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The potential water quality impacts of hard-rock lithium mining: Insights from a legacy pegmatite mine in North Carolina, USA

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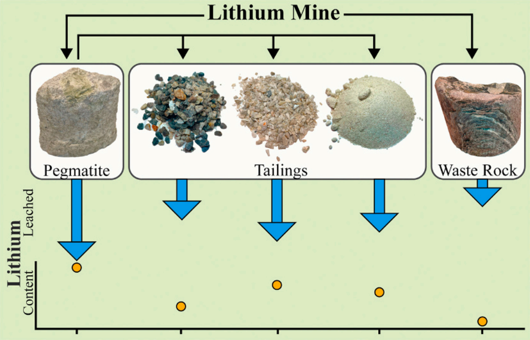
HIGHLIGHTS

- Water quality of waters near lithium mine show low regulated contaminant concentrations but elevated Li, Rb, and Cs
- Analysis of Li ore and waste solids reveals low concentrations and leachability of contaminants.
- Leaching experiments show alkaline conditions induced by interactions with phosphate and spodumene minerals.
- Phosphate and spodumene impact on water quality is a short-term outcome of hard-rock lithium mining.
- Prevalence of Li, Rb, and Cs in natural waters and leaching experiments highlights their importance.

GRAPHICAL ABSTRACT



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ABSTRACT

The global green energy transition has spurred increased lithium exploration and extraction, yet the water quality impacts from lithium mining are understudied. This study investigates the potential water quality impacts from a legacy hard-rock lithium mine through comprehensive geochemical analyses of groundwater, surface waters, ore grade rocks, tailings, and waste rocks from a mine site in North Carolina, USA. The concentrations of regulated contaminants (e.g. As, Pb) in both groundwater and surface water emerging from the mine site were low, below drinking water and ecological standards. Yet Li (up to 46.8 mg/L), Rb (up to 169 μg/L), and Cs (up to 21 μg/L) were elevated relative to local background waters. Leaching experiments of the pegmatite ores, waste rocks, and tailing consistently demonstrate low mobilization of regulated contaminants and high leachability of Li, Rb, and Cs. Leaching experiments also reveal that water-rock interactions of the rocks and solid wastes from the mine site generate alkaline conditions, and that both phosphate and spodumene minerals are primary sources of Li and play a major role in formation of alkaline conditions during early stages of water-rock interactions. Over longer time scales, their direct impact on water quality is decreased. Given the

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global interest in hard-rock lithium mines, our findings highlight the potential occurrence of Li, Rb, and Cs in water resources adjacent to hard-rock lithium mines.

1. Introduction

The clean energy transition from fossil fuels-based energy has spurred international exploration and increased extraction of critical raw materials (CRMs) to be used in the development of clean energy technologies. Lithium, in particular, is expected to have the largest increase in demand of any CRM due to this transition, with primarily uses in Li-ion batteries for electric vehicles and grid storage (IEA, 2021). Globally, Li is produced from two major resource types, Li-rich brines and Li-rich pegmatites, the latter accounts for >50 % of global production (Bradley et al., 2017b; Jaskula, 2023). Although Li brines are typically larger resources, Li pegmatite occurrences are more common around the world, with plans in numerous countries to exploit these deposits (Fig. 1A) (Bradley et al., 2017b; Kesler et al., 2012). Hard-rock Li deposits are typically found in Lithium-Cesium-Tantalum (LCT) pegmatites, so called for their characteristic enrichments of these metals (Bradley et al., 2017a; London, 2008). The heightened demands have led to exploration of new and unused Li deposits, including in the Tin Spodumene Belt (TSB) of North Carolina (NC), USA (Fig. 1B).

Globally, the environmental impacts of Li mining have been relatively understudied, while in many cases CRM mining has been shown to have extensive environmental impacts. This is becoming a topic of major debate for the green energy transition (Asif and Chen, 2016; Hao et al., 2015; Kesler and Simon, 2015; Pell et al., 2021; Wall et al., 2017). For example, heavy metal contamination and acid rock drainage (ARD) have been shown to afflict natural waters as a result of CRM (e.g., Cu, Zn, rare

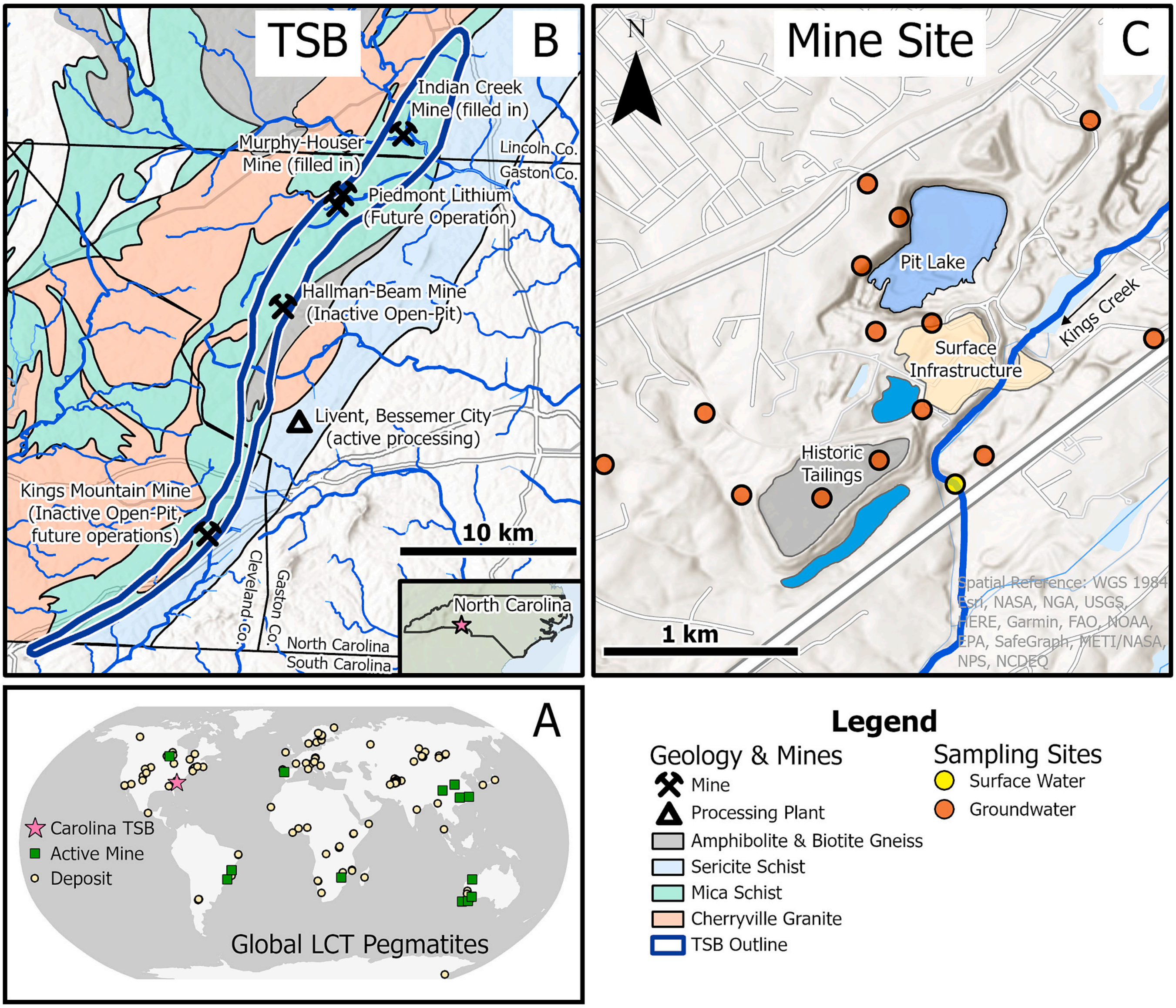

Fig. 1. (A) Map of worldwide LCT pegmatite occurrences with active hard-rock Li mines shown as green squares. Several other deposits have active mines currently mining other commodities. (B) The geology and surface hydrology are shown in the Tin Spodumene Belt (TSB) and the location of the Kings Mountain Mine (KMM). The dark blue TSB Outline shows the vicinity in which the dikes and sills of Li-rich pegmatite are known to exist. (C) Map of the hard-rock KMM Li mine site with relevant surroundings including the pit lake and historic tailings. Kings Creek as shown flows south with headwaters just out of frame.
Data sources (Bradley et al., 2017a; Karl et al., 2013; Natural Resources Canada, 2022; NCGS, 1985; Shaw, 2021).

earth element) and coal mining (Dudka and Adriano, 1997; Kesler and Simon, 2015; Liu et al., 2022; Nordstrom, 2011; Vengosh and Weinthal, 2022). LCT pegmatites are not typically enriched in many of those same heavy metals, but are known to be enriched in a suite of other trace elements, including Li, Cs, U, Ta, Tl, Be, B, F, P, Mn, Ga, Nb, Sn, and Hf, which could have environmental effects upon mobilization into the aquatic system (Bradley et al., 2017a). Despite known enrichments of many incompatible elements, there can be huge variations in elemental composition between and within LCT pegmatites (Bradley et al., 2017b; London, 2008). Previous studies have shown varied and inconsistent results related to the environmental effects of hard-rock Li mining. There have been reports of elevated Li and B in waters from a Li mineral processing facility in Pennsylvania (US EPA, 2019), Li in waters associated with an abandoned Li mine site in Canada (Roy et al., 2022), and Li in surface waters near hard-rock deposits in Central Europe (Toupal et al., 2022). While a water quality study of tailings ponds from an LCT pegmatite in South Dakota has reported pH in the 8–9 range with low concentrations of heavy metals (Rahn et al., 1996), a study of tailings affected surface waters near a hard-rock Li mine in China also reported high pH but with elevated levels of As, Pb, and Mn (Gao et al., 2021). Although impacts on water quality from Li mining in NC have not been directly studied, water quality fluctuations in a stream have been previously attributed to a Li ore processing facility upstream (NCDENR, 2010, 2007). Overall, mine wastes are one of the world's largest waste streams, and are often a source of environmental contamination in water resources near mines (Hudson-Edwards et al., 2011). Since LCT pegmatite mining is expected to generate large amounts of solid waste (Bradley et al., 2017a; Nassar et al., 2022), understanding the potential risks of water resources around Li mining with associated solid wastes is the focus of this study.

Given the variability in literature on the environmental impacts induced from hard-rock Li mining, and the fact that Li concentrations in waters have not regularly been reported, this study aims to evaluate the potential water quality impacts resulting from legacy hard-rock Li mining through investigating a broad range of trace metals. The evaluation is based on understanding the ambient water quality and the leachability of trace elements from natural ore rocks, tailings, and waste materials in a legacy Li mining site. Here we focus on evaluating elements relevant to water quality standards as regulated by the US EPA for drinking water and ecological quality (i.e., aquatic life; US EPA, 2009, 2021a, n.d.) combined with analyzing other elements known to be enriched in LCT pegmatites, including Li, Cs, Ta, Rb, P, and Nb (Bradley et al., 2017a; Černý, 1992). We hypothesize that elements commonly found in LCT pegmatites will be found in elevated concentrations in waters around the mine as compared to background levels. Integrating the leaching test results with the observed water quality data, this investigation can provide a unique and useful observation on the potential impact of hard-rock Li mining on water quality.

2. Background and methods

2.1. Background and site description

In NC, Li was historically mined from the LCT pegmatites of the TSB (Fig. 1B), where pegmatite bodies occur as dike and sill intrusions, primarily into mica schist and amphibolite host units (Horton, 2008; Kesler, 1942; Swanson, 2012). Spodumene, a pyroxene, is often the primary Li-bearing mineral in such pegmatites and is separated from other minerals for Li extraction (Bradley et al., 2017b; Garrett, 2004; Norton, 1973). The pegmatite had historically been exploited in small-scale tin mining operations, and later in small-scale Li mining operations during the first half of the 20th century. Lithium gained economic importance in the 1950s and during this period, operations were expanded at the Kings Mountain Mine (KMM, formerly the Foote Mine) and other open pit operations commenced nearby (Fig. 1B–C). Eventually all mining operations ceased by the early 1990's when the South American Li brines became more economically viable (DOE, 2010; Garrett, 2004; Mohr et al., 2012; Swanson, 2012). For much of this period the TSB was the most productive global Li deposit (Bleiwas and Coffman, 1986; Broadhurst, 1956; Garrett, 2004; Kunasz, 1976; Quan, 1976). With renewed interest in Li exploration, and the projected shortcomings of the global Li supply, mineral exploration of the TSB has begun again with operations intended to recommence in the coming years. The KMM is currently an inactive operation in Cleveland County, NC with a pit lake, waste rock dumps (WRDs), and tailings storage facilities (TSFs) (Fig. 1C). Kings Creek is the only surface water outflow from the KMM with headwaters in the mine site. The hydrogeologic environment is characterized by massive and foliated crystalline rocks featuring fracture flow (Daniel III et al., 1997). In the TSB, the granitic pegmatites are typically intruded into amphibolite and mica schist host rock units (Horton, 2008).

2.2. Sample collection

Solid samples collected from the KMM (Fig. 1C) include three whole rock pegmatite samples from outcrops within the existing pit and one from a fresh core, two waste rock (amphibolite) samples collected from fresh cores, five tailing samples from a recent pilot spodumene separation project (comprising various streams such as magnetic separates, dense media separates, and cleaner concentrates), and one tailings sample from the TSF of the original operation. Additionally, two amphibolite and eight pegmatite ore samples were collected from outcrops and waste piles external to the KMM. All solid samples were oven dried at 60 °C and crushed in a ring and puck mill. Mineralogical information can be found in the supplemental text.

Water samples were collected from monitoring wells around the KMM screened in bedrock unless otherwise noted ($n = 14$) in March of 2023. Kings Creek ($n = 11$) was sampled along a 33 km transect, and background streams within the TSB ($n = 26$) across several sampling trips in 2023 and 2024. Water sampling followed established U.S. Geological Survey (USGS) protocols including immediate filtration and preservation in HDPE bottles (USGS, 2006). Samples for trace element analyses were preserved in acid washed bottles and acidified to pH <2 with HNO_3.

2.3. Bulk digestions

All solid samples were homogenized, dried, and crushed prior to digestion. Digested following an HF + HNO_3 procedure described elsewhere using 50 mg of sample (Wang et al., 2020). The accuracy and precision of the digestion were assessed by comparison to reference concentrations (Jochum et al., 2005; OREAS, 2019a) for external granite and LCT pegmatite standards, USGS G2 and OREAS 753 (Table S1).

2.4. Leaching experiments

The U.S. Environmental Protection Agency (EPA) Leaching Environmental Assessment Framework (LEAF) method 1313 is designed to evaluate the partitioning of constituents between liquid and solid phases at a wide pH range (US EPA, 2017). This followed a slightly modified procedure using extraction fluids with an initial target pH range of 2 to 12 (i.e., 2, 4, 5.5, 7, 8, 9, 10.5, and 12) prepared via titration of 0.01 M HNO_3 and 0.01 M NaOH (typically KOH) solutions in HDPE bottles. Na was not measured due to the use of NaOH. Pre-weighed solid samples were mixed with each extraction fluid in a liquid to solid ratio of 10:1 (mL:g) and placed in metal free centrifuge tubes on a platform shaker at 180 rpm for 24 h. Final solutions were filtered (0.45 μm); one aliquot was taken for pH measurement immediately after extraction and another was acidified for trace metal analysis. While the pH range likely exceeds the expected natural field conditions, understanding the conditions that control the leachability of trace elements is important when comparing to the natural pH of waters interacting with these solids. A detailed explanation of the method choice can be found in the supplemental text.

Two separate water leach experiments were conducted: (1) A 24-hour leaching following the same protocol at the LEAF 1313 leaches using DI water (resistivity >18.0 MΩ/cm) as the extraction fluid and 10:1 liquid to solid ratio; and (2) A 25-days second DI water leaching experiment using a 5:1 liquid to solid ratio, using two Li ore samples. The leachate solutions were decanted, filtered, and collected.

2.5. *Analytical techniques*

For water, solid, and leachate samples, trace and major elements were analyzed on a Thermo Fisher X-Series II inductively coupled plasma mass spectrometer (ICP-MS), while radionuclides were analyzed on a Canberra Broad Energy 5030 Germanium Gamma detector (Vinson et al., 2018; Wang et al., 2022). Major anions in waters were measured via ion chromatography with a Thermo Dionex IC DX-2100, and total alkalinity and bicarbonate were determined via titration to pH 4.5 with 0.02 N HCl.

The major mineralogy was determined using an Anton Paar XRDynamic 500. Mineral phases were identified and quantified using Profex (Doebelin and Kleeberg, 2015). Phosphate minerals are known to occur regularly in the pegmatites but at relatively low abundance, typically <1 wt%, and in many different phases making them difficult to quantify with XRD (Hodges, 1983; Swanson, 2012; White, 1992). Thus, phosphate mineral abundance is estimated from bulk P concentration and calculated assuming all P is in the fluorapatite phase ($Ca_5(PO_4)_3F$).

2.6. *Calculations and statistics*

Enrichment factors (EFs) are concentration ratios, $EF = [X]_{sample} / [X]_{reference}$, where [X] is the concentration of the element. Solid samples are compared to the mean baseline soil compositions (0–5 cm) for the USA (Smith et al., 2013) as a representative environmental baseline with the exception of B, Cs, and Ta, for which concentrations were not available and upper continental crust values were substituted (Rudnick and Gao, 2014). Nonparametric analyses were performed using Spearman's rank correlation where rho (ρ) represents the correlation coefficient and *p*-value (p) represents statistical significance.

3. Results and discussion

3.1. *Water quality of natural water resources at the mine site*

Water quality analysis of the surface waters and groundwater from the KMM site (Figs. 1, S1) reveals that none of the investigated water samples had contaminates levels that exceed the EPA drinking water standards (US EPA, 2009). This includes F, Be, and Tl, which are considered elements of concern in hard-rock Li mines due to their natural enrichment in LCT pegmatites (Bradley et al., 2017a; Liu et al., 2023; Toupal et al., 2022). The concentrations of As, Se, Cd, Ni, Cu, and Pb that can potentially pose risks to freshwater ecological systems did not exceed the threshold values defined by the EPA chronic criterion concentrations of aquatic life (US EPA), except for Fe (>1000 μg/L) in two, and Cu (>9 μg/L) in three groundwater samples. From the

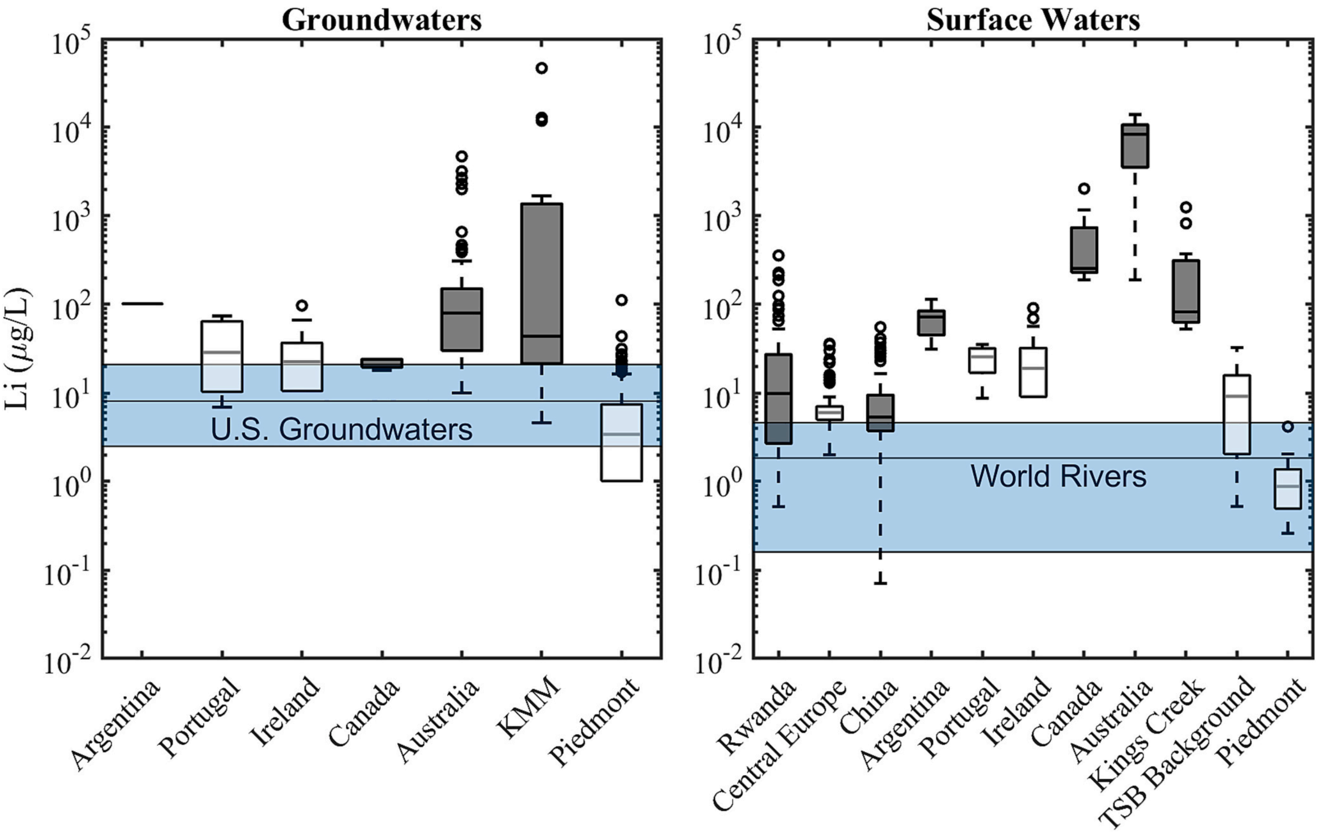

Fig. 2. Lithium concentrations (log scale) in groundwater and surface waters at the KMM as well as data reported for other LCT pegmatite deposits and mines (CELC, 2020; Marqués et al., 2023; Rodrigues et al., 2019; Toupal et al., 2022). Boxes highlighted in black include samples from active and legacy LCT pegmatite mines, including the active Jiajika mine in China and Greenbushes mine in Australia (Gao et al., 2019; Nieder et al., 2014; Roy et al., 2022; Talison Lithium, 2020, 2018). The data from Ireland is simulated from descriptive statistics assuming a normal distribution (Kavanagh et al., 2017). For reference, the groundwater data was compared to Li concentrations in groundwaters of the NC Piedmont (Coyte et al., 2020) and to the inter-quartile range and median of groundwaters used for drinking water across the U.S.A. (blue box and black line) (Lindsey et al., 2021). Lithium contents in surface waters were compared to the range and mean concentration (blue box and black line) reported for world rivers (Gaillardet et al., 2014).

unregulated elements known to be enriched in LCT pegmatites, only Li, Rb, and Cs were notably elevated, while Ta and Nb were typically below detection (<0.01 μg/L and < 0.02 μg/L, respectively) and PO_4^{3-} was below 0.5 mg/L in all but one groundwater sample.

In groundwater, Li concentrations varied considerably across four orders of magnitude, from 4.6 μg/L to 46.7 mg/L (median 41.4 μg/L; Fig. 2). Of these, two outlier samples with the highest concentrations were detected in an open borehole adjacent to the mine pit lake (46.7 mg/L) and a well screened in the historic TSF (12.8 mg/L). The rest of the samples had Li contents below 1700 μg/L (mean 225 μg/L; Fig. 2). The open borehole and the TSF likely have greater water-rock interaction since the borehole was drilled within the last few years and might have residual pulverized rock, while the TSF is largely characterized by crushed rock, both increasing the surface area of minerals and likely induce intensive water-rock interactions. By comparison, the regional aquifer with groundwater of lower Li content is characterized by a fracture flow due to the crystalline aquifer lithology, which likely induces a lower degree of water-rock interactions (Daniel III et al., 1997). Furthermore, the Li concentrations in the groundwater from the mine site were significantly higher than from the NC Piedmont regional

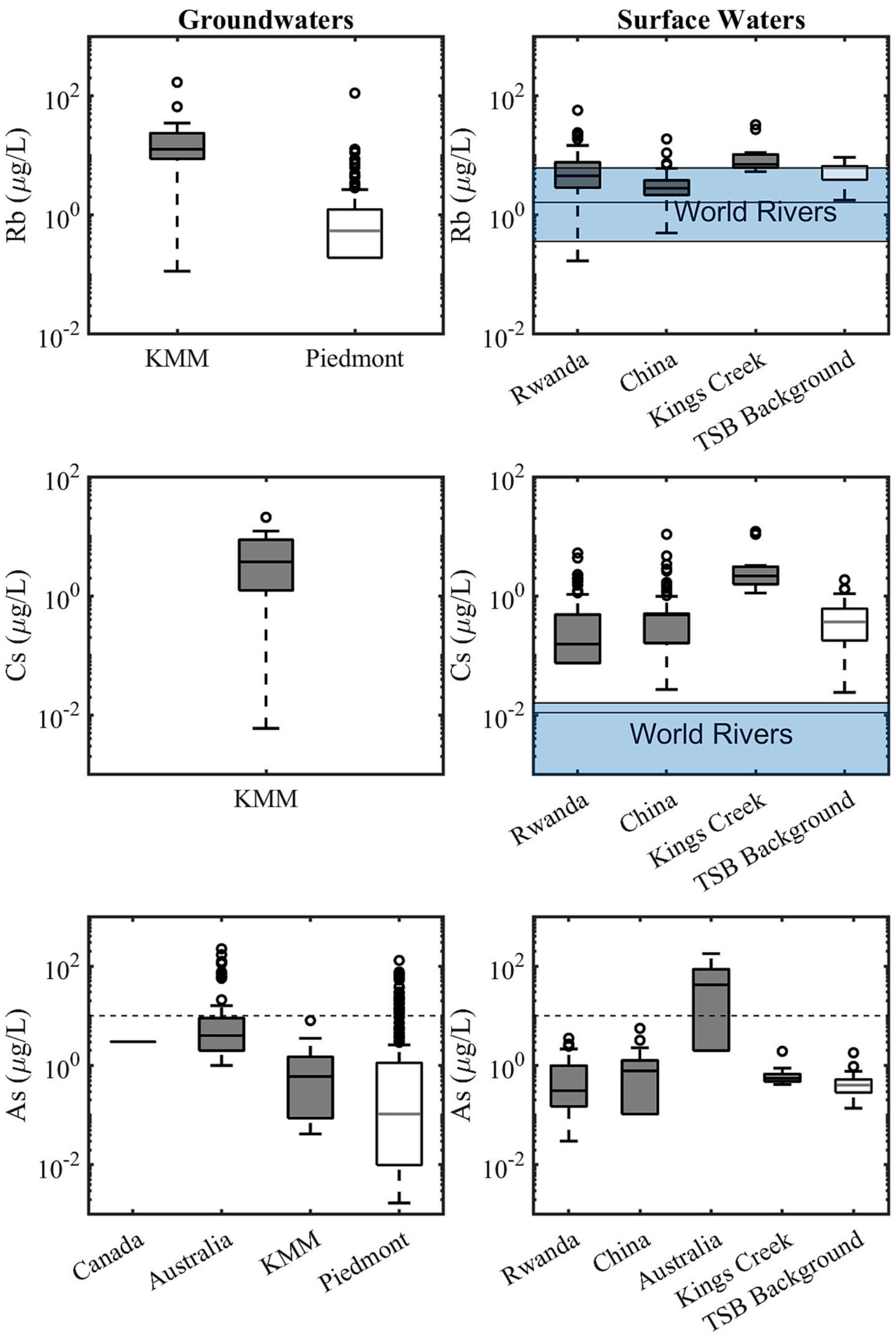

Fig. 3. Rubidium, Cs, and As concentrations (log scale) in groundwater and surface waters associated with LCT Li pegmatite sites. Highlighted black boxes include samples from active and legacy LCT pegmatite mines. Groundwater concentrations are compared to those from the NC piedmont (Coyte et al., 2020) and surface waters are compared to the range and mean concentrations (blue box and black line) reported for world rivers (Gaillardet et al., 2014). For As, the EPA maximum Contaminant Level standard of 10 μg/L is shown as a dashed line (US EPA, 2009).
Data for other sites compiled from (CELC, 2020; Gao et al., 2021; Nieder et al., 2014; Talison Lithium, 2020, 2018).

groundwater ($p < 0.01$), which typically has Li concentrations <7 μg/L. In comparison to other Li mine sites, groundwater adjacent to other LCT pegmatites has typically lower Li concentrations, yet those from the active Greenbushes mine in Australia are similar (median 80 μg/L versus 225 μg/L; Fig. 2) (Talison Lithium, 2020, 2018).

Both Rb and Cs co-occurred at relatively elevated concentrations in the groundwater of the KMM, up to 169 and 21 μg/L, respectively (Fig. 3). Rubidium typically exceeded 8 μg/L (median = 12.4 μg/L), while Cs exceeded 1 μg/L (median = 5.3 μg/L) in the groundwater samples. In comparison to the regional Piedmont groundwater, Rb (median 0.54 μg/L; Fig. 3) is much lower ($p < 0.01$), while Cs has not been previously reported. Cesium, however, has been found at median concentrations of 0.02 and 0.096 μg/L in carbonate and fractured bedrock aquifers, and up to 5–6 μg/L in a fractured granitoid aquifer in Sweden, generally lower than our results at the KMM (Frengstad et al., 2000; Mathurin et al., 2014; Morgantini et al., 2009).

Kings Creek is the only surface water draining the mine site and was sampled systematically for 33 km downstream of the KMM to its confluence with the Broad River (Fig. S1). Elevated Li, Rb, and Cs were detected at the upstream section downflow from KMM compared to the levels in the background streams from the TSB region (p < 0.01). Yet further downstream, each of these elements was diluted with distance (Fig. 4). Emerging from the KMM, Li content was 828 μg/L and decreased to 52.7 μg/L after 33 km, remaining above the background level (<32 μg/L). Rubidium began at 27.4 μg/L and decreased to 7.9 μg/L, within the background range (<9.3 μg/L) after 4.2 km. Cesium began at 10.8 μg/L and dropped to 1.1 μg/L after 28.4 km, reaching background levels (<1.9 μg/L) (Fig. 4). The relative rates of decrease to background levels of these elements along the downstream flow correspond to the magnitude of their enrichment in the most upstream sample downstream from the KMM relative to the mean background level, with enrichment following Li > Cs > Rb. While the Kings Creek samples had higher concentrations relative to those of the background streams within the TSB region (mean Li of 10.5 μg/L), these background samples were also more concentrated than those reported for surface waters in the Piedmont (mean 1.16 μg/L, p < 0.01) (Anderson et al., 1988), suggesting that the Li concentrations are naturally elevated in surface waters of the TSB also away from legacy mine sites.

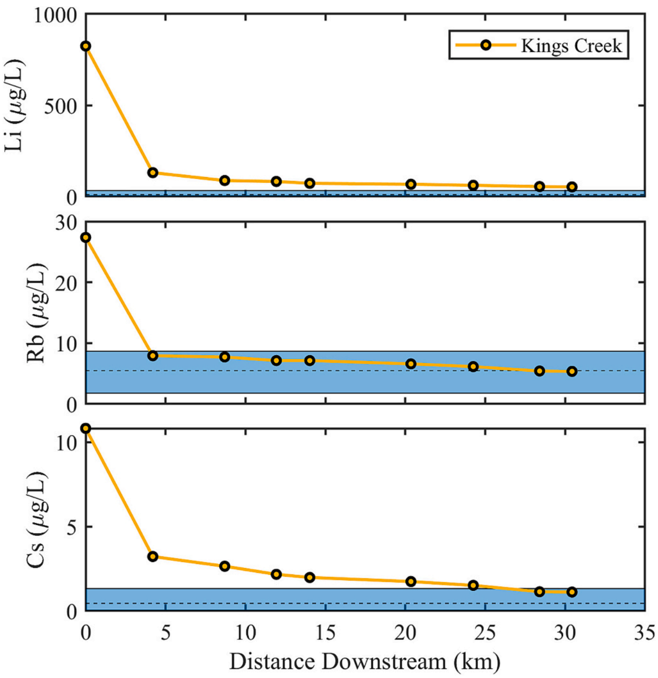

Fig. 4. Profiles of Li, Rb, and Cs concentrations in surface water of Kings Creek over 33 km flow downstream from the KMM outfall before its confluence into the Broad River. At the outfall from the KMM, Kings Creek had elevated concentrations of Li, Rb, and Cs, which gradually decrease with flow distance to background levels shown as the shaded blue region.

By comparison, the Li concentration in the most upstream sample of Kings Creek, at the KMM, is roughly the same level as that of a legacy LCT pegmatite mine reported in Canada (Roy et al., 2022), yet lower than surface waters near the active Greenbushes mine in Australia (Talison Lithium, 2020, 2018), and higher than those around the active Jiajika mine in China (Fig. 2) (Gao et al., 2019). Additionally, stream water surveys of Li concentrations near other global LCT pegmatite deposits, typically yield lower concentrations (Fig. 2). Rubidium and Cs tend to be reported less frequently than Li, and therefore, data comparisons are less readily available. For surface waters, Rb and Cs in Kings Creek at the KMM were higher than those from a legacy LCT pegmatite mine in Rwanda and the Jiajika mine (Gao et al., 2019; Nieder et al., 2014). Nearly all of these values including those from TSB background streams fall within or just above the range of concentrations of world rivers (Fig. 2) (Gaillardet et al., 2014).

While As was not found at concerning levels in the KMM waters (max: 8.0 μg/L relative to EPA MCL 10 μg/L), it has been recorded at higher concentrations in groundwater and surface waters from the Greenbushes mine in Australia, up to 224 μg/L (mean 32 μg/L) (Talison Lithium, 2020, 2018). Yet in the Jiajika mine, tailings-affected surface waters had As concentrations above background streams levels, yet still below water quality thresholds (up to 5.6 μg/L) (Gao et al., 2021). Similar low As concentrations have been reported for surface waters at an LCT pegmatite mining district in Rwanda (up to 3.5 μg/L) (Nieder et al., 2014). The As concentrations in waters from Greenbushes are significantly higher ($p < 0.01$) than those from the KMM and Jiajika mines, while the As concentrations in KMM groundwater are indistinguishable from the range of groundwaters within the Piedmont of NC (Fig. 3) (Coyte et al., 2020).

The pH of nearly all investigated natural waters fell within a circumneutral range (pH 6–8); the exceptions were four groundwater samples with pH 5.2–5.9, screened in mica schist and phyllite units, and the high pH of the open borehole as previously discussed. The pH range reported in this study is generally similar to pH values reported from waters associated with a Li pegmatite mine in Canada (Roy et al., 2022) and in China (Gao et al., 2021), but are mostly lower than values reported at a pegmatite mine in North Dakota (pH of 8–9) (Rahn et al., 1996). Our data show that the pH generally correlates with electric conductivity ($\rho = 0.62, p < 0.05$) and bicarbonate content ($\rho = 0.85$, $p < 0.01$) in the KMM groundwater (Fig. S2). This indicates that intensification of water rock interactions enhances solute mobilization and induces alkaline conditions (see discussed in Section 3.2.3). Additionally, the Li/Na ratio in groundwater generally increases with pH ($\rho = 0.54$, $p < 0.05$), indicating preferential mobilization of Li over Na with higher pH.

Overall, these results indicate that Li, Rb, and Cs are naturally elevated in waters around the TSB region, with relatively high concentrations in groundwater associated with the legacy Li mine. It is clear from the Kings Creek investigation that the legacy of mining has further increased the concentrations of Li, Rb, and Cs in the surface water near the outfall, above background levels. Lithium, Rb, and Cs are not regulated as contaminants, and their health impacts at the levels found in water are poorly constrained. However, it has been suggested that Li concentrations in river water up to 103.9 μg/L downstream from a Li processing site in China could result in Li bioaccumulation in aquatic life and potentially affect human health (Yang et al., 2024). While none of these elements are regulated as contaminants, their unusually high concentrations in waters associated with Li mining and processing facilities might warrant further investigation. Given the increasing attention on and the use of Li, the US EPA has added Li to its unregulated contaminant monitoring rule for public water systems and its drinking water contaminant candidate list (US EPA, 2021b, 2021a).

3.2. Chemical compositions and leachability of solid materials

To understand the observed water quality variations in natural waters, we have investigated the different solids that can influence the water quality around the KMM and TSB region. We analyzed the bulk elemental compositions of ore grade LCT pegmatite, waste rock (amphibolite), and tailings materials, as well as conducted leaching experiments to evaluate the magnitude of leachability of the different elements extracted from the solid sources into the aquatic phase.

3.2.1. Bulk composition of lithium ore and solid wastes

To evaluate the possible enrichment of certain elements in the solid samples, we compared their concentrations in the solids to those of the mean baseline surface soil (0–5 cm) for the USA (Smith et al., 2013), and these ratios are defined as Enrichment factor (EF). The soil baseline was selected as a reference since soils are one of the most important solid media influencing water quality (Drever, 1997). Elemental enrichment above the baseline could be indicative of potential sources of water contamination. In the pegmatites, three elements were highly enriched with EFs >10 (Fig. 5), including Li (EF = 369), Be (EF = 116), and Ta (EF = 17), while other elements common in LCT pegmatites were moderately enriched (EF > 5), including Nb, Tl, Rb, and Cs (Bradley et al., 2017a, 2017b). Other elements such as P and U had mild enrichment (EF > 2) relative to the soil baseline (Fig. 5) but were not uniquely elevated. Reported trace element values for LCT pegmatites in Canada and Australia show similar enrichment trends and are generally known to have similar elemental concentrations. Importantly, our study found low concentrations of potential contaminants (e.g., As, Se) in the Li ores, similar to results reported in previous studies, indicating that this pattern occurs also in global LCT pegmatites (Fig. S3) (Barnes, 2010; OREAS, 2019a).

The tailings samples are composed of residual material following bulk pegmatite rock crushing and mineral separation to concentrate spodumene. Of the average composition of the tailing samples, Li, Be, Ta, and Cs were highly enriched (EF > 10), while Nb, Tl, Rb, U, P, and Fe were moderately enriched (EF > 5) (Fig. 5).

The other type of waste material expected to be produced from hard-rock Li mining is waste rock, which are the rocks surrounding the pegmatite ore that are removed for access to the pegmatite. It is primarily made up of an amphibolite rock unit, which the pegmatite has intruded into. Waste rock samples showed a high degree (EF > 10) of enrichment in Li and Mn, while Co, Sc, and V were moderately enriched (EF > 5). Similar waste rock compositions might be expected at other LCT pegmatite deposits since they commonly form under similar geological conditions, hosted in upper greenschist and lower amphibolite facies (Bradley et al., 2017a; Černý, 1992). However, the exact compositions will likely vary depending on the protolith. It is important to note that several common contaminants such as Pb, As, and Ba were lower in the KMM pegmatite, tailings, and waste rock samples relative to the soil baseline levels (Fig. 5). These results are consistent with a previous report of low concentrations of heavy metals relative to the upper continental crust reported for pegmatites and wastes from a mine site in Rwanda (Lehmann et al., 2014).

A comparison of the average tailings chemistry to the original pegmatite composition from the KMM samples shows a relative enrichment of Sc (EF = 26.3), V (EF = 871), Cr (EF =155), Ni (EF = 52), Cu (EF = 154), Mo (EF = 10.6), and Ba (EF = 24.6), indicating that the tailings become enriched in minerals containing these elements as they are separated from the parent pegmatite. The absolute concentrations of these elements, however, are not higher than those of the soil baseline (Fig. 5).

While radioactivity was not measured in waters, it does merit discussion since mining operations can concentrate radioactive minerals into different waste fractions that can induce human and environmental health risks. In Australia, for example, LCT pegmatite ores contain <65 Bq/kg for combined U and Th, while solid wastes become 10-fold enriched, to a range of 7500–75,000 Bq/kg (Cooper, 2005). Here we measured the U and Th progeny nuclides of ^{226}Ra and ^{228}Ra in the Li ore and waste rocks. Total Ra activities ranged from 22 to 201 Bq/kg in the pegmatites, 26 to 297 Bq/kg in tailings, and 7.5 and 45 Bq/kg in waste rocks. Assuming that the rocks and waste solids are in secular equilibrium, these activities should be indicative of those of U and Th. The

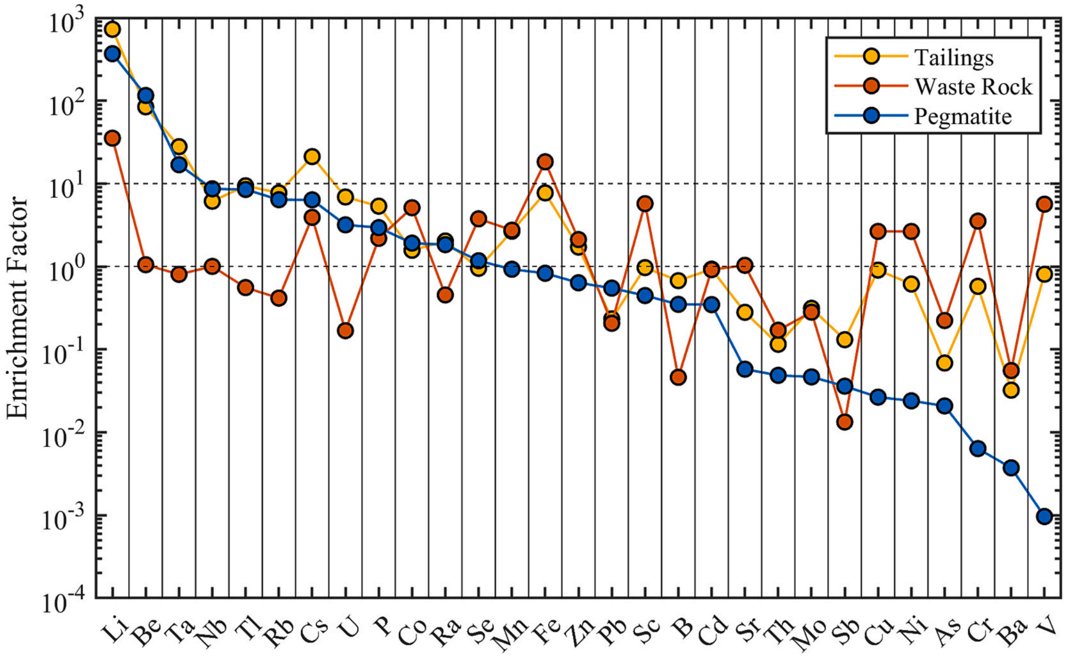

Fig. 5. The average enrichment of select elements measured in pegmatite ore, waste rock, and tailings relative to their mean concentrations in USA soils (Smith et al., 2013). Horizontal dashed lines show enrichment factors of 1 and 10, from which can be classified as highly enriched. Elements are arranged in decreasing enrichment order for the pegmatite samples. Several trace elements were enriched in the tailings and waste rock as compared to their concentration in the pegmatite ore with apparent lower enrichment factors.

average activity of the pegmatite (107 Bq/kg) is greater than the levels reported in Australia, however the high degree of enrichment in the tailings was not observed with a mean total Ra activity of 119 Bq/kg in tailings samples, indistinguishable from the pegmatite. For reference, the average activity of combined ^{226}Ra and ^{228}Ra in the USA soil baseline ranges from 58.5 Bq/kg in topsoils (top 5 cm) to about 64.3 Bq/kg in deeper C horizon soils (~100 cm deep), as calculated from U and Th concentrations (Smith et al., 2013). Soil from the nearby Lake Norman State Park has an activity of 114 Bq/kg (Wang et al., 2021). While the Ra levels of background solids can vary considerably, the Ra values we report for the KMM ore, waste rocks, and tailing are relatively low and not appreciably different from both the soil baseline and local soil.

Consistent with previous studies, only a few elements (Li, Be, Ta) are highly enriched in the whole rock pegmatites (London, 2008), of which only Be is regulated as a contaminant in drinking water (US EPA, 2009). While the waste rock and tailings samples show enrichment of several elements relative to the Li pegmatite, there are no high enrichments (EF > 10) of common contaminants over the soil baseline reference. Despite these low concentrations of potential contaminants in the solids, their potential leachability is as important in determining the potential influence of these solids on water quality as low concentrations can have an outsized effect if they are highly leachable.

3.2.2. *LEAF-1313 leaching experiments*

Following the U.S. EPA LEAF 1313 method, the solubility of trace elements was investigated at a wide range of $pH_{initial}$ (controlled pH at experiment start), from 2 to12. While the full range of pH tested is unlikely to occur at the natural mine conditions, this testing can indicate the pH conditions necessary for trace elements to become soluble. This is important for understanding how changes in water chemistry can affect water quality. Furthermore, this test can provide general information of the buffering capacity of the investigated solid samples.

Similar to the findings from the water leaches (Section 3.2.3), all samples demonstrated alkaline tendencies and buffered the solution in the $pH_{initial}$ range of 4–9, with an average final pH in this range of 8.8 for pegmatites, 7.9 for waste rock, and 8.8 for all but one tailings sample. One sample from the original TSF had a relatively low buffered pH of 6.7 (Fig. 6A).

The greatest leachable fraction (i.e., the percentage of the elements in the leachate solution relative to the bulk concentration) for nearly all elements was at $pH_{initial}$ of 2. At higher $pH_{initial}$, the percentage of the elements in the leachates drastically reduced (Fig. S4). Only B, Al, Sc, and V (in pegmatites only) exhibited similar leachable percentages at both lowest and highest $pH_{initial}$ of 2 and 12 (Fig. S4), with lower leachability at neutral $pH_{initial}$. The most proportionally extractable elements during the LEAF 1313 experiment were Mg, P, Ca, Cr, Mn, Fe, Co, Ni, Y, La, and Ce, in particular at lower $pH_{initial}$. In each of these elements, the percent leachability was positively correlated with the proportion of P leached (Fig. S5) from pegmatite and tailings samples (waste rock excluded due to low total P content). This suggests that these elements are being leached from phosphate minerals within the pegmatite and tailings, which are known to occur as minor minerals in pegmatites of the TSB (0.5–1.9 wt% in our samples) (Horton, 2008; Swanson, 2012; White, 1992, 1981).

On the other hand, elements that are typically silicate bound showed the lowest percentage of leachability at any $pH_{initial}$, with Li and K leaching <2.5 % of bulk pegmatite concentrations, and Al leaching even less at <0.3 %. Both Li and K show a marked decrease in leachability above $pH_{initial}$ of 2, with proportional leaching ranging from <0.1 % to ~1 % in individual experiments (Fig. S4). Rb and Cs follow a similar trend to that of Li and K, but with total leaching at much lower levels. Other trace elements, regulated for water quality, including Ba, Tl, Cu, Zn, Cd, Pb, and U also followed this pattern with the highest leachability at lower $pH_{initial}$, while others including Se (<0.0036 mg/kg), Mo (<0.0003 mg/kg), Sb (<0.0001 mg/kg), and Th (<0.0004 mg/kg) were typically below detection. Tantalum (<0.0001 mg/kg) and Be (<0.00003 mg/kg), elements enriched in the pegmatites, were also typically below detection in the leachate solutions.

These findings show that most elements are not highly leachable under circumneutral pH. Instead, low-pH conditions would be required to induce high-degrees of leachability. Given that our data show that water-rock interactions of the whole rock pegmatites, waste rock, and tailings give rise to alkaline conditions, we predict that such water-rock interactions would lead to low mobilization of trace elements from these

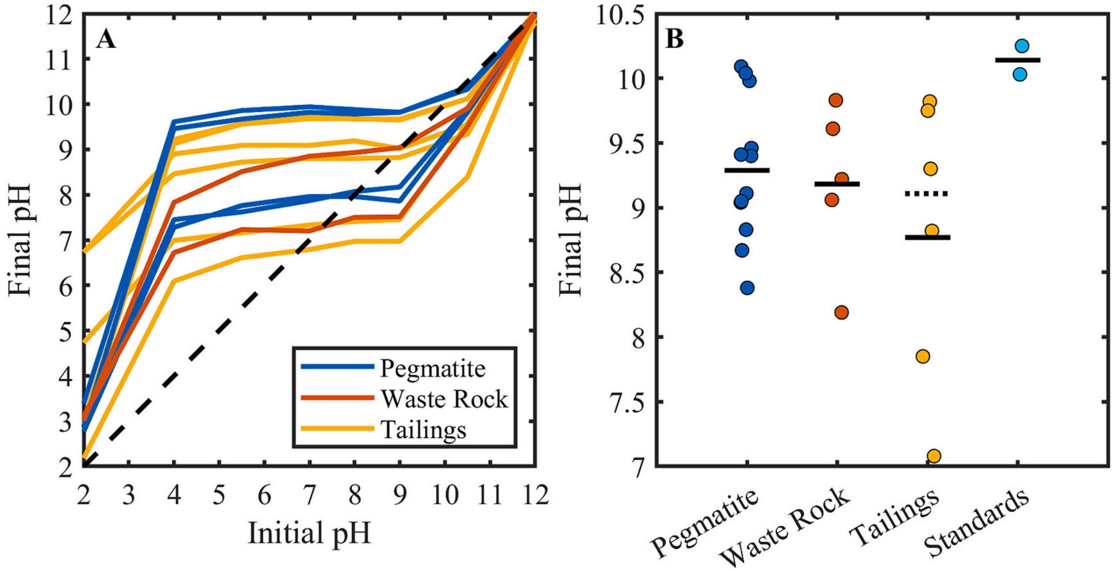

Fig. 6. (A) Evolution of pH in effluents interact with different solid materials colored by sample group during the LEAF 1313 experiment, where initial pH is the starting pH of the leaching solution and final pH is the pH after the completion of the experiment. The black dashed line represents a 1:1 ratio between the initial pH and final pH. Values above this line indicate that the final pH increased from the initial value. The results show that all samples were relatively well buffered between initial pH of 4 and 9. (B) Variations of final pH of the DI water leaching experiment. The black horizontal lines represent the mean final pH of each solid group. The dashed black line in the tailing column shows the average final pH of tailings samples from the pilot plant, excluding the sample from the TSF of the original KMM operation. Note that the water started at a pH of 7 and rose to alkaline conditions in all experimental solid samples. The two standards shown are a pegmatite and spodumene concentrate (OREAS, 2019a, 2019b).

solid sources. Despite the relatively high leachability of a few contaminants, the absolute concentrations leached into solution were low, indicating if they are leached into natural waters, the concentrations would likely be low. Nonetheless, Li, Rb, and Cs concentrations were all notable in the leachates. A few of these elements do display oxyanion tendencies with greater leachability at higher pH, yet the same low leachability is observed (e.g., As, Cr, and Se). Shown in Section 3.1, the generally circumneutral pH of groundwater and surface water from the Li mine site suggests that ambient water quality conditions would not initiate a high degree of leachability of any elements.

3.2.3. *Short-term water leaching experiments*

Deionized (DI) water leaching experiments were employed to understand the natural buffering of the solids in the Li mine, as well as the leaching characteristics of trace elements independent of acid or base solutions. While DI water would not be found in natural conditions, this is used to understand how water quality is affected solely as a function of the solid composition and it's reactivity with water.

Water leaching show high proportions (>5 %) of P, V, Co, Ni, and As in leachates from whole rock pegmatite and tailings, while little to none is observed in the waste rocks (Fig. S6). Other elements of concern to water quality such as Cr, Sb, Pb, and U had low leachability, up to a few percent. Lithium leaching was below 1 % of total Li content, while the highest proportions of Li were leached from waste rocks (Fig. S7). The findings of the LEAF 1313 leaching experiments indicate that Li, Rb, and Cs concentrations were all notable, which is consistent also for the water leachates experiments. Thallium is known to concentrate in LCT pegmatites and has been identified as a potential water quality issue (Phan et al., 2021), however, the water leaching experiments yield low Tl concentrations and percent in the leachates (Fig. S7).

At least three mineral groups in LCT pegmatites and mines can be asserted for Li mobilization to water resources (Roy et al., 2023, 2022; Toupal et al., 2022): (1) lithium-rich phosphates, (2) spodumene, and (3) lithium-rich micas. Li-rich phosphate minerals are known to occur in LCT pegmatites and are found at the KMM (total phosphates 0.5–1.9 wt %) (Hanahan, 1985; London, 2008). The most common phosphate minerals at the KMM are two lithium bearing varieties of ferrisicklerite and montebrasite and Fe and Ca dominant varieties of fluorapatite and vivianite (Hodges, 1983; Swanson, 2012). Lithium and PO_4^{3-} in the water leachates of the pegmatite samples show a strong correlation ($\rho = 0.62$, $p < 0.05$; Fig. 7A). If all the Li in solution was from phosphate dissolution, the molar ratio of Li:PO_4^{3-} would be <1 (Fig. 7A), which is the maximum ratio for montebrasite or ferrisicklerite. In all but 2 of the leaching experiments, this ratio was >1 (mean 2.17), suggesting that some of the Li is derived from phosphate dissolution, but indicating that phosphate minerals cannot explain all Li mobilization to the water leachates. Roy et al. (2023) favored partial spodumene dissolution from leaching experiments of LCT pegmatites from Canada, following an elevated weatherability model for spodumene (London, 2017; Roy et al., 2023). In this model, spodumene ($LiAl(SiO_3)_2$) is susceptible to alteration, as Li is preferentially released during weathering due to the small atomic size of Li having induced a weakness in the crystal structure during cooling (Cameron et al., 1973; London, 2017; Singh and Gilkes, 1993). This could also explain why no trends are observable between Li and Al in the leachates, if Al remains in the residual solid. While it is possible that Li could also be derived from Li-rich micas, there are no apparent correlations between Li and either K or Rb, which should co-occur from mica dissolution. Given the strong correlation between phosphate and Li, the relative dominance of spodumene (12–34 wt%) over Li rich micas (<1 %) (Swanson, 2012), and the comparative weatherability of phosphate minerals (0.5–1.9 wt%) to silicate minerals it would seem likely that the majority of mobile Li in the water leachates is from lithium-bearing phosphates and spodumene.

Despite a relatively low percent leachability of Li, the total concentration is relatively high (Fig. S7), indicating that Li is a major outcome of water-rock interactions with pegmatites, tailings, and waste rock samples. The other alkali elements, Rb and Cs, which follow similar leaching characteristics, are likely to co-occur at elevated concentrations from water-rock interactions of pegmatite rocks and waste materials. These results are consistent with the relative enrichment of Li, Rb, and

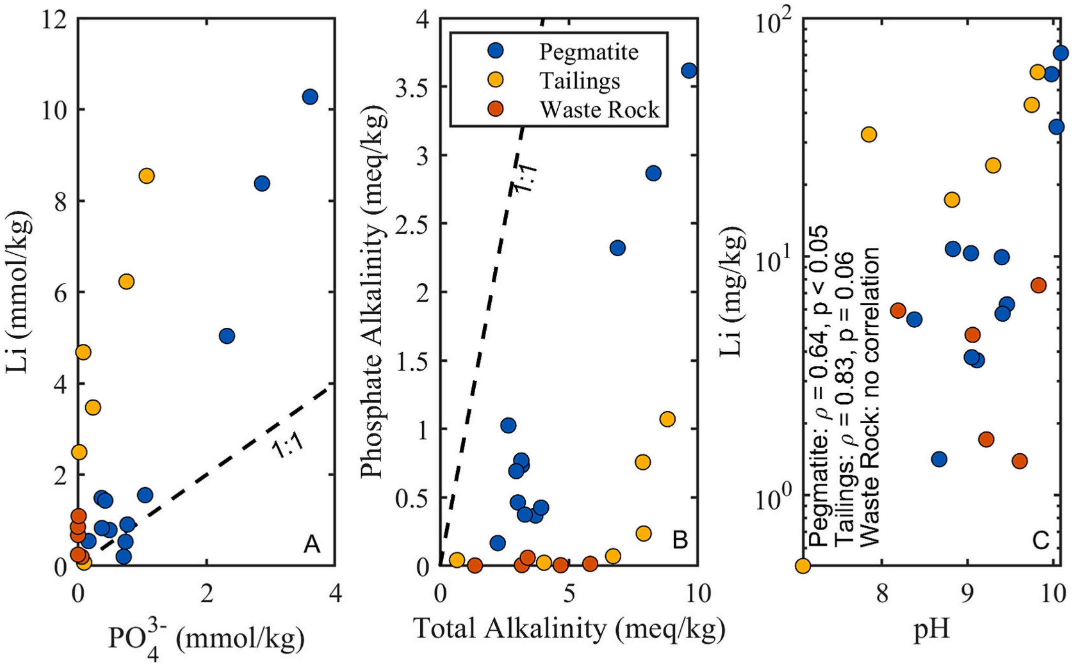

Fig. 7. (A) The molar concentrations of phosphate versus that of lithium in water leachates extracted from leaching experiment tests on pegmatite, tailing, and waste rocks from KMM. The 1:1 line shows the expected ratio in phosphate mineral dissolution. (B) Total alkalinity versus the fraction of total alkalinity that can be explained by phosphate ions in solution. The 1:1 line shows the expected phosphate mineral dissolution to the overall alkalinity that derive from leaching of the pegmatite and tailing samples. (C) Leached Li extracted from the different solid materials versus pH of the leachates. The associated correlation, particularly for tailing suggest that spodumene may assert some control on alkalinity occurrence in the water leachates.

Cs we detected in the natural waters in the mine area (Section 3.1).

All solid samples leached with deionized water displayed an alkaline tendency, with pegmatite leaching generally resulting in the highest pH (mean 9.3), followed by waste rock (9.2), and tailings (8.8) (Fig. 6B). pH in all samples was also associated with an increase in total alkalinity ($\rho = 0.77$, $p < 0.01$; Fig. S8). Given the high concentration of phosphate ions in the leachates for pegmatite rocks and the correlation with total alkalinity (Fig. 7B; $p < 0.001$), the occurrence of phosphate ions may have a strong influence on alkalinity. The pH of all leachates fell between 7.1 and 10.1, which corresponds to HPO_4^{2-} and $H_2PO_4^-$ being the dominant phosphate species and converting primarily to $H_2PO_4^-$ during titration to the pH 4.5 endpoint. Assuming that all phosphate in leachates is one of these two species, phosphate can account for between 7 % and 39 % (mean 23 %) of the total alkalinity in the water leachates (Fig. 7B). Vivianite ($Fe_3(PO_4)_2$) is one of the common phosphate minerals in the TSB pegmatites and has been shown to preferentially release P under alkaline conditions, where Fe(II) is oxidized to Fe(III) precipitates (Metz et al., 2023; Thinnappan et al., 2008). The solubility of vivianite has also been shown to increase from circumneutral pH to pH > 8 (Metz et al., 2023; Thinnappan et al., 2008), a trend seen in some of the individual pegmatite and waste rock samples during the LEAF 1313 leach, where P leaching is greater at higher pH than circumneutral pH. This could explain why there is no observable trend in water leachates between P and Fe or Mn, which would both be redox sensitive, and in part why P increases with pH.

3.2.4. Long-term water leaching experiments

We also conducted a DI water-leaching test over 25 days for two of the pegmatite samples. These tests revealed that the pH, alkalinity, PO_4^{3-}, and Li decreased considerably after the first day of leaching, following strong decreasing trends ($\rho < -0.78$, $p < 0.01$; Fig. 8). The coordinated decrease between Li and PO_4^{3-} and their high degree of correlation ($\rho = 0.71$, $p < 0.05$; Figs. 8, S9) further reinforce our interpretation that Li is dominantly derived from phosphate mineral dissolution during early-stage leaching. Additionally, the fraction of phosphate contribution to total alkalinity decreases from 30 to 50 % to 2–4 % during the 25-day interval coupled with a pH decrease (to circumneutral values), demonstrating that phosphate mineral dissolution also exerts a strong influence on pH. In contrast, Rb and Cs leaching showed no discernable trend during the 25 days and roughly parallel the leaching behavior of K (Fig. S10), lending further evidence they are leaching from K bearing minerals. While this test was only conducted for 25 days, it demonstrates the importance of phosphate minerals in the initial stages of water-rock interactions. We hypothesize that soluble phosphate minerals would become exhausted relatively quickly and that pH might reach a circumneutral equilibrium after long-term water-rock interactions. The open borehole with pulverized rock might be the closest corollary to the water leaches since it had both the highest Li and PO_4^{3-} concentrations, and phosphate can account for 24 % of the total alkalinity in groundwater collected from this well. The rest of the groundwater samples are from cased monitoring wells and would not be interacting with fresh mineral surfaces and phosphate was accounts for only <0.1 % of total alkalinity.

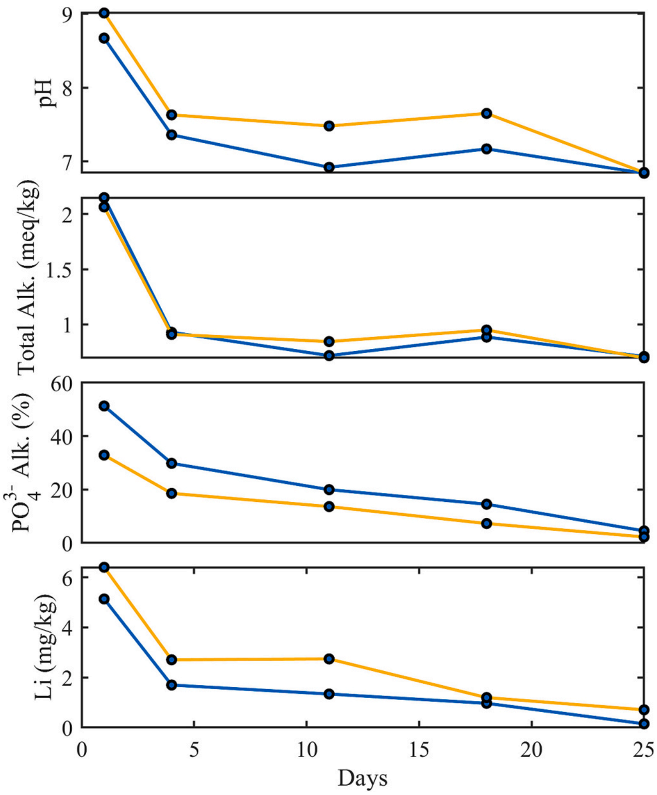

Fig. 8. Results of the 25-day leaching experiment on two pegmatite samples showing a decrease in pH, buffering capacity (as Total Alkalinity), and the relative contribution of the phosphate ion to that buffering capacity with time. The leaching of Li also decreases with time suggesting that the earliest leaching of Li is derived from phosphate and spodumene minerals.

Since only a fraction of total alkalinity and pH can be explained by phosphate dissolution, other minerals should contribute to the elevated pH in the leachates. Roy et al. (2023, 2024) observed neutral to alkaline conditions during water leaching experiments of LCT pegmatites and tailings in Canada, and that the leachate pH decreased with repeated leaching over time. This led to the conclusion that feldspar dissolution induces the alkaline leachate pH, where cation exchange sites with H^+ are eventually exhausted after repeated reactions with water (Mamonov et al., 2020; Roy et al., 2024, 2023, 2022). This was demonstrated in a TSF at a legacy LCT pegmatite mine, where leachate pH was circumneutral at the surface and increased with depth. A trend attributed to regular wetting and drying cycles at the surface, the first section to react with slightly acidic rainwater, reducing the buffering capacity of feldspars (Roy et al., 2023, 2022). This could explain our sample from the TSF of the original KMM operation, which had been collected from the surface and resulted in the lowest leachate pH of 7.1 (Fig. 6B). Feldspars could and likely contribute to the alkalinity of the leachates, yet albite ($NaAlSi_3O_8$; 16–34 wt% bulk pegmatite composition) and K-Feldspar ($KAlSi_3O_8$; 2–19 %) are the dominant feldspars (Kesler, 1961; Swanson, 2012), which would suggest that Na and K are exchanging with H^+ yet neither element correlates with pH in any of the experiments. Lithium, however, shows a high correlation with pH in pegmatites ($\rho = 0.64$, $p < 0.05$) and in tailings samples ($\rho = 0.83$, $p = 0.06$; Fig. 7C). This indicates that preferential release of Li from spodumene ($LiAlSi_2O_6$, 12–34 %) is accompanied by H^+ exchange and a major source of alkalinity in the leachates.

For comparison, leaching experiments of an LCT pegmatite sample from the Bynoe Pegmatite Field in Australia (OREAS, 2019a) and a spodumene concentrate from the Greenbushes region in Australia (OREAS, 2019b) resulted in elevated pH (>10) during the water leaching experiments (Fig. 6B). In the spodumene concentrate, neither phosphates nor feldspars should be important mineral impurities, and therefore, the majority of alkalinity is controlled by spodumene. Given that this was among the highest pH of all recorded samples and the Li/PO_4^{3-} ratio was 192, we suggest that spodumene may be an important contributor to both alkalinity and to dissolved Li. Furthermore, spodumene has been classified as a rapid weathering mineral as compared to the other major pegmatite minerals (albite, K-feldspar, quartz, and muscovite), which are considered slow to very slow weathering (Kwong, 1993; Swanson, 2012). This weathering process typically forms clays, a process observed at the KMM, resulting in the release of LiOH, increasing both dissolved Li and pH (Krainov, 1973; Schwartz, 1937; Tien et al., 1975).

Leaching of the amphibolite waste rock also resulted in alkaline leachates. Here, hydrolysis of silicate minerals and cation exchange on the surface of feldspars (62–70 wt% amphiboles, 30–38 % feldspars) may be a primary mechanism that can explain the elevated pH of water leachates. Sulfide minerals can be found as accessory minerals in these amphibolite rocks (Horton, 2008) and could cause lower pH conditions to develop. However, humidity cell testing of amphibolite host rocks at another LCT pegmatite deposit in Canada showed that ARD was unlikely to develop (CELC, 2020). While the exact mineralogy between different pegmatite ores may differ, the leaching experiments conducted in our study demonstrate the potential high buffering capacity of amphibolite waste rocks.

Given the neutral pH of the one TSF sample, the findings of Roy et al. (2023), and our 25-day leaching experiment showing that leachate pH decreases over time toward circumneutral values, we suggest that tailings materials from other hard-rock Li mines might follow a similar trend. This would imply that the alkalinity detected in the 24-hour leaching tests reflect fresh mineral surfaces interacting with water during the first stages of water-rock interaction, while their buffering capacity would decrease with repeated wetting and drying. Feldspars have typically been considered the source of high pH in waters and leachates of pegmatite mines in South Dakota and Canada (Rahn et al., 1996; Roy et al., 2023, 2022); however, we suggest that both spodumene and phosphate minerals may contribute to alkalinity and Li occurrence in water resources associated with Li ores. While spodumene is a major mineral phase, phosphate minerals are only minor occurrences and may contribute to the early stages of water-rock interactions.

4. Conclusions

This study presents integrated data of the bulk chemical composition of Li-rich pegmatites, waste rock, and tailings, results of leaching experiments, as well as water quality of groundwater and surface associated with a legacy hard-rock Li mine in NC. The results of this study indicate that common regulated contaminants (e.g., As) likely do not pose risks to the quality of water resources around the Li mine, since they were typically found at low concentrations in the bulk rocks and waste solids, require acidic conditions to be mobilized in high proportions, and were not found at concerning levels in the natural waters in the vicinity of the mine. In contrast, elevated concentrations of the rare alkali metals of Li, Rb, and Cs were detected in water resources directly associated with the mine in comparison to natural background levels in common aquifers in the Piedmont region of NC.

Mining inherently exposes fresh mineral surfaces directly through excavation, increased fractures from blasting, and fresh deposition of ground waste material, all of which could be exposed to water (Schultze et al., 2022). As suggested from the water leaching experiments presented in this study and reported in Roy et al. (2022, 2023), trace elements like Li, Rb, and Cs would be most prone to leach out during the first exposures to water before ion exchange sites on mineral surfaces are exhausted. The same understanding may be applied to pH, as H^+ exchange capacity decreases with repeated and continued exposure to water. The findings presented in this study indicate that in the immediate aftermath of mining, crushed rock, and phosphate minerals inparticular might exert a strong influence on water quality affecting the total alkalinity and could be a major source of Li. However, over longer time scales the pH of waters impacted by a legacy Li mine would likely be circumneutral to slightly alkaline, and given the high weatherability of spodumene (Kwong, 1993; London, 2017) this would be controlled by feldspars.

Our data suggest that it would seem unlikely that ARD would develop from water-rock interactions in Li mine areas given the alkaline conditions that developed during the leaching experiments and the pH data detected in natural waters within the mine vicinity. While longer-term leaching experiments show that this buffering capacity could decrease over time, we would not expect acidic conditions to develop due to the low sulfide mineral content of the rock units investigated in the study (Horton, 2008; Swanson, 2012). Furthermore, ARD has not been recorded in the TSB geological area or associated with the historic lithium mines at the KMM or the Hallman-Beam Mine (HBM), another legacy Li mine in the TSB region (Fig. 1B). Additionally, the pH of Long Creek a drainage adjacent to the HBM has been regularly monitored during and after the period when the HBM was operational and consistently maintained a pH between 6 and 8, never recording acidic conditions (National Water Quality Monitoring Council, 2021). Furthermore, the neutral to slightly alkaline properties of pegmatites and tailings from several global LCT pegmatite mines further indicate that ARD is unlikely to result from hard-rock lithium mining (Lehmann et al., 2014; Ndoli et al., 2013; Rahn et al., 1996; Roy et al., 2024, 2023, 2022).

The results presented in this study are consistent with previous water quality reports globally from Li pegmatite mines, where water quality is characterized by elevated pH and Li concentrations (Gao et al., 2021; Rahn et al., 1996; Roy et al., 2022). Furthermore, at many historic LCT pegmatite mines, other elements like Sn or Be and not Li were the commodity of interest, indicating that Li bearing minerals may still occur in historic mine wastes (Bradley et al., 2017a; London, 2008). At the KMM, spodumene in tailings wastes from historic mining are reported at 5.1 wt% (Browning et al., 1964), while 1–12 wt% spodumene were found in tailings from legacy pegmatite mines in Russia and in Canada (Chen et al., 2022; Roy et al., 2022; Yusupov et al., 2015). These solid wastes could be a major source of Li and possibly Rb and Cs to water resources near legacy LCT pegmatite mines.

We conclude that monitoring for the occurrence of Li, Rb, and Cs in water resources near LCT pegmatite deposits from legacy and new Li mines should be a focus of future assessments on the impacts of hard-rock Li mining to water resources. Further understanding the role of phosphate and spodumene minerals in contributing to alkalinity and Li in waters at LCT pegmatite deposits could be important for constraining their influence on water chemistry.

CRediT authorship contribution statement

Gordon D.Z. Williams: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Sam Saltman:** Methodology, Investigation, Data curation. **Zhen Wang:** Supervision, Methodology, Investigation, Data curation. **D. Morgan Warren:** Writing – original draft, Validation, Investigation. **Robert C. Hill:** Methodology, Investigation, Formal analysis. **Avner Vengosh:** Writing – review & editing, Writing – original draft, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

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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Appendix A. Supplementary data

Supporting Information (SI) includes 10 Supplemental figures (S1 to S10) and 2 supplemental tables in a separate excel file (Tables S1 and S2). Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.177281.

Data availability

All data are included in the paper

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