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The Water Quality Impacts of Legacy Hard-Rock Lithium Mining and Processing

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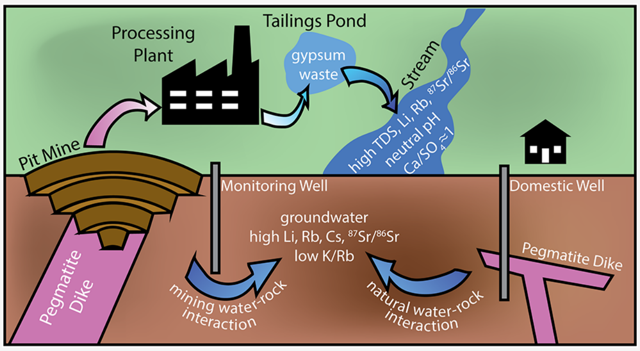

ABSTRACT: Hard-rock Li mining from Li-pegmatites is rapidly expanding to support the clean energy transition, yet associated water-quality impacts remain understudied. Here, we evaluate surface water and groundwater in the Tin-Spodumene Belt (TSB), a legacy Li mining and processing district in North Carolina. Streams emerging from legacy Li mining and processing sites show markedly elevated total dissolved solids (TDS), Li, and Rb, and circumneutral pH, but no exceedances of regulated contaminants. These solute increases are attributed to gypsum dissolution from historical processing wastes. Groundwater across the TSB exhibits widespread enrichment in Li, Rb, and Cs due to natural pegmatite water-rock interaction, although groundwater within a mine shows greater degrees of interaction. A geospatial analysis reveals no effects on quality of domestic wells located near the historic Li mining and processing. Our results indicate that the waste gypsum residues from Li processing drive the long-term surface water quality impacts, while groundwater chemistry is governed by natural geochemical processes between the Li pegmatite and its host rocks. The global abundance of Li pegmatite deposits and the increasing demand for Li exploration suggests that similar water quality issues may become widespread as new mines and processing facilities open and expand.

KEYWORDS: *pegmatite, critical mineral, arsenic, water-rock interaction, pit mine, gypsum*

1. INTRODUCTION

Lithium (Li) is among the most important elements to the clean energy transition for its use in Li-ion batteries for electric vehicles and grid storage.[1,2] Production of Li is expected to increase 13- to 42-fold by 2040 over 2020 levels, and consequentially, increased mining efforts are needed.[1,2] This heightened demand has spurred increased exploration and mining activities around the world including at hard-rock Li deposits[3,4] known as Lithium−Cesium-Tantalum (LCT) pegmatites,[5] which account for more than 50% of global Li production.[6,7] Since LCT pegmatites are globally distributed (Figure 1) they are projected to become increasingly important sources of Li as new deposits are found and new mines open.[8,9] The environmental impacts of mining hard-rock Li deposits may thus likely become globally widespread in impacted areas.

While Li exploration has increased during the past decade, the number of active mines across the globe is still relatively small. Therefore, this early stage of Li exploration provides a unique opportunity to investigate the potential environmental outcomes of hard-rock Li mining before the onset of large-scale operations projected globally. Since many other mined metals and materials produced from large-scale hard-rock operations (e.g., coal, copper, iron) have shown extensive negative impacts on the environment,[10,11] this has become an important topic of debate for the green energy transition.[1,12] Yet it is important to note that the geology and geochemistry of most of these deposits are distinct from that of hard-rock Li deposits. For example, the adverse environmental impacts of mining are largely associated with the occurrence and oxidation of sulfide minerals that produce acid mine drainage that leaches various metals into water resources.[11,13,14] However, such sulfide minerals are typically minor in pegmatite ores and wastes, which are instead dominated by felsic minerals like feldspars and quartz.[15,16] This would suggest that the environmental impacts of hard-rock Li mining are less likely to follow the common environmental paradigms typically associated with hard-rock mining.

Most studies on the environmental impacts of hard-rock Li mining have focused on the types and quantity of waste generated (e.g.,[15,17]) and the carbon and water footprints of the Li-ion battery supply chain (e.g.,[18−20]). Studies on the

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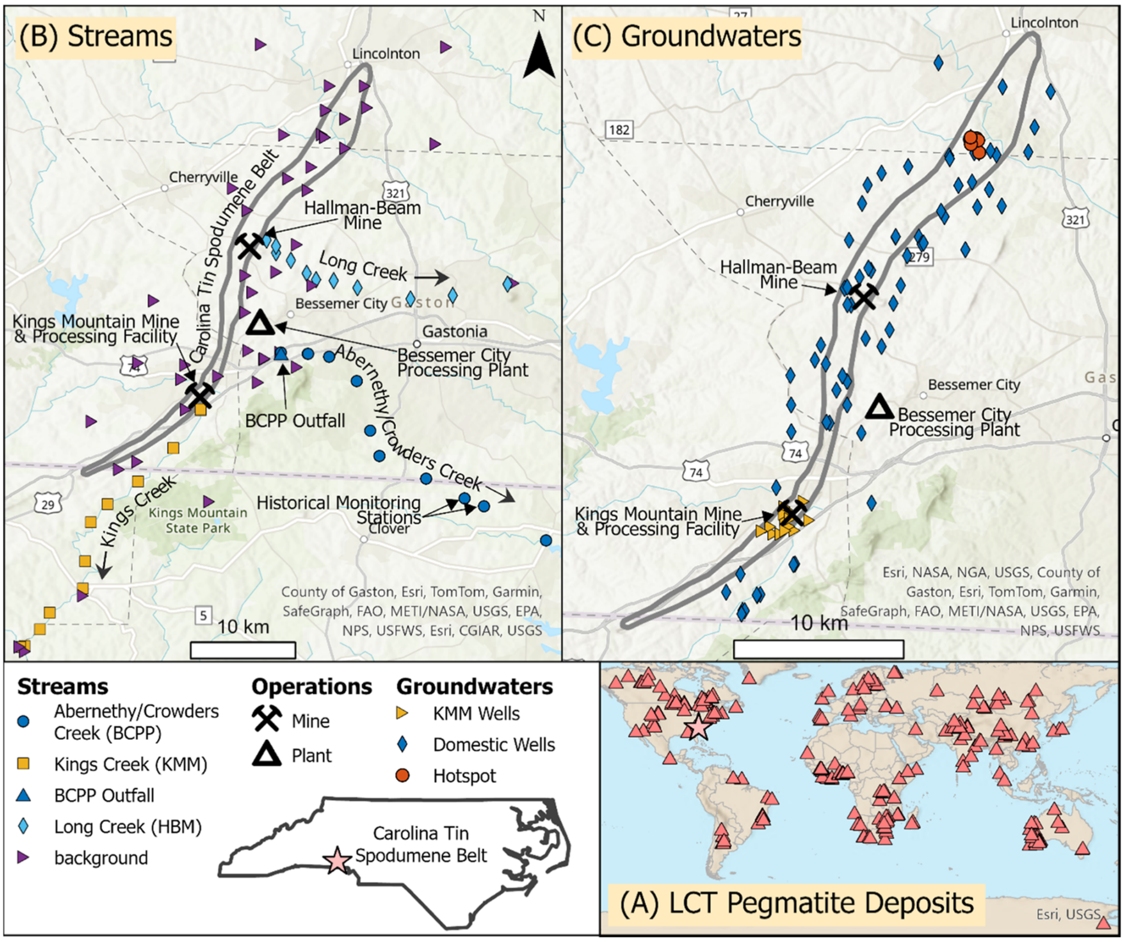

Figure 1. (A) Map of global LCT pegmatites which are found throughout the world. Data from ref 4. (B−C) Maps of stream and groundwater samples collected from throughout the TSB for this study and their relationship to different features discussed in the text. The TSB is shown as the gray outline highlighting the approximate extent of where LCT pegmatite dikes occur, modified from ref 56.

water quality impacts of LCT pegmatite mining, however, have been limited and primarily focused on leaching studies of ores and waste materials[21−28] or water quality parameters, including trace metal concentrations, within and surrounding Li mines and deposits[21,27,29−36] and processing regions.[37] A focus of these studies has been on the occurrence and concentration of Li in waters since it is currently an unregulated constituent in drinking water, but nonetheless, is found in relatively high concentrations around Li mining and processing regions.[27,37] Rubidium (Rb) and cesium (Cs) co-occur with Li in LCT pegmatites[38] and have also been reported in elevated concentrations in waters around mining and processing facilities.[27,33,37] Recent work has, however, tried to characterize baseline Li concentrations in drinking waters and to establish preliminary recommended thresholds;[39−43] Rb and Cs, however, remain under investigated and unregulated despite their co-occurrence in other Li deposit types (e.g., closed-basin brines,[44] volcano sedimentary basins,[45] and geothermal waters[46]). Previous studies on hard-rock Li deposits and mines, however, have not developed geochemical fingerprints to link elevated Li concentrations and other variations in water quality to mining and processing operations and cannot differentiate these from natural processes. Thus, establishing the ultimate source of variations in water quality remains uncertain, which is the focus of this study.

While Li is not currently regulated as a contaminant, the US Environmental Protection Agency (USEPA) has added it to the Drinking Water Contaminant Candidate List[47] reflecting both its increasing environmental prevalence from anthropogenic activities[48] and the lack of scientific consensus on the health impacts of chronic exposure to low Li concentrations.[49] Despite this, various studies (e.g.,[50−53]) have provided evidence that Li contamination in humans and ecosystems can lead to adverse health and ecological outcomes. Additionally, Li is a highly fluid mobile element prevalent at varied concentrations in water systems[39,43] and is not effectively removed by water treatment plants.[37,43] Furthermore, the increasing mining and production of Li and the end-of-life recycling and disposal of Li-ion batteries have been suggested as potentially new and significant sources of Li fluxes to water resources.[54,55] Reflecting these concerns, the US Geological Survey (USGS) has proposed a nonregulatory health-based screening level (HBSL) of 10 μg/L Li that is commonly used as a threshold to detect potential Li contamination.[41]

Here we investigate groundwater and surface water throughout the largest legacy Li mining region in the USA, the Carolina Tin Spodumene Belt (TSB) in North Carolina, which still has active Li chemical processing facilities, aiming to evaluate possible water quality impacts from mining and processing. This region with the legacy of Li mining provides a unique opportunity to evaluate how water-rock interactions with Li-rich pegmatites in mines and the surrounding area have affected the quality of nearby surface water and groundwater. We hypothesize that the exposure of rocks in legacy Li mines increases water-rock interactions with ambient waters and thus intensifies water quality impacts. The objective of this study is to develop geochemical tools that can help to delineate between naturally occurring contaminants derived from water-rock interactions with the Li-rich pegmatite and surrounding lithologies from the anthropogenic impacts associated with Li mining and processing. Here, we demonstrate how the unique geochemistry of hard-rock Li deposits naturally influences the quality of associated water resources, and how legacy processing wastes have a direct impact with a distinctive geochemical fingerprint on the quality of associated surface waters.

2. METHODS

2.1. Site Descriptions and Sampling. The TSB is an ~40 km long belt between Gaffney, South Carolina and Lincolnton, North Carolina (Figure 1).[57] Throughout the TSB, LCT pegmatites occur as hundreds of individual dikes of varied sizes and mineral composition, which have intruded into the local metamorphic rocks, primarily into amphibolite gneiss and mica schist rock units.[58−60] Because of this, the occurrence of pegmatite dikes is not consistent throughout the TSB[58] and the locations of many dikes are still unknown and remain unmapped. A few clusters of dikes have been identified throughout the TSB, which have given rise to two mines with currently existing open pits, the Kings Mountain Mine (KMM), and the Hallman Beam Mine (HBM) (Figure 1). Both ceased mining operations in the late 1980s to early 1990s.[61] Today both open-pits remain nonoperational and have become pit lakes; however, both mines have active aggregate quarries adjacent to them.

In addition to mining, the KMM had a Li processing facility extracting Li from spodumene and the HBM had one offsite at the nearby Bessemer City processing plant (BCPP) (Figure 1).[62] Today neither facility extracts Li from spodumene (operations ceased in the 1990s)[63,64] but do still convert Li-carbonate feedstock from brine operations in Nevada, Chile, and Argentina into other Li products (e.g., Li metal, LiOH).[3] Additionally, each location has the legacy of mining and processing with various nearby tailings storage facilities and waste-rock piles.

The headwaters of Kings Creek are within the mine site of the KMM and the adjacent aggregate quarry and it is the only drainage from the KMM; legacy tailings ponds do spill into Kings Creek.[65] Kings Creek was sampled at its exit from the mine and at regular intervals along its 33 km course before flowing into the Broad River (n = 12 samples over 3 sampling campaigns) (Figure 1). At the HBM, an unnamed tributary to Long Creek flows adjacent to the legacy tailings ponds. This stream was sampled both upstream and downstream of the HBM and along its flow for ~21 km in Long Creek until its discharge into the South Fork of the Catawba River (n = 21 samples over 4 sampling campaigns). At the BCPP, an outfall drains a tailings pond and discharges into Abernethy Creek. This discharge was sampled and is termed the BCPP outfall (n = 4 over 4 sampling campaigns). Abernethy Creek was sampled upstream, downstream, and along its flow into Crowders Creek and eventually into Lake Wylie ~35 km downstream (n = 23 over 4 sampling campaigns).

In total, 99 stream samples were collected in this study (including the stream profiles) from throughout the TSB and surrounding areas over a three-year period, covering different seasons (summer, winter, spring). These surface waters can be categorized as (1) background samples (n = 51) without a direct connection to mining or processing activities, and (2) stream waters that are downstream (n = 48) of mining and processing activities.

Groundwater samples were also collected from throughout the TSB and surrounding area (n = 93 across 6 sampling campaigns). The groundwater samples were primarily collected from domestic wells (n = 79) with additional samples from monitoring wells (n = 14) within the KMM site as described in Williams et al.[27] Well depths ranged from ~10−170 m with a median depth of ~60 m, however, depth data were not available for all wells. The geologic units associated with each well were determined by location from the North Carolina geologic map,[66] however since the pegmatite dikes are largely unmapped wells cannot be spatially associated with it and are instead associated with the larger geologic units. Some samples from the KMM wells are however known to be associated with the Li pegmatite.[27]

2.2. Analytical Methods. All water samples were collected following US Geological Survey (USGS) standard protocols[67] and were filtered with 0.45 μm mixed cellulose ester syringe filters. For trace element and cation analysis, samples were collected in acid washed HDPE bottles and acidified with HNO_3 (Fisher Optima grade). For anions, alkalinity, and strontium (Sr) isotopes, samples were collected in washed HDPE bottles without headspace. For groundwater sampling, wells were flushed through continuous pumping until physical field parameters were stabilized. Groundwater samples were collected directly from the wellhead.

Trace elements, Li, Rb, Cs, Sr, uranium (U), thallium (Tl), beryllium (Be), arsenic (As), antimony (Sb), barium (Ba), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), and selenium (Se) were measured with a Thermo Fisher X-Series II inductively coupled plasma mass spectrometer (ICP-MS).[27] Major cations, sodium (Na), potassium (K), magnesium (Mg), and calcium (Ca) were measured on a Thermo Scientific Aquion ion chromatograph and anions, fluoride (F), chloride (Cl), bromide (Br), nitrate (NO_3), sulfate (SO_4), and phosphate (PO_4) on a Dionex DX-2100 ion chromatograph.[44,68] Bicarbonate (HCO_3) was determined by alkalinity titration to pH 4.5 with 0.02 M HCl.[27]

For Sr-isotope measurements, ~500−1000 ng of Sr were purified using Sr-specific resin (Eichrom) and analyzed with a ThermoFisher Triton thermal ionization mass spectrometer (TIMS) at Duke University.[69,70] NIST SRM 987 was measured repeatedly to monitor analytical accuracy and reproducibility with an average of 0.710252 ± 10 (n = 71), in good agreement with accepted values.[71]

All new data are available in Tables S1 (streams) and S2 (groundwater), however the geographic location of private well waters have been randomly altered up to 0.5 km to protect the privacy of well owners.

3. RESULTS AND DISCUSSION

3.1. Surface Water. Water quality of all surface waters in the TSB region, including background streams and those downstream of the three mining and processing facilities (KMM, HBM, BCPP) (Figure 1) revealed low concentrations of contaminants regulated by the USEPA. Contaminants that

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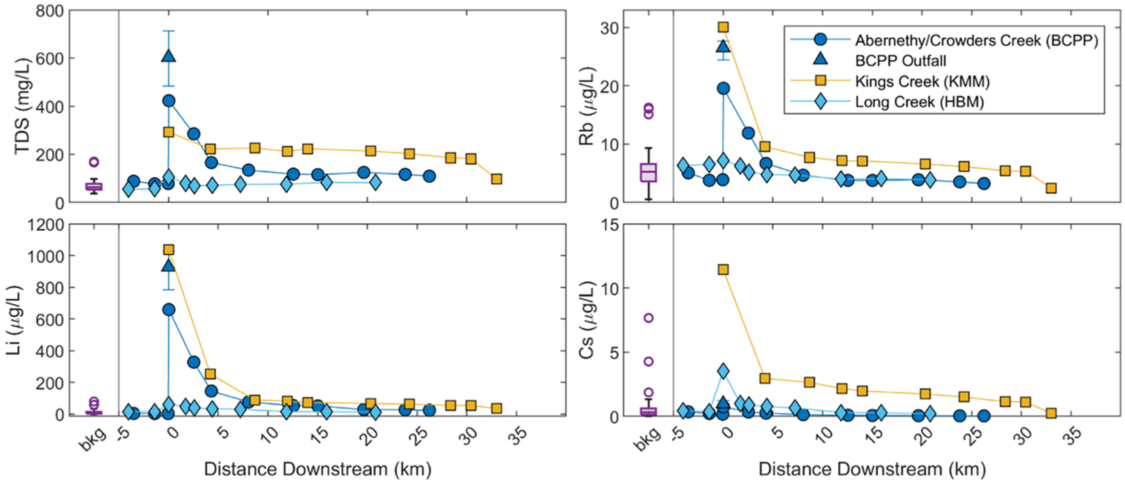

Figure 2. Stream transects of (1) the BCPP outfall discharges to Abernethy/Crowders Creek; (2) Kings Creek drains the KMM mine site and processing facility; and (3) Long Creek passes by the HBM mine site. The boxplot shows the distribution of TDS, Li, Rb, and Cs in the background streams. A distance of zero is set as the point the stream passes by a mine site or processing facility. Brackets on the BCPP outfall show the range of concentrations measured over four sampling events. Several other points were sampled at multiple times, but the variations are much lower and only the mean concentration is shown. bkg = background.

can occur in LCT pegmatites such as F, U, Tl, Be, and As, as well as all other elements regulated in drinking water (Sb, Ba, Cd, Cr, Cu, Pb, Se, NO_3)[72] were found below the USEPA maximum contaminant levels in all investigated stream waters in this study. Lithium, however, is an unregulated contaminant, previously found in high concentrations in natural waters of the TSB[27] and around other LCT pegmatites[21,23,27,29,33−36] and the USGS has a recommended HBSL for Li set at 10 μg/L.[41] This threshold was exceeded in 39% of background streams and 98% of streams downstream of mining and processing facilities in the TSB region.

Although no streams exhibited exceedances of USEPA drinking water thresholds, stream chemistry is clearly altered downstream of Li mining and processing sites compared to background stream waters. Three stream transects were investigated: (1) Kings Creek, draining the KMM and processing facility; (2) Long Creek, flowing past the HBM; and (3) Abernethy/Crowders Creek, which receives discharge from the BCPP Li processing outfall (Figure 1).

Background streams are characterized by low TDS (∼36−170 mg/L) and moderate concentrations of Li (0.2−79 μg/L), Rb (0.6−16 μg/L), and Cs (<0.01−7.7 μg/L) (Figure 2). In contrast, the BCPP outfall shows elevated TDS (483−713 mg/L, $n = 4$), Li (785−1027 μg/L, $n = 4$), and Rb (24−28 μg/L, $n = 4$), with relatively low Cs (0.86−1.2 μg/L, $n = 4$). These elevated concentrations persist after the outfall in Abernethy/Crowders Creek for ∼ 5−10 km downstream before dilution to background levels (Figure 2). Kings Creek, draining the KMM site, exhibits similarly high TDS (286−300 mg/L), Li (824−1249 μg/L, $n = 2$), Rb (27−33 μg/L, $n = 2$), and Cs (11−12 μg/L, $n = 2$) immediately downstream of the KMM. These values decline to background levels within ∼10−15 km. TDS and Cs, however, remain elevated until the creek joins the Broad River (∼33 km downstream; TDS ≈70 mg/L, Li ≈3 μg/L, Rb ≈16 μg/L, Cs ≈1 μg/L). Long Creek, which drains the area near the HBM mine, shows modest increases in TDS (from 57 mg/L before up to 106 mg/L after), Li (14 up to 60 μg/L), and Cs (0.4 up to 3.5 μg/L) concentrations as the stream passes the mine site. These concentrations decline with downstream dilution, while Rb remains relatively constant (∼3−6 μg/L). Overall, Li, Rb, and Cs concentrations in these impacted streams are notably higher than those reported around other hard-rock Li mining and processing sites (Figures 2, S1).[27]

Impacted streams were also characterized by neutral to moderately alkaline pH (Figure S1), with no systematic trend downstream. This is slightly higher than the circumneutral pH of background streams (Figure S1). These results align with leaching tests of ores and waste materials from the TSB and other pegmatite regions, which suggest that neutral to alkaline conditions should prevail upon water-rock interactions and that acid mine drainage is unlikely to form from interacting with these rocks.[21,22,27,31] The low concentrations of trace metals such as As, Tl, and U further demonstrate that (1) the typical adverse environmental outcomes associated with acid mine drainage[14,73] are absent for surface water interacting with Li pegmatites and wastes, and (2) the slightly alkaline conditions do not enhance oxyanions release (e.g., As, Cr), which can occur in alkaline mine discharges.[13,73]

Since only Kings Creek and Abernethy/Crowders Creek show large increases in TDS, Li, and Rb, and since these streams drain processing facilities, the primary solute inputs may be attributed to processing activities rather than legacy mining operations. Supporting evidence comes from the elevated Ca and SO_4 concentrations and the molar Ca/SO_4 ratios of ∼1 detected in the BCPP outfall, Kings Creek, and Abernethy/Crowders Creek. Elevated Ca and SO_4, combined with Ca/SO_4 ratio of ∼1, suggests that the streams were impacted by the dissolution of Ca-sulfate ($CaSO_4{\cdot}XH_2O$), hereafter called gypsum. In contrast, background surface waters are characterized by greater Ca/SO_4 ratios, ranging from ∼2−13 (Figure 3). The high Ca/SO_4 ratios in the background waters reflect the typically greater abundance of Ca-rich minerals than sulfide/sulfate minerals in greenschist to amphibolite (mica schist and amphibolite) rock units,[74] such as those surrounding the TSB.[59] Gypsum does not occur naturally in this region.[59] However, the traditional approach to extract Li from spodumene involves leaching with sulfuric acid and neutralization by Ca-(hydroxide/carbonate).[61,75] This forms a waste gypsum byproduct, which has been reported

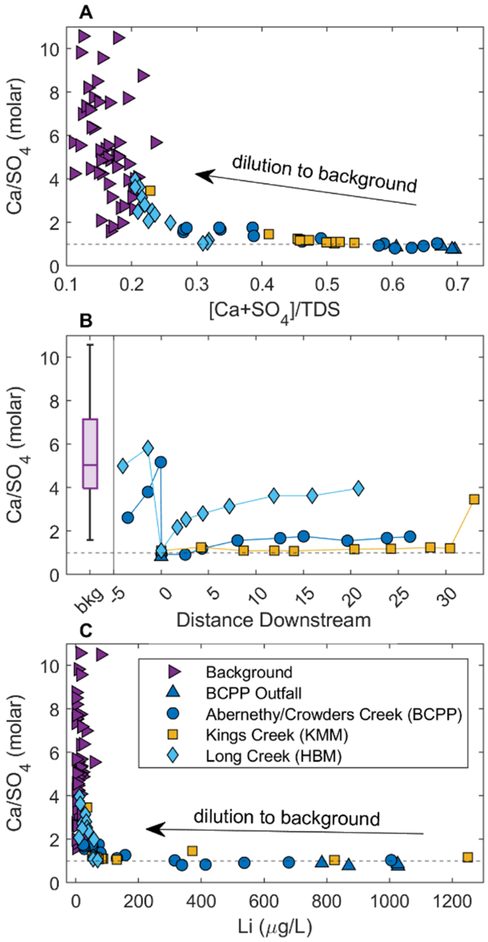

Figure 3. (A) Ca/SO_4 versus $[Ca + SO_4]/TDS$ in streams downstream from legacy Li mines and processing sites. Discharge or leaking of water from tailings ponds produces a Ca/SO_4 ratio ∼1 and high a $[Ca + SO_4]/TDS$ signature due to the dissolution of waste-gypsum produced during the processing of spodumene. Background waters have much higher Ca/SO_4 ratios with and a lower $[Ca + SO_4]/TDS$ signature reflecting natural water-rock interactions with the local silicate-rich geology. (B) Ca/SO_4 ratio versus distance downstream from Li mines and processing sites. The Ca/SO_4 ratio of all streams draining or flowing past mining and processing facilities converges on unity, suggesting that these are the source of the gypsum dissolution signature. bkg = background (C) Ca/SO_4 ratio versus Li concentrations in background streams and streams downstream from legacy Li mines and processing sites. Elevated Li concentrations are associated with waste-gypsum dissolution in mining and processing impacted waters. The dashed line in all graphs is a Ca/SO_4 ratio of 1.

in waste residues from processing of various Li-pegmatite ores.[28,76−78] Additionally, Na-sulfate ($Na_2SO_4 \cdot XH_2O$), hereafter called thenardite, can also form during spodumene processing and has also been reported as a waste by-product.[75,78] This mineral also does not occur naturally around the TSB.[59] Therefore, we posit that in the TSB region, gypsum and thenardite and their dissolution products should only be found in relation to wastes from Li extraction processes and consequently, can be an indicator for anthropogenic activities that affect water quality.

Given the relatively high solubility of gypsum and thenardite compared to the silicate minerals that dominate the local geology, their dissolution should leave a clear chemical signature in waters impacted by processing activities, whereas background waters should primarily reflect interaction with the local silicate-rich bedrock (pegmatites, amphibolites, mica schists, granites). This is evident in Figures 3 and S2, where streams affected by Li processing (Kings Creek and Abernethy/Crowders Creek) show both Ca/SO_4 ratios and $(Ca+ 1/2Na)/SO_4$ ratios ∼1 and derive large fractions of their TDS from dissolved Ca and SO_4 (i.e., high $[Ca + SO_4]/TDS$). In the BCPP outfall, the Ca/SO_4 ratios were just below 1 (∼0.76−0.90), while the $(Ca + 1/2Na)/SO_4$ ratios were slightly higher and closer to 1 (0.83−1.00), suggesting that thenardite and gypsum are both likely dissolving and control the stream chemistry. This is however less clear in Kings Creek and Long Creek where Ca/SO_4 ratios are just above 1 nearest the outfall. The Ca/SO_4 ratio near 1 in all impacted streams and the high fraction of Ca and SO_4 contributing to TDS suggests that thenardite dissolution is relatively minor in comparison to gypsum. Therefore, the elevated salinity of these streams primarily reflects gypsum dissolution and only Ca/SO_4 and $[Ca + SO_4]/TDS$ will be discussed in the following sections.

By contrast, background surface waters are clearly distinguishable and had higher Ca/SO_4 ratios with a much lower contribution of Ca and SO_4 to TDS (i.e., low $[Ca + SO_4]/TDS$). A few samples from Long Creek that has no Li processing facility also show Ca/SO_4 ratios close to 1 and intermediate $[Ca + SO_4]/TDS$ values, suggesting a possible influence from historical BCPP processing wastes that may have been impounded at this site when it was operated by the same company.[61] Stream profiles for all three impacted streams further support this interpretation, where Ca/SO_4 ratios are higher upstream, but converge on 1 at mine or processing facility discharge points before increasing again downstream as background inflows dilute the gypsum dissolution signal (Figure 3B).

Elevated Li concentrations downstream of mining and processing sites are consistently associated with $Ca/SO_4 \approx 1$ (Figure 3C), indicating that Li may be released during gypsum dissolution, where it is entrapped in waste residues. Similar but weaker trends are observed for Rb, and to a lesser extent Cs, U, and F, which remain well below USEPA drinking water thresholds (Figure S3), suggesting that waste-gypsum dissolution elevates these elements above natural background levels and yet not above regulatory levels.

It is important to note that processing of hard-rock ores in this region ceased approximately three decades ago, and the elevated Li concentrations observed today reflect the long-term release of Li from legacy waste residues. Historical water-quality monitoring data[79,80] for stations along Abernethy/Crowders Creek, located ∼ 25 km downstream of the BCPP (Figure 1B), show that between 1976 and 1993, during the period of active hard-rock ore processing, Li concentrations were substantially higher, ranging from 230 to 940 μg/L (no contemporary Ca or SO_4 concentrations data were reported). In comparison, our present measurements at the same location are ∼25 μg/L. These historical values are similar to those that we measured directly at the BCPP outfall (785−1027 μg/L), despite being recorded 25 km downstream and presumably diluted during transport. This indicates that (1) Li concentrations in processing outfalls during historical active ore processing were likely far higher than those currently observed, and (2) that the release of Li from processing wastes can persist for decades after operations have ceased.

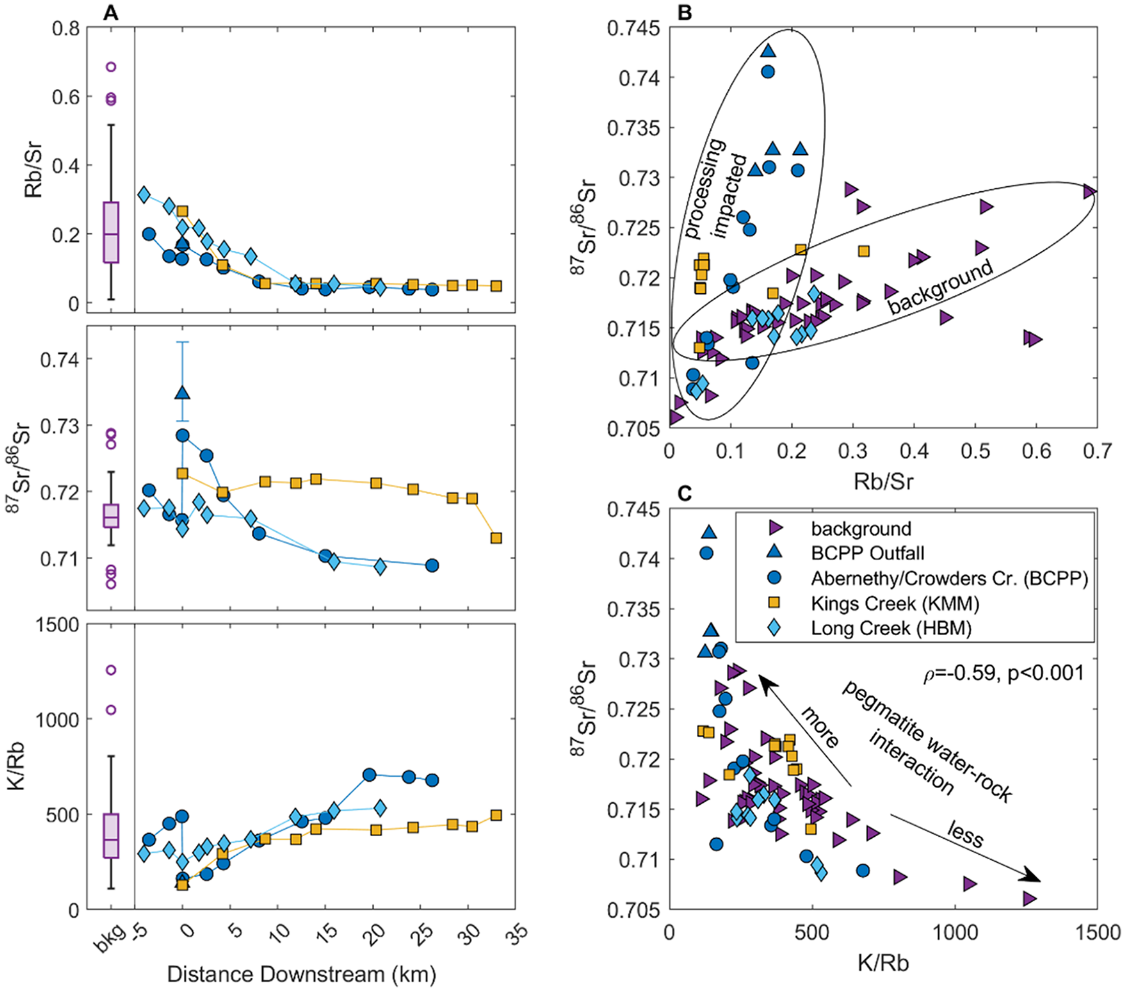

Figure 4. (A) Variations of Rb/Sr, ^{87}Sr/^{86}Sr, and K/Rb ratios in surface waters versus distance of the streams downstream from Li mines and processing sites. The stream profiles assessing the effectiveness of Rb/Sr, ^{87}Sr/^{86}Sr, and K/Rb ratios in evaluating the direct impacts of pegmatite mining on water quality. Boxplots show the range of background (bkg) values. (B) ^{87}Sr/^{86}Sr versus Rb/Sr in surface waters. Evaluation of Rb/Sr and ^{87}Sr/^{86}Sr demonstrating that gypsum dissolution from processing wastes has added Sr to the gypsum wastes and lowered the Rb/Sr and presumably also the ^{87}Sr/^{86}Sr signal, but still distinguishing them from background values. The waters from Long Creek flowing past the HBM without a processing facility have values overlapping with background streams. (C) ^{87}Sr/^{86}Sr versus K/Rb in surface waters. The inverse correlation between ^{87}Sr/^{86}Sr versus K/Rb in the streams reflects the magnitude of the water interaction with the pegmatite rocks characterized with high ^{87}Sr/^{86}Sr and low K/Rb ratios.

Seasonal and weather-related variability can influence elemental concentrations. Although this was not the primary focus of this study, we observed some temporal variations in the concentrations of regularly monitored streams. The most notable example is the BCPP outfall, which was sampled four times between 2022 and 2025, showing Li concentrations ranging from 785 to 1027 μg/L. These variations do not correspond to expected seasonal trends (e.g., dilution during wetter summer months). Instead, the two highest Li concentrations coincided with rainfall events within 3 days prior to sampling (0.8−1.4 cm cumulative precipitation), whereas the lowest values occurred during periods of little to no rainfall (<0.03 cm) (Figure S4). Although this pattern is somewhat counterintuitive, since increased precipitation would typically dilute solute concentrations, it may reflect enhanced discharge from tailings ponds as water levels rise following rainfall. By comparison, the background sample location immediately upstream of the outfall showed little variation consistently ranging from ∼2−3 μg/L.

While gypsum dissolution provides a clear signal of processing wastes from hard-rock Li ores, it does not directly trace mining impacts. LCT pegmatites are geochemically distinct from the surrounding regional geology because they formed through extreme fractional crystallization of granitic magmas.[81] This process drives strong elemental fractionation where incompatible elements (e.g., Rb) become concentrated in the pegmatite, while compatible elements (e.g., Sr) are removed earlier during crystallization. As a result, pegmatites are enriched in Rb, depleted in Sr, and show preferential Rb enrichment over K, yielding distinctly high Rb/Sr and low K/Rb ratios.[82] Furthermore, the extremely high Rb/Sr ratios allow radiogenic ingrowth of ^{87}Sr from ^{87}Rb decay, producing highly radiogenic ^{87}Sr/^{86}Sr ratios.[83,84] The TSB pegmatites are no exception and are known to have high Rb/Sr (7−304), low K/Rb (29−43), and highly radiogenic ^{87}Sr/^{86}Sr ratios (0.8822−4.3369), in sharp contrast to the local host rocks, which have much lower Rb/Sr (0.02−0.27), higher K/Rb (75−653), and lower ^{87}Sr/^{86}Sr (0.7073−0.7111).[27,84−86] These geochemical ratios can therefore serve as indicators of water-quality impacts linked directly to pegmatite signatures or mining activity.

Our analysis of Rb/Sr and $^{87}Sr/^{86}Sr$ ratios in surface waters reveals that Rb/Sr ratios in all the impacted surface water samples overlap with those of background streams and did not appreciably increase in any stream profile near mine sites or processing facilities (Figures 4A, S1). Rb/Sr ratios did, however, decrease with the flow of streams away from the TSB into regional geology characterized by much lower Rb/Sr values. In contrast, $^{87}Sr/^{86}Sr$ ratios in the BCPP outfall is distinctly high (radiogenic, 0.7306−0.7425) and this signature was transmitted downstream into Abernethy/Crowders Creek before being diluted to background values by inflows (Figure 4A). Although no outfall was directly sampled at the KMM, a similarly elevated $^{87}Sr/^{86}Sr$ signature was detected in Kings Creek and persisted throughout its run to the Broad River, although values remained near the upper limit of the background range (Figure 4A). By comparison, $^{87}Sr/^{86}Sr$ values in Long Creek did not change appreciably as it passed the HBM site. Overall, our data show that Rb/Sr ratios in impacted streams are not distinct from background streams, whereas $^{87}Sr/^{86}Sr$ ratios could be a useful geochemical tracer, specifically for the BCPP outfall. We suggest that the muted Rb/Sr signal likely reflects additional Sr input during industrial gypsum formation, where Sr substitutes for Ca in Ca-(hydroxide/carbonate). This addition of Sr would reduce Rb/Sr ratios in wastes and residues. This would also result in modification toward a lower $^{87}Sr/^{86}Sr$ ratio, since most industrial Ca-(hydroxide/carbonate) is derived from marine limestones,[87,88] which have much lower marine like $^{87}Sr/^{86}Sr$ ratios.[89] This interpretation is confirmed by plotting Rb/Sr against $^{87}Sr/^{86}Sr$ (Figure 4B); waters downstream of processing sites plot above the background trend, reflecting Sr addition and a lowered Rb/Sr from gypsum dissolution. Importantly, Long Creek waters plot with background samples, reflecting the absence of a processing facility and minimal influence from gypsum dissolution at this site, which is consistent with the lower TDS and Ca and SO_4 contents of that stream.

Despite the influence of gypsum-derived Sr on $^{87}Sr/^{86}Sr$, the conservative behavior of Sr isotopes and the highly radiogenic signature of pegmatites infers that this isotope ratio is a reliable tracer of water-rock interactions with pegmatite minerals. This is evident in the elevated $^{87}Sr/^{86}Sr$ values in Kings Creek and Abernethy/Crowders Creek reflecting the impact of pegmatite rocks on the streamwater geochemistry (Figure 4A−B).

In contrast, we posit that the K/Rb ratio should be relatively independent of gypsum and primarily controlled by regional geology, with pegmatites characterized by distinctively low values in comparison to the ratios in regional rocks.[82,85] In stream profiles, the lowest K/Rb ratios indeed occur where streams pass mining and processing sites, and yet these ratios were overlapping with the lowest values detected in background streams (Figure 4A). This indicates that both background and mine-impacted waters are strongly influenced by water-rock interactions with pegmatites composed of low K/Rb ratios.

When K/Rb is plotted against $^{87}Sr/^{86}Sr$, all samples, both background and downstream of processing/mining, fall on an inverse correlation, where low K/Rb corresponds to high $^{87}Sr/^{86}Sr$, even with the influence of gypsum dissolution (Figure 4C). Low K/Rb and high $^{87}Sr/^{86}Sr$ indicate stronger pegmatite influence, while the opposite pattern reflects greater contributions from regional host rocks. Most waters downstream of mining and processing facilities plot in the middle of this trend, overlapping with background streams, demonstrating that there is no evidence that mining has led to greater pegmatite water-rock interaction than that already present in background waters. This indicates in turn that downstream waters show little to no additional geochemical modification that could be induced from legacy pegmatite mining, apart from processing wastes that elevate Ca and SO_4.

Taken together, the geochemical tracers presented in this study demonstrate that the impacts of Li processing at both the KMM and BCPP facilities likely stem from legacy operations and not from current contributions of Li processing. Neither facility currently processes hard-rock ores[63,64] but rather are importing brine-derived Li carbonate from Chile, Argentina, and Nevada for refining into products such as Li hydroxide.[3] The distinction between legacy hard-rock mining and processing and current Li processing is demonstrated by two geochemical signatures: (1) gypsum dissolution, which only occurs from hard-rock processing, and (2) distinctly high $^{87}Sr/^{86}Sr$ ratios in the BCPP outfall and in Kings Creek. By contrast, Sr in brine-derived Li carbonate feedstocks would have much lower Sr isotope ratios (<0.7200).[90−92] In addition, all waters downstream of mining and processing sites show relatively low K/Rb ratios, consistent with influences from the pegmatite but not distinct from background waters, demonstrating the pervasive influence of the pegmatite on waters in the region. Overall, we suggest that surface water impacts of legacy Li mining and processing are driven primarily by the dissolution of processing wastes such as waste-gypsum that were historically generated. This leads to increases in downstream salinity, while the direct impacts of mining are related to water-rock interactions with the Li pegmatite that are not distinct from the composition of natural background. Besides gypsum, waste residues also include aluminosilicates from spodumene decomposition, which can also be enriched in Li, Rb, and Cs.[28,76−78,93] These waste residue phases may explain elevated concentrations of these metals in downstream waters (Figure 4C). We rule out the impact of modern processing discharge of Li due to the different Sr isotope composition of Li brines. Further support comes from the consistent association of elevated Li with Ca/SO_4 ratios ∼1 (Figure 3C), indicating that processing waste-residues are a major source of Li and other solutes to impacted waters. In summary, we show that streams impacted by legacy lithium mining and processing are slightly saline, have circumneutral pH, high Li, Rb, and Cs and no elevated concentrations of regulated contaminants. Furthermore, streams impacted by processing wastes have distinctive geochemical fingerprints (Ca/SO_4 ∼1, high $^{87}Sr/^{86}Sr$) that are different from that of background streams.

3.2. Groundwater.

This study evaluates the quality of groundwater collected from domestic wells within the TSB region (Figure 1) and from monitoring wells within the KMM mine site reported by Williams et al.[27] Groundwater from the KMM site have Li concentrations ranging from 4.5 μg/L to 47 mg/L, Rb from 0.1 to 169 μg/L, Cs from <0.1 to 21 μg/L, and TDS from 35 to 630 mg/L, with the highest values detected in wells near tailings sites and/or from relatively new boreholes as described in Williams et al.[27] By comparison, groundwater from domestic wells throughout the TSB region (Figure 1) had Li concentrations ranging from 0.5 to 890 μg/L, Rb from 0.1 to 17 μg/L, Cs from <0.1 to 66 μg/L, and TDS from 25 to 317

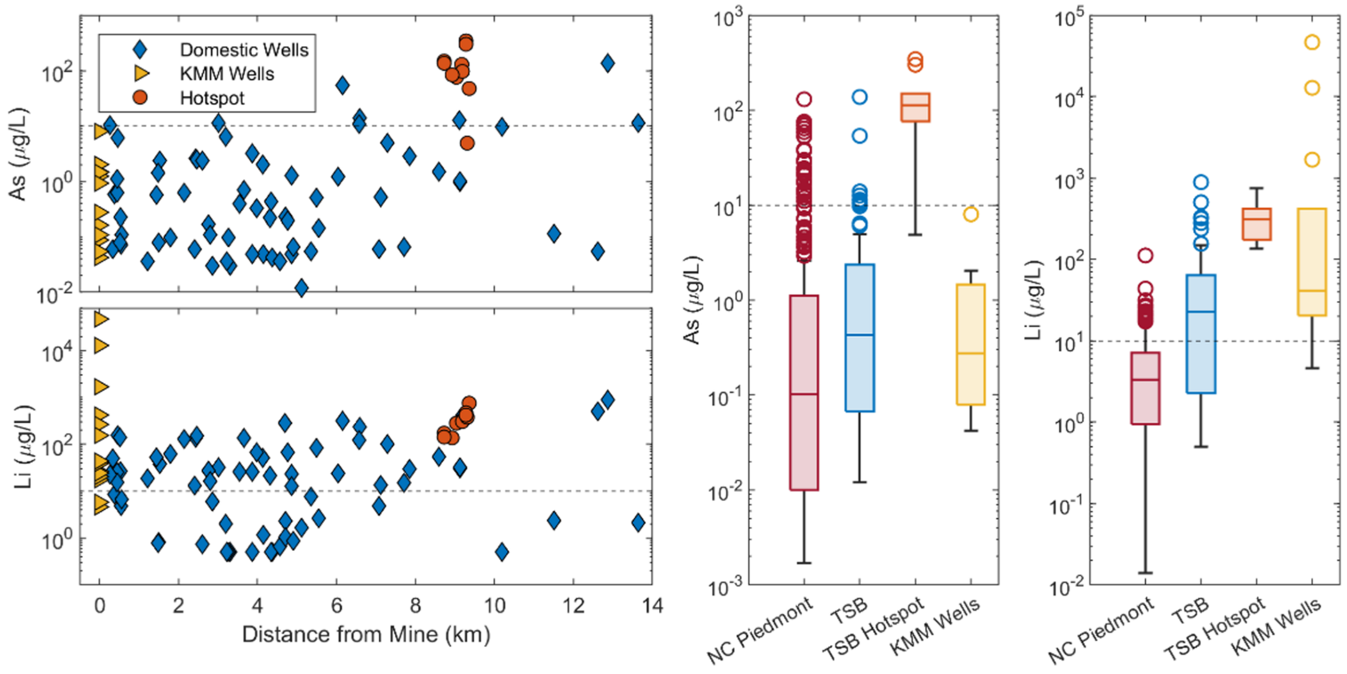

Figure 5. As and Li concentrations in groundwaters from the TSB region versus distance to the nearest legacy Li mine or processing plant (left) and in box plots for groundwater in KMM site, hot spot cluster, across the TSB region, and NC Piedmont aquifers (right). Elevated As was found in the hot sport cluster, while As in the TSB and KMM were similar to the variations of the NC Piedmont.[95] Li concentrations are elevated throughout the TSB and highest in the KMM and hotspot groundwaters. Dashed lines represent the USEPA MCL for As of 10 μg/L and the USGS HBSL for Li of 10 μg/L.

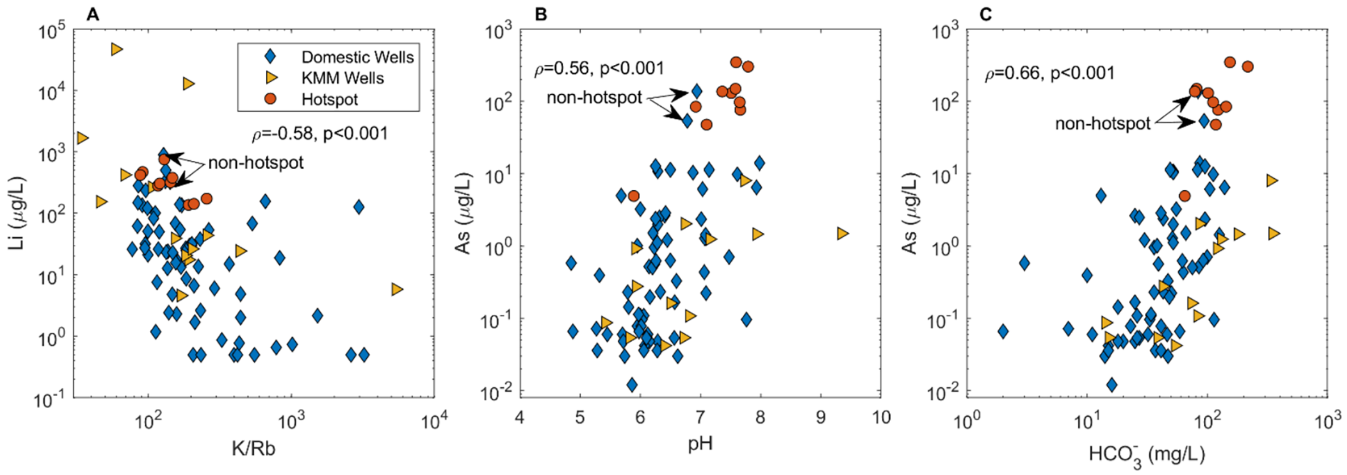

Figure 6. (A) K/Rb vs Li in groundwaters from throughout the TSB where lower K/Rb and higher Li concentrations indicate greater water-rock interaction with the pegmatite. This is demonstrated by several samples from monitoring wells at the KMM with both the highest Li and lowest K/Rb and by the samples from the hotspot which have the same signature of pegmatite water-rock interaction. (B and C) As concentrations are a function of pH and bicarbonate (HCO_3^-) concentration. Samples from the hotspot have both the highest As concentrations and some of the highest pH and bicarbonate concentrations while samples from the KMM have similar pH and bicarbonate but much lower As. Samples listed as nonhotspot have similar chemistry as those in the hotspot and are ~ 4−5 km away.

mg/L (Figures 5, S5). Comparing these Li variations to the USGS HBSL for Li (10 μg/L),[41] 69% of all well waters exceed this threshold, a far greater exceedance rate than found in domestic and public supply wells elsewhere in the USA.[39] In addition, our data also show a wide range in As concentrations, ranging from <1 to 346 μg/L in the TSB groundwater, with 22% exceeding the USPA drinking water threshold of 10 μg/L.[72] In contrast, groundwater associated with the KMM site show much lower As levels, ranging from <1 to 8 μg/L (Figure 5).[27] Our data set (n = 79) reveals that no other regulated water quality parameters were regularly exceeded in the domestic wells across the TSB region.

Since As is a common concern near mine sites[94] and Li concentrations can be elevated in groundwaters at Li mines,[27] we examine groundwater As and Li concentrations in relation to distance from legacy Li mine sites in the TSB region (Figure 1). Our geospatial analysis of the domestic wells across the TSB region reveals that neither As nor Li show any relationship with distance from mine sites or processing plants (Figure 5). We have, however, identified a "hotspot" ~9 km north of the closest mine (HBM), where the highest concentrations of both As and Li occur in a well cluster (Figures 1 and 5) reaching values as high as 346 and 890 μg/L, respectively. Figure 5 shows how these As and Li concentrations in groundwater from the hotspot cluster compare to other groundwater in the TSB region and throughout the North Carolina Piedmont, demonstrating that As is significantly ($p < 0.001$) more concentrated in the hotspot cluster relative to the KMM groundwater, other domestic groundwaters from the TSB, and the overall NC Piedmont groundwater. Li concentrations in groundwater from the hotspot cluster are also significantly higher than other groundwaters of the TSB and the NC Piedmont, but similar to those of the KMM groundwater. Importantly, aside from this hotspot cluster, As concentrations in wells throughout the TSB region and the KMM groundwater were low in comparison to groundwaters from the NC Piedmont, inferring that As is typically not elevated in groundwater from the TSB region.

Since the water quality in the hotspot is unique, we further evaluate the mechanisms that could control the unusually high As and Li concentrations detected in these wells. Throughout the NC Piedmont region, elevated As concentrations are well-known to be a water quality issue in groundwater and is typically associated with the geology of the local aquifers.[96] In the area around the TSB these As anomalies have been associated with a local mica schist rock unit.[97] Given the same mica schist unit is located within the TSB and associated with the location of the hotspot cluster, we suggest that this rock unit may be the source of the elevated As. However, other nearby wells in the TSB are also located in the same mica schist unit but do not have the same unusually high As concentrations. Uniquely, the high As in the hot spot cluster co-occurs with elevated Li, which could indicate that the local groundwater is also interacting with the Li pegmatites. Using the same K/Rb ratio indicator discussed previously in the surface waters (section 3.1), we can see consistent ratios between groundwater from the hotspot cluster and groundwater from the KMM, which is known to interact with the pegmatite rocks.[27] Consequently, the similar characteristics to what was observed in the KMM groundwaters of relatively low K/Rb ratios and high Li concentrations indicate that water-rock interactions with Li-rich pegmatites also impacted the groundwater in the area of the hotspot cluster (Figure 6). To resolve this apparent mismatch, we posit the groundwater from the hotspot cluster reflects the local aquifer composed of both mica schist and Li pegmatites which have generated a unique groundwater chemistry with the co-occurrence of Li and As anomalies.

In addition to As and Li, the pH and the bicarbonate concentration of the hotspot groundwater are among the highest throughout the TSB and similar to some of the KMM groundwater samples. Throughout the region As concentrations increase with increasing pH and bicarbonate concentration (Figure 6), suggesting greater As mobilization under alkaline conditions. The leaching of As from subsurface solids into groundwater is well-known to increase at higher pH and bicarbonate concentrations,[98−101] and water-rock interactions with pegmatites in the TSB (and elsewhere) have been shown to produce alkaline conditions during silicate hydrolysis.[27,31] Since this hotspot has the geochemical characteristics of relatively intense water-rock interactions with the pegmatite (low K/Rb, high Li, high pH) and mica schist is known to be a source of As, we posit that this region has a subsurface contact of a pegmatite dike(s) intruded into the mica schist. We speculate that the high pH and bicarbonate concentrations generated by interacting with the pegmatite could have induced favorable conditions for increased mobilization of As from the mica schist. In contrast, the groundwater from the KMM also have high pH but low As, reflecting the fact that the pegmatite is intruded into an amphibolite gneiss unit, not associated with high groundwater As, rather than the mica schist unit[27,59,102] resulting in low As groundwater. Most of the other investigated groundwaters from throughout the TSB do not have as strong a signature of pegmatite water-rock interaction as detected in the hotspot (Figure 6) and also have much lower As concentrations even in cases where the wells were located within the mica schist rock unit. We have detected two other wells with the highest As (54 and 138 μg/L) outside of the hotspot cluster in two different locations ∼4−5 km away from the hotspot. These also have the same geochemical characteristics (high pH, high Li, low K/Rb) as those from the hotspot cluster, indicating the presence of the pegmatite, and they are also set in the mica schist unit. This suggests that the same water-rock interactions with the pegmatite have led to elevated pH and bicarbonate and the subsequent mobilization of As from the mica schist (Figure 6). In summary, we suggest that the co-occurrence of As and Li are derived from codependent water-rock interactions that include both the pegmatite and the mica schist rocks, resulting in unusually elevated As and Li in the groundwater hotspot.

Our data consistently show that groundwater chemistry within the TSB region is influenced by extensive water-rock interactions with the Li pegmatites that are distributed in the local aquifers. The Li concentrations in groundwater throughout the TSB are relatively elevated in comparison to the Piedmont groundwater, and Li is inversely correlated with K/Rb ratios (Figure 6). Further evidence for the strong influence of pegmatite related water-rock interactions comes from the clear correlation between Rb/Sr and ^{87}Sr/^{86}Sr observed in the groundwater, where elevated Rb/Sr signatures correspond to uniquely radiogenic Sr isotope ratios (Figure S6) in which the pegmatites are the likely sole source.[27,86,103] For example, groundwater within the legacy Li mine in KMM has extremely radiogenic ^{87}Sr/^{86}Sr ratios (up to 0.762) and high Rb/Sr (Figure S6), demonstrating that the pegmatites have a strong influence on the chemistry of groundwater throughout the TSB region.

In the earlier section we show that gypsum dissolution from processing wastes is the primary source for the impact on water quality of surface waters. Yet none of the geochemical signatures indicative of gypsum dissolution were detected in the investigated groundwater; Ca/SO$_4$ of groundwaters was >4 in nearly all samples and the Rb/Sr ratios were similar to the streams if not much greater in the groundwaters.

Overall, our analysis of the groundwater shows that the groundwater quality in the TSB region is primarily influenced by natural water-rock interactions in which the local aquifer rocks are composed of Li pegmatites. Within the area of the legacy Li mine in Kings Mountain (the KMM) the intensity of these water-rock interactions is apparently much higher, causing much higher Li content associated with low K/Rb and high Rb/Sr and ^{87}Sr/^{86}Sr ratios in the local groundwater. Given the strong natural influence on water quality from the pegmatites, we can see that Li is naturally elevated and this would also explain the elevated Rb and Cs previously reported from waters in the region.[27] Additionally, we present evidence

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that the natural co-occurrence of the Li pegmatite with an As-rich geologic unit can amplify the mobilization of As into the groundwater, explaining the exceptionally high As concentrations (up to ∼350 μg/L) detected in the hotspot cluster.

The results of this study indicate that aquifer geology plays a major role in groundwater quality and that mining activities may have increased the intensity of water-rock interactions due to increased mineral surface areas from blasting/fracturing and rock crushing as described in Williams et al.[27] At the KMM site the Li pegmatites were intruded into an amphibolite gneiss unit rather than the nearby mica schist, which results in elevated Li but not As. In contrast, in scenarios that involve Li pegmatite intrusion into the mica schist can lead to elevated As concentrations in groundwater as shown by the arsenic hotspot in the northern section of the TSB. Consequently, such increased water-rock interactions could lead to As contamination in groundwater if a new Li mine is located where the pegmatite dikes intrude into the mica schist unit (or other As-rich rock units). Future mining prospects should investigate the proximity to As-bearing rocks to prevent potential water quality issues induced from water-rock interactions between pegmatites and As-containing rocks.

As with surface waters, groundwater elemental concentrations can also be influenced by seasonal and weather-related variations, particularly for redox-sensitive elements in shallow groundwater.[104] However, nationwide surveys of drinking water wells in the United States have shown that such variations in As[105] and Li[39] concentrations are typically minor. To evaluate potential temporal variability in our study area, we conducted follow-up sampling of several wells within the arsenic hotspot. All resampled wells yielded similar concentrations to those measured previously. For example, the well with the highest As and Li concentrations in 2022 (346 and 469 μg/L, respectively) still exhibited exceptionally high but slightly lower values three years later (301 and 418 μg/L). This indicates minor variability over time but no significant change in magnitude. Although these data do not capture short-term seasonal or weather-related effects, they demonstrate the long-term stability of groundwater As and Li concentrations in the region. We acknowledge that this study was not specifically designed to evaluate seasonal variation; however, the consistency of concentrations across sampling campaigns and years suggests that temporal variability does not significantly affect our observations.

3.3. Broader Implications for Water Quality and Li Mining. Pegmatite deposits are currently the primary source of global Li production,[7,9] are globally distributed (Figure 1),[8] and are projected to continue to be important sources of future Li production.[106] This means that as new mines open around the world, their water-quality impacts may become relatively widespread in comparison to that of brine deposits which are geographically restricted to a few regions (i.e., the central Andes and Tibetan Plateau). The results presented here are from a legacy Li mining region reflecting the lasting impacts decades after mining operations ceased. These lasting impacts and the similarities between Li pegmatite deposits can however provide important information and guidelines on the potential water quality impacts at other new, operational, and legacy Li pegmatite deposits.

The mineralogy of Li rich pegmatites can vary, and yet they form from a highly fractionated endmember of granitic magmas, meaning that they are often enriched (e.g., Rb) and depleted (e.g., Sr) in the same elements,[82] and are also similarly characterized by radiogenic $^{87}Sr/^{86}Sr$ signatures.[83,84] This suggests that the tracers we have developed in this study for understanding the relative degree of water-rock interactions with Li-pegmatites (e.g., high Rb/Sr, $^{87}Sr/^{86}Sr$, and Li concentrations, low K/Rb) should be applicable to other worldwide pegmatite deposits. Furthermore, these pegmatites are typically intruded into similar lithologies as at the TSB (i.e., into mica schists or amphibolites in the upper greenschist to amphibolite facies).[107] This means that the potential water-rock interactions with the pegmatite and surrounding lithologies at other deposits may lead to similar water quality issues as those observed at the TSB. Furthermore, Williams et al.[27] compiled a data set of Li and As concentrations in ground and surface waters around pegmatite deposits throughout the world, which demonstrated that Li is consistently elevated in water resources associated with these deposits. However, As concentrations were elevated only at a few deposits such as at the Greenbushes mine in Australia. This might suggest that the codependent water-rock interactions between the pegmatite and As-bearing rocks discussed at the TSB are also happening at these deposits.

The dissolution of gypsum from Li processing wastes is the major water quality impact on surface waters in the TSB and gypsum is found in processing wastes from several different Li pegmatite processing operations.[28,76−78] The use of sulfuric acid to extract Li from spodumene (and other Li minerals) followed by neutralization with lime/limestone is one of the most common methods for extracting Li.[108,109] Other methods for Li extraction, however, such as alkaline digestion are becoming increasingly common and may produce different waste solids,[108,109] potentially leading to different water quality impacts. Given the common occurrence of gypsum in waste residues and use of the sulfuric acid leaching method, we posit that gypsum dissolution will likely be a frequent impact to water quality at other processing facilities as well. It is important to note that the gypsum impact associated with Li extraction from Li-minerals (e.g., spodumene) is from a processing facility and not directly from mining activities. Globally, mineral concentrates are not always processed at mine sites, meaning that this processing impact on water quality may be exported elsewhere or even overseas. This could be the case with Australian Li-pegmatite mining where most spodumene is exported to China for processing and refinement.[3,110]

Our findings clearly show that Li concentrations in water resources can be extremely elevated in a region with Li mining and processing activities, consistent with findings from other regions.[27,29,33,35−37] Lithium is not currently regulated as a contaminant with a defined drinking water standard and its toxicity remains unresolved,[49] with various studies suggesting that it may or may not have toxic properties (e.g.,[50,111,112]). Regardless, increased mobilization of Li from anthropogenic activities to water resources[48] highlights the need for further research on the toxicity of Li and of other co-occurring and unregulated elements like Rb and Cs.

Overall, our findings demonstrate that water quality impacts in the TSB are primarily governed by two distinct processes: (1) the natural geochemical influence of Li-rich pegmatites on groundwater chemistry and (2) the dissolution of gypsum waste-residues from legacy processing which elevates solute loads in surface waters. While mining activities themselves do not appear to induce water-quality changes beyond those already imposed by natural pegmatite water-rock interactions, the persistence of processing-derived gypsum signatures highlights the long-term legacy of Li processing wastes in shaping surface water salinity and trace-element concentrations (i.e., elevated Li and Rb). Importantly, the co-occurrence of Li pegmatites with As-bearing lithologies underscores the potential for localized groundwater contamination under specific geologic settings. As global demand for Li accelerates and pegmatite mining expands, our results emphasize the need for site-specific assessments that integrate geological context and industrial processing waste characterization with geochemical monitoring to anticipate and mitigate future water-quality risks.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c13682.

Additional figures (Figures S1 through S6) showing variations in trace element concentrations and geochemical tracers in ground and surface water investigated in this study (PDF)

Major and trace element chemistry (Tables S1 and S2) and associated sample information on ground and surface waters investigated in this study (XLSX)

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Notes

The authors declare no competing financial interest.

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