSL used in these experiments are provided in Table 1.

Table 1
Properties of templeton silt loam from the Lincoln University dairy farm.

	Total[a]	Extractable[b]
pH	5.6	n/a
C (%)	2.0	n.d
N (%)	0.18	n.d
CEC (cmol kg^{-1})	12.5	n/a
Li (mg kg^{-1})	31.8	0.13
Mg (mg kg^{-1})	855	16.6
K (mg kg^{-1})	1401	14.0
Na (mg kg^{-1})	136	44.9
Ca (mg kg^{-1})	3005	n.d.
P (mg kg^{-1})	518	0.71
S (mg kg^{-1})	193	8.5

[a] Pseudototal using a conc. HNO_3 extract.
[b] Extracted in a 0.05 M $Ca(NO_3)_2$ solution.

We used a batch experiment to determine the influence of pH and spiked Li solution concentration on Li sorption by the TSL. Soil was first dried and passed through a 2 mm Nylon sieve. The sorption experiment was conducted using 40 ml centrifuge tubes with 4 replicates per treatment. Three replicates per treatment were used for element analysis with duplicate pH determinations. TSL (5 g) was added to 30 mL of 0.05 M $Ca(NO_3)_2$ (BDH AnalaR $Ca(NO_3)_2 \cdot 4H_2O$) solution spiked with varying amounts of Li (LiCl, Sharlau). The initial solution Li concentrations were 0, 1, 3, 10, and 30 mg/L. The solution pH was adjusted to values ranging of 2.8, 4.3, 5.2, 6.5 & 7.4 after addition of the soil by using HNO_3 (BDH ARISTAR nitric acid 70%) or KOH (BDH AnalaR KOH) to decrease or increase pH, respectively. Centrifuge tubes containing solutions without soil were prepared as a control for each Li concentration to determine the extent of any Li sorption by the tubes or filter papers; this was found to be negligible. The tubes were continuously agitated (2 h) on an end-over-end shaker in order to reach sorption equilibrium (determined in a previous experiment). The pH of the slurry was measured after shaking. The tubes were centrifuged for 10 min at 3300 rpm. The supernatant was decanted and filtered (Whatman 52, pore size 7 µm). Solution element concentrations were determined using ICP-OES (Varian 720 ES).

The soil was amended with Li (LiCl, Sharlau) to provide concentrations of 0 (control), 1, 3, 10, 30 & 100 mg/kg. Lithium was added to soils using a thoroughly cleaned concrete mixer to which 250 mL of solution containing the appropriate Li concentration was applied. Pots (2.5 L) were planted with *Beta vulgaris* L. (beetroot), *Lactuca sativa* L. (lettuce), *Brassica nigra* L. (black mustard), and *Lolium perenne* L. (perennial ryegrass), with five replicates per treatment. Larger pots (7.5 L) were prepared for *Helianthus annuus* L. (sunflower) spiked with Li at 0, 10 and 30 mg/kg (three replicates per treatment). Plants were sown with three seeds per pot and later thinned out to one seedling, with the exception of *L. perenne*, which was established by sowing 50 seeds per pot. Pots were arranged in a fully randomised single block design on a glasshouse bench.

After three months growth, when the edible portions of the plants were mature, they were harvested using scissors ca. 1 cm from the soil surface. The harvested plants were thoroughly washed in deionised water and transferred into pre-labelled paper bags and oven dried for at 105 °C to constant weight. The dry plant samples were ground into a fine powder and stored in labelled plastic bags.

The *B. vulgaris* leaves and bulbs were harvested, leaves were washed & dried and the bulbs were peeled, washed and dried separately. The *H. annuus* plants were divided into six samples: Leaves were divided into equal thirds by their height on the stem. Henceforth, these are referred to as bottom leaves, middle leaves and top leaves. The shoots, flower and roots were analysed separately.

Soil and pasture samples from 69 sites had been collected in an earlier study at sampling locations across New Zealand (Reiser et al., 2014). Pasture samples (ca. 0.2 m^2) were cut 2–3 cm above ground and dicotyledonous weeds removed. These samples were predominantly perennial ryegrass, *Lolium perenne*, which comprise some 70% of NZ pastures (Roberts and Longhurst, 2002). From each sampling location, a 10 cm soil corer was used to obtain a bulked sample from 10 to 15 composite cores.

Plant samples were washed in deionized water and dried at 60 °C until a constant weight was obtained. Subsequently, they were milled using a Cyclotech type 1093 cyclone grinder with an aluminium rotor. Plant material (0.5 g) was digested in 5 ml HNO_3. The digests were diluted with Milli Q (Barnstead, EASYpure RF, 18.3 MΩ-cm) to a volume of 25 ml and filtered with a Whatman 52 filter paper (pore size 7 μm). Soil samples were dried for 7 d at room temperature, the aggregates separated, and then passed through a 2 mm nylon sieve. Soil sub-samples (0.5 g) were microwave digested in 5 ml aqua regia (BDH Aristar HNO_3 69%, BDH Aristar HCl 37%).

Pseudo-total element concentrations (henceforth referred to as “total”) were determined in the acid digests using ICP-OES (Varian 720 ES). Wageningen (ISE 921, IPE 100) and NIST (1573a) QA reference materials were analyzed in the same sample sets. Recoveries ranged from 91 to 112%. The C and N content of soil and plant material was determined using an Elementar Vario MAX CN element analyzer.

$Ca(NO_3)_2$-extractable elemental concentrations in soil were determined using ICP-OES. Soil (5 g) was weighed into centrifuge tubes ($V = 40$ ml) and 30 ml 0.05 M $Ca(NO_3)_2$ (BDH AnalaR $Ca(NO_3)_2 \times 4H_2O$) was added. Tubes were agitated for 2 h on an end-over-end shaker, subsequently centrifuged for 10 min at 15′000 rpm and then filtered using a Whatman 52 filter paper. All extracts were stored at 4 °C before the analysis. Soil pH was measured in Milli Q at a solid: water ratio of 1:2.5 using a Metler Toledo pH meter. The grain size distribution of the soils was determined using the pipette method according to the reference methods of the Swiss Federal Agricultural Research Stations (FAL et al., 1996).

Minitab® 16 was used for ANOVA with Fisher's Least-Significant-Difference post-hoc test to compare means.

3. Results & discussion

3.1. Lithium in soils

Lithium concentrations from the 69 pastoral soils from around NZ are shown in Table 2, with comparable silt loam values to the Dairy Farm TSL soil (Table 1). It has been suggested that soil Li concentrations in the low mg/kg range may indicate contamination from small amounts of Li-grease which can contain >1% Li (Zeng and Li, 2014), presumably from agricultural machinery.

Soil Li was strongly positively correlated with the clay fraction (Table 3), which is consistent with the findings of Schrauzer (2002a). Lithium may substitute for Mg in silicate matrices due to their similar ionic radii of 90 p.m. and 86 p.m. respectively. There was a strong positive correlation between Li and Mg as well as between Li and Al, B, Fe, K, Mn and Zn.

The K_D results (Fig. 1) show that, as with other trace element cations, sorption increases exponentially with increasing pH and proportionately decreases with increasing Li concentrations. The magnitude of the sorption is substantially lower than other trace element cations, where K_D values generally range from <1 to 7. This indicates that Li ions that are added to soils are likely to be mobile relative to other trace elements and therefore more likely to be mobilised, to leach through soil or to interact with soil organisms and plant roots. This is consistent with observations elsewhere that monovalent cations with a large hydrated radius are weakly sorbed via outer-sphere binding (Sposito, 2016). Similarly, inner-sphere binding, if it occurs at all, is likely to be weak due to the low polarizability of the Li ion and large energy difference between the bonding orbitals of Li and those of potential ligands in soil. The high mobility added Li^+ in the TSL contrasts sharply with the low mobility of the geogenic Li, which has a K_D of 245. These results further support the hypothesis that geogenic Li is associated with silicate matrices or in the interlayers of layer silicates and thus only exposed to soil solution during the weathering processes of these minerals.

Table 2
Mean [Li] (mg kg^{-1}) dry weight for NZ pasture soils. Values in brackets are the standard error of the mean. Values with the same letter are not significantly different at the 5% level.

Texture	N	Soil Li	N	*L.perenne* Li
Silty clay loam	7	48.8 (11.1)^a	5	1.4 (0.6)^a
Clay loam	15	24.9 (4.3)^b	6	1.2 (0.2)^a
Silt loam	9	24.3 (4.4)$^{b\ c}$	6	1.8 (0.7)^a
Loam	17	21.6 (2.7)$^{b\ c}$	9	1.8 (0.6)^a
Sandy loam	13	11.7 (2.6)^c	7	1.7 (0.6)^a
Sandy clay loam	4	10.9 (3.2)$^{b\ c}$	2	0.9 (0.6)^a

Table 3
Correlation matrices between soil Li Vs soil texture, soil Li Vs Li pasture and soil Li Vs other elements.

Elements	Soil Li and soil properties	Pasture grass Li and soil properties	Pasture grass Li and other elements in pasture grass
Sand	−0.53 (S**)	0.05 (NS)	
Silt	0.36 (S)	0.03 (NS)	
Clay	0.43 (S**)	−0.14 (NS)	
pH	0.06 (NS)	−0.04 (NS)	
Al	0.59 (S**)	−0.10 (NS)	0.90 (S**)
B	0.65 (S**)	0.17 (NS)	−0.02 (NS)
Ca	0.12 (NS)	0.16 (NS)	−0.16 (NS)
Cu	0.13 (NS)	0.04 (NS)	−0.002 (NS)
Fe	0.56 (S**)	0.09 (NS)	0.90 (S**)
K	0.56 (S**)	0.20 (NS)	−0.14 (NS)
Mg	0.68 (S**)	−0.23 (S)	−0.23 (NS)
Mn	0.45 (S**)	0.07 (NS)	0.06 (NS)
Mo	0.15 (NS)	0.21 (NS)	0.21 (NS)
P	0.08 (NS)	−0.07 (NS)	−0.07(NS)
S	−0.22 (S)	0.08 (NS)	0.08 (NS)
Zn	0.60 (S**)	−0.15 (NS)	−0.15 (NS)

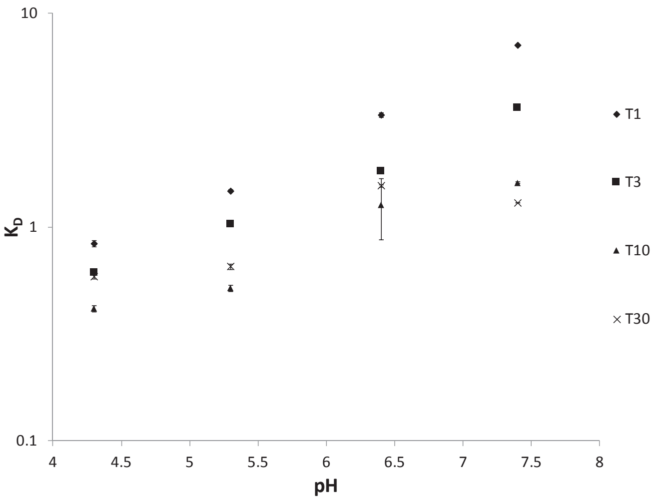

Fig. 1. K_D as a function of pH at varying Li concentrations. T1 – T30 indicate the initial solution concentrations in mg/L. Error bars represent the standard error of the mean ($n = 3$). The background Li concentration in the soil was 25 (mg/kg).

3.2. Lithium in plants

Lithium concentrations in pasture grasses from the 69 sites (Table 2) indicate that geogenic soil Li was mostly unavailable for plant uptake as the average bioaccumulation coefficient (plant/soil concentration quotient) was only 0.14. Pasture grass Li was strongly correlated with plant Al and Fe (Table 3), which is consistent with the plant Li arising from dust particles incorporated into the cuticular wax of plant leaves (Robinson et al., 2008). There were no significant correlations between plant Li and any soil property except for a significant but weak negative correlation with soil Mg.

When Li was added to soil in the pot experiment, there was significant plant uptake (Fig. 2A), with Li concentrations in the leaves of all plant species exceeding 1000 mg/kg (dry weight) at $Ca(NO_3)_2$-extractable concentrations of just 5 mg/kg Li in soil, representing a bioaccumulation coefficient of >20. There was no significant biomass reduction even though these plants accumulated 500–1000 mg/kg Li in their leaf tissue (Fig. 2b). Tissue concentrations above 1000 mg/kg resulted in reduced biomass production and necrosis on the older leaves, consistent with the findings of Kalinowska et al. (2013) for lettuce plants. These results indicate that the phytotoxicity of Li is low when compared to other non-essential trace elements. This also indicates that food chain protection is not provided by Li toxicity as was suggested by Chaney (1980).

The effect of increasing plant Li concentration had species-specific effects on the uptake of other elements (Table 4). In *L. perenne*, leaf Li concentration was negatively correlated with B, Mg, Mn, & Mo and positively correlated with Ca, Fe, K, & Zn. This differed in *L. sativa* and *B. vulgaris*, which, for example, had positive correlations between Li and Mg. It is likely that elevated Li concentrations change the function of plant transporters of essential elements, but we have insufficient information to speculate on the nature of these changes.

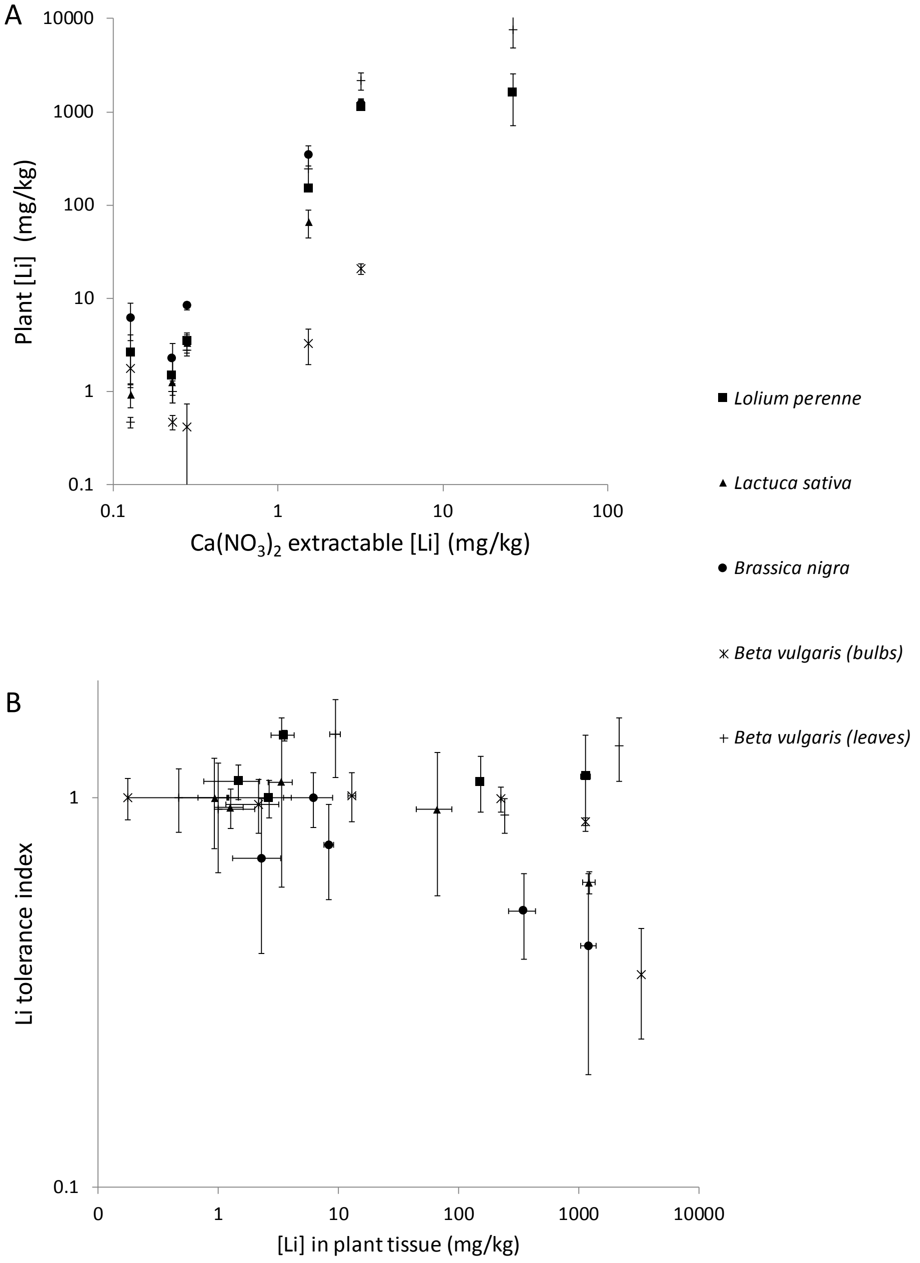

Fig. 2. (A) Plant uptake of Li as a function of extractable Li in soil and (B) Li tolerance index (treatment/control biomass quotient) as a function of Li in plant tissue. Bars represent the standard error of the mean (n = 5).

Table 4
Correlation matrix of Li in plants versus essential elements in plants.

Element	*L. perenne*	*L. sativa*	*B. vulgaris*, leaves	*B. vulgaris*, bulbs
B	−0.84 (S**)	−0.68 (S**)	0.06 (NS)	0.31 (NS)
Ca	0.58 (S**)	0.19 (NS)	0.31 (NS)	0.09 (NS)
Cu	0.07 (NS)	0.72 (S**)	0.21 (NS)	−0.37 (NS)
Fe	0.49 (S*)	0.32 (NS)	0.30 (NS)	−0.15 (NS)
K	0.52 (S*)	0.32 (NS)	−0.66 (S**)	0.26 (NS)
Mg	−0.78 (S**)	0.48 (S)	0.69 (S**)	0.28 (NS)
Mn	−0.64 (S**)	0.02 (NS)	0.54 (S*)	0.47 (S)
Mo	−0.80 (S**)	0.00 (NS)	0.10 (NS)	0.16 (NS)
P	−0.19 (NS)	0.52 (S*)	−0.25 (NS)	−0.44 (S)
S	0.14 (NS)	0.27 (NS)	0.19 (NS)	0.02 (NS)
Zn	0.44 (S)	0.71 (S**)	0.72 (S**)	0.72 (S**)

The bulbs of *B. vulgaris* had Li concentrations that were tenfold lower than the leaves (Fig 2a), which indicates that Li may be primarily translocated in the xylem and accumulates in the leaves. This was tested by growing *Helianthus annuus* in Li-spiked soil and measuring Li concentrations in the major plant organs (Fig. 3). The highest Li concentrations occurred in the bottom leaves of the plant, with the shoots, roots and flowers having lower concentrations. This pattern of accumulation is consistent with an element that is primarily transported in the xylem and with limited phloem mobility (Evangelou et al., 2016). The aboveground portions of the plants grown in soil spiked with Li at 10 and 30 mg/kg accumulated high Li concentrations of 27 and 756 mg/kg respectively. The biomass production of the *H. annuus* grown in the lower soil concentration was not significantly different from the control (data not shown), indicating that Li was not phytotoxic at these concentrations but, at the higher soil concentration, the plants exhibited necrosis on the lower leaves. The soil Li concentration that caused toxicity in this study (30 mg/kg) was similar to the findings of Hawrylak-Nowak et al. (2012), who reported a significant reduction of the biomass of *H. annuus* growing in nutrient solution containing 50 mg/L Li. These authors reported that the growth and concentrations of chlorophyll, carotenoids, and malondialdehyde were unaffected in *H. annuus* growing in solutions containing 25 mg/L Li despite the leaves containing >400 mg/kg Li.

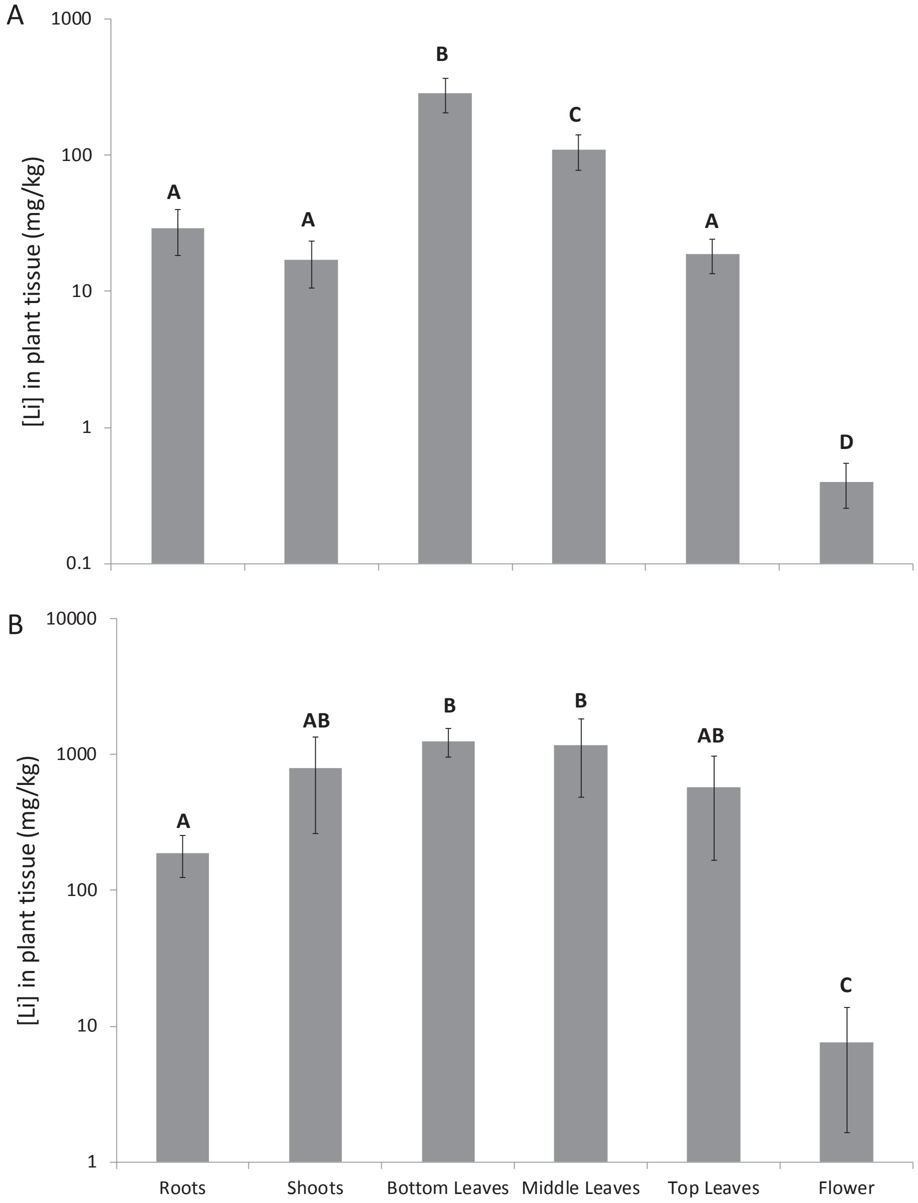

Fig. 3. Li concentrations in different parts of *H. annuus* growing in soil spiked with 10 mg kg^{-1} (A) and 30 mg kg^{-1} (B). Bars represent the standard errors of the means (n = 3).

3.3. *Potential health risks*

Our results indicate that soil contamination with Li may result in plant Li concentrations that are increased by several orders of magnitude, with leaf crops likely providing the highest source of Li in the diet. Even with these increases, it is unlikely that humans would experience acute Li toxicity, which occurs when blood serum reaches 15 mg/L (Aral and Vecchio-Sadus, 2008a). Assuming that ingested Li is distributed evenly in the aqueous phase of the body, a 70 kg individual (assuming 60% water content) would need to consume 630 g of plant material (dry matter) with a Li concentration of 1000 mg/kg Li to reach toxicity thresholds. Nevertheless, the chronic ingestion of such plant material may have measurable effects on humans in the context of the known medicinal effects of this element, and as has been observed in patients taking Li-based antidepressant medication over extended periods (Aral and Vecchio-Sadus, 2008a).

4. Conclusions

The low toxicity of Li has probably resulted in this element being largely overlooked as an emerging environmental contaminant. The pervasion of Li-based batteries into society will likely result in greatly increased incidences of soil contamination. Lithium is mobile in the soil-plant system and therefore soil contamination with Li may result in rapid and extreme increases in Li concentrations in groundwater and in food crops. While increases in Li consumption may be beneficial in some cases, the effect on society of prolonged exposure to elevated Li concentrations is unknown. Being relatively phloem-immobile, leafy vegetables will likely provide the largest source of Li in foods. Further work is warranted to elucidate the transfer of Li from plants to animals and humans.

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