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Quality of Wastewater from Lithium-Brine Mining

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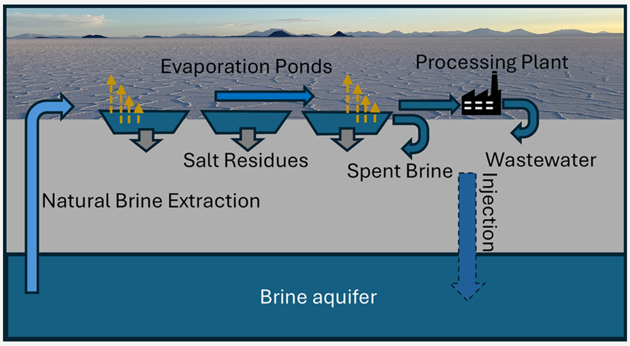

ABSTRACT: The sustainability of lithium mining is one of the critical factors for a successful transition to renewable energy. A potential practice to alleviate brine level decline and loss of adjacent fresh groundwater from brine pumping in the salt pans (salars) is through injection of spent brines into the subsurface. The quality and possible impacts of injecting spent brines have not been fully investigated. Here we present data for major and trace elements in natural brines, brines and salts from evaporation ponds, and wastewaters from a lithium processing plant at the Salar de Uyuni in Bolivia, the largest known global lithium deposit. The investigation reveals that evaporation of natural brines results in highly saline brines (TDS ≈ 360 g/kg) with low pH (3.2) and elevated concentrations of lithium, boron, and arsenic (up to ∼50 mg/kg) that could modify the chemical composition and mineral saturation upon release to the environment. The extremely high arsenic concenrations and low pH also have potential environmental impacts. In contrast, the processing plant generates saline and low-saline wastewater streams with high pH (∼10) and lower solute concentrations that could dilute the natural lithium reservoir, while the high pH limits their disposal options.

KEYWORDS: *Lithium, water quality, spent brines, contamination, wastewater*

1. INTRODUCTION

The transition to renewable energy requires increased extraction of critical raw materials, including Li, which is an essential component of Li-ion batteries. Approximately 40% of global Li production is currently extracted from continental closed-basin brines, of which the Lithium Triangle (LT) in South America (Figure 1) accounts for 50−85% of known Li-brine resources, with China and the western United States accounting for the rest.[1−6] While Li extraction at the Salar de Uyuni (SDU; Figure 1) in Bolivia has not progressed beyond pilot operations, it is the single largest Li deposit globally.[7] The traditional approach for extracting Li from brine resources is via pumping of subsurface brines into a series of evaporation ponds, allowing Li to evapoconcentrate, while precipitating undesired salts (e.g., halite, sylvite, carnallite) before extracting Li-salts in a processing plant.[8,9]

In addition to water availability, a critical component of the sustainability of Li-brine mining is the quality of spent brines (i.e., residual brine after lithium extraction) and the wastewaters that are generated from processing plants. Previous research on environmental impacts of Li-brine mining has almost exclusively focused on issues related to water and land use, documenting shrinking wetlands, changes in salt pan (salar) areas, lowering of groundwater tables, and the concurrent environmental and ecological impacts, including land subsidence.[5,9−15] The carbon and water footprints associated with the Li mining and battery industries have also been addressed.[5,16−18] Spent brines and wastewaters can enter into the environment through at least three pathways: (1) injection into the subsurface, (2) discharge at the salar surface, and (3) leaking of storage and evaporation ponds.[8,9,19] Injection of spent brines to the underlying brine aquifer is being considered to alleviate drawdown of subsurface brine resources and land subsidence of the salar basins.[19,20] There is, however, no information available about the quality of evaporated and spent brines generated during Li enrichment or about wastewaters from processing plants. This study aims to fill this gap and provide detailed information about the chemistry of spent brines and wastewaters from Li-brine mining.

Here we present, for the first time, a broad spectrum of major and trace elements in evaporated and spent brines, solid wastes, and wastewaters from the SDU. We hypothesize that the quality of evaporated brines and wastewaters have the potential to pose environmental hazards if they are leaked or recycled back to the natural environment. Given the importance of Li-brine deposits to global Li production, the

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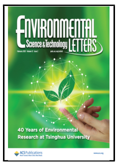

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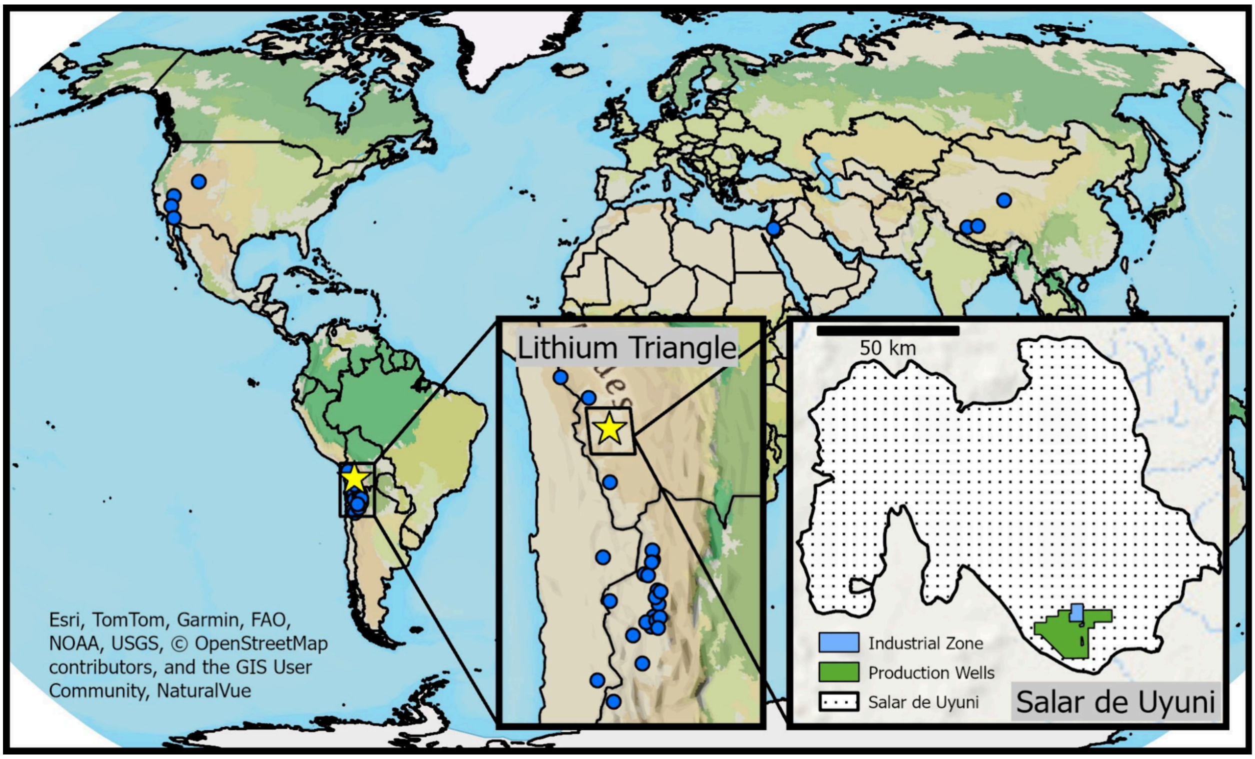

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Figure 1. Map of the global Li-brine deposits (blue dots)[3,21] and the Salar de Uyuni (SDU). The majority of such deposits are found in the Lithium Triangle of South America, where the SDU (yellow star) is considered the largest deposit.[7] At the SDU, the production wells region (green region) is where brines are being pumped, while the industrial zone (blue region) includes evaporation ponds, spent brine pools, and processing plant operations. For reference, the surface area of the SDU is ∼10,000 km^2,[22] while the production well zone is ∼250 km^2, and the industrial zone is ∼28 km^2.

data presented in this study are critical for understanding the potential environmental impacts of Li mining.

2. METHODS AND MATERIALS

2.1. Background Information and Sample Collection. The lithium operation at the SDU pumps brines from ∼16 to 50 m below the subsurface, from which aggregate samples were collected where each sample represents a mix of several wells (natural brine, $n = 8$).[23] This brine is derived from a subsurface brine aquifer, primarily composed of halite and sand units underlain by impermeable lacustrine sediments (Figure S1).[22,24,25] See supplemental text and Figure S1 for hydrogeology and more information.

Natural brines are pumped into an eight-pond evaporation sequence, where Li_2SO_4 and other salts precipitate in the seventh pond and these salts are harvested and redissolved for the processing facility (Figure S2). The residual brines in ponds 7 and 8 are considered spent brines despite their high Li concentrations due to a high Mg/Li ratio inhibiting separation. Brines and salts were sampled from each of the eight ponds (Figure S2).

2.2. Analytical Methods. Major cations (Li^+, Na^+, K^+, Mg^{2+}, Ca^{2+}) and anions (Cl^-, SO_4^{2-}) were measured with ion chromatography (IC), while B and trace metals were measured on an inductively coupled plasma−mass spectrometer (ICP-MS).[26,27] For detailed information, see supplemental text.

2.3. Mixing and Saturation Indices. Saturation indices and mixing were modeled using PHREEQC (v 3.7.3) with a Pitzer ion interaction model and the pitzer.dat database for its applicability to high-salinity solutions.[28,29] Two-component mixing simulations were conducted between wastewaters and spent brines with the average natural brine. These mixing simulations evaluate changes in the chemistry and mineral precipitation/dissolution. Changes in porosity are estimated from these mixtures reacting in a bulk medium representative of the brine aquifer. See supplemental text for more details.

3. RESULTS AND DISCUSSION

3.1. Major and Trace Elements in Brines and Wastewaters. Analyses of major and trace elements of the natural brines, brines from evaporation ponds, and wastewaters from the processing plant (Table S1) reveal Cl, Na, SO_4, K, Li, and B as the major dissolved solutes. Among trace elements, only As, Rb, and Cs were found in elevated concentrations relative to natural brines (Figure 2). In the evaporation ponds, Li, B, and As generally behave conservatively, becoming progressively enriched during evaporation, while total dissolved salts (TDS) rises to a maximum concentration of 358 g/kg (Figure 2). Rb and Cs precipitate into K-salts in ponds 4−7, while Li precipitates as Li_2SO_4 in pond 7 (Figures S2−S4, Table S2). Li and B increase to maximum concentrations of 4,620 and 5,800 mg/kg, respectively (Figure 2). At the Salar de Atacama (SDA), similarly high concentrations of B (6,600 mg/kg) but much higher concentrations of Li (∼25,000 mg/kg) have been reported in evaporated brines.[8,30] Arsenic increases from ∼4.3 mg/kg in natural brines to a maximum concentration of 49.6 mg/kg in pond 8, while Rb and Cs reach their maximum concentrations in pond 4 at 35.8 and 1.4 mg/kg, respectively. This level of As is extremely high, ∼1400-fold higher than the U.S. EPA Criterion Continuous Concentration (CCC) value for potential ecological effects upon chronic discharge of saltwater effluents, with an As

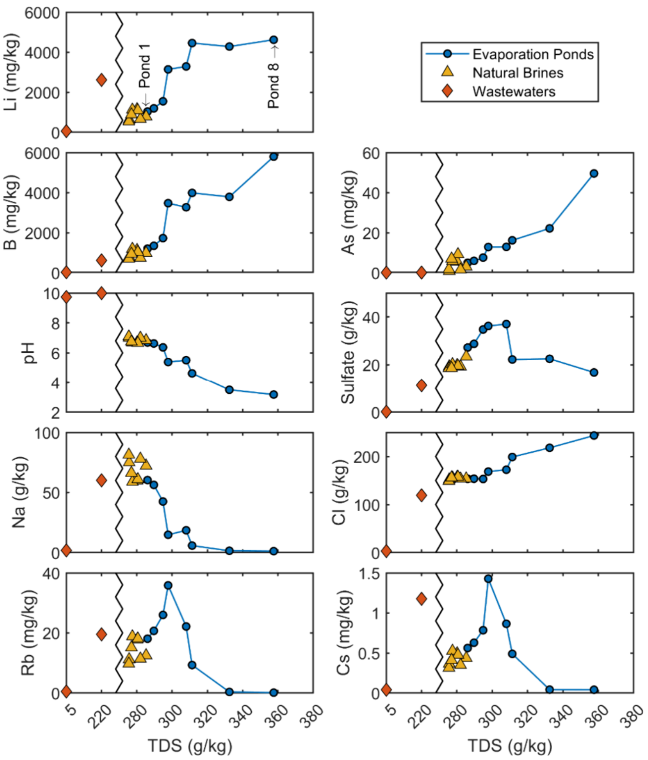

Figure 2. Plots of Li, B, As, pH, SO_4, Na, Cl, Rb, and Cs relative to total dissolved salts (TDS) in natural brines, wastewaters from the lithium carbonate plant, and brines from evaporation ponds at the SDU. In the evaporation ponds, TDS increases from pond 1 to pond 8, with the first and last ponds as labeled in the Li concentration graph; all other ponds are sequentially in order. Figure S2 has a flowchart describing the location of each sample. Note the break in the *x*-axis where TDS values of the wastewaters are on a different scale.

threshold value of 36 μg/L.[31] These results are consistent with previous studies indicating As enrichment in brines and evaporite minerals and reflects that As is highly concentrated in waters and brines throughout the LT.[32−38] The pH of the evaporated brines decreases with evaporation from circumneutral (6.7−7.1) in the natural brines to 3.2 in the most evaporated brine. Similar decreases in pH have been reported at other Li-brine operations, down to 2.5 at the SDA[30] and 4.8 in the Qaidam Basin.[39] We propose that evaporation and increasing boron concentrations cause intensified borate buffering, where heightened salinity increases the dissociation constant of boric acid and dissociation to borate increases the H^+ activity, as has been suggested in hypersaline environments.[30,40] The processing plant generates two wastewater streams: saline wastewater with a TDS of 214 g/kg and relatively elevated levels of Cl, SO_4, Na, Li, and B, and low-saline wastewater with a TDS of 6 g/kg and low concentrations of all major elements. Arsenic concentrations in both the saline and low-saline wastewater streams were relatively low, at <0.06 mg/kg, while the pH was high (>9.7; Figure 2).

Salts in the evaporation ponds precipitate in the order of halite, sylvite, mixed potassium salts, lithium sulfate, and bischofite (Figure S2).[41] These salts are enriched in K, Cl, SO_4, Na, Mg, Li, B, and Rb (Figure S4, Table S2). Solid waste from the processing plant contains relatively elevated levels of K, Li, B, Al, Mn, Fe, Zn, and As (24 mg/kg; Table S2).

3.2. Implications for Injection or Leaking of Spent Brines.

Pumping of subsurface Li-rich brines has been suggested as the cause of declining brine levels and surface subsidence at salars, combined with freshwater resource depletion in adjacent aquifers.[5,9,15,42,43] Furthermore, inflow of low-saline groundwater into aquifers containing salt deposits, due to overpumping of brines, could cause salt dissolution and sink holes, as demonstrated along the periphery of the Dead Sea.[44,45] As such, injection of waste brine back to the subsurface is considered a method for alleviating these issues by maintaining hydraulic pressure.[9,19] The natural deep brines at the SDU are typically saturated with halite; however, reinjection of waste brines could modify the natural equilibrium and introduce precipitation or dissolution of evaporite minerals. This could be particularly problematic for brine recovery if supersaturation and salt precipitation reduce the aquifer permeability. On the other hand, reinjection of dilute wastewater could enhance dissolution of subsurface evaporites, triggering further land subsidence.[46]

To demonstrate these potential impacts, we model the mixing of spent brines with the average natural deep brine and the potential for porosity changes upon precipitation or dissolution induced by blending these brines. The mixing of natural brines with spent brines would increase the concentrations of Cl, SO_4, and B in the mixed solution, resulting in supersaturation of halite and various sulfate (e.g., gypsum) and borate (e.g., hydroboracite) minerals. Following the same approach, injection of the wastewaters from the processing plant would result in undersaturation of halite and gypsum but supersaturation of calcite. The modeling results indicate that this potential precipitation/dissolution outcome can potentially affect the porosity of the underlying aquifer. Both the natural brine and the spent brines are in equilibrium with halite and gypsum; however, when the brines are mixed together this equilibrium is altered, resulting in precipitation of halite and gypsum (up to 45 g per kg of brine) and a decrease in porosity from ∼13.5%[47] to ∼13.1% (Figure 3). The converse is true for wastewaters, which are undersaturated with halite and gypsum, so any mixing with the natural brine would lead to dissolution of halite and gypsum and relatively minor precipitation of calcite, resulting in dissolution of up to 262 g in the low-saline wastewater and potentially increasing the porosity up to 15.7% (Figure 3). In both cases, halite, the primary mineral at the SDU, would be the most affected.[22,48]

Injection of different spent brines would change the chemistry of the natural brine. For example, in a 1:1 mixing scenario, the spent brines could decrease the pH (from 6.8 to 5.0; Figure 3) and increase Li (from 860 to 2,780 mg/kg), As (from 4.4 to 35.9 mg/kg), and B (from 940 to 3,420 mg/kg) concentrations. Injection of acidic spent brine could enhance leaching from subsurface solids such as the lacustrine sediments that are characterized by detrital materials, clays, and carbonates.[25,48,49] Likewise, injection of the basic wastewaters would raise the pH (to ∼7.6−8.3; Figure 3). Injection of the saline wastewater would increase Li (to 1,690 mg/kg), while mixing with the dilute wastewater would decrease the Li concentration (to 521 mg/kg). Mixing with both wastewater sources would decrease the B and As concentrations. Overall, injection of spent brines and wastewaters could induce changes to solute concentrations including long-term lithium dilution or enrichment of the underlying brines.

The heightened density of the spent brines (>0.1 g/mL higher than that of natural brines) is expected to lead to

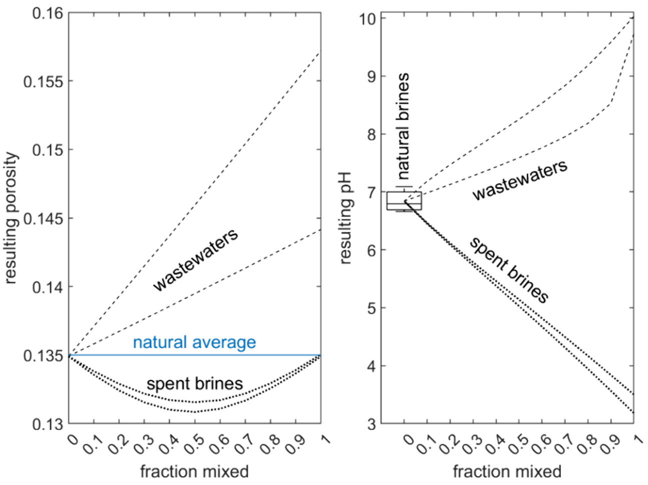

Figure 3. Results of mixing scenarios between natural brine and spent brines and wastewaters. Left: the resulting porosity from the dissolution or precipitation of halite and gypsum in the halite aquifer after mixing with wastewater or spent brine. Note that spent brines and natural brines are in equilibrium with halite and gypsum, while their mixing would result in precipitation or dissolution. Right: mixing curves between spent brines from evaporation ponds and wastewaters from the Li carbonate plant with the natural brines, showing the modeled pH changes. The spent brines would decrease the pH, while the wastewaters would raise it in the impacted brines.

density-driven mixing, where the denser spent brines sink through the natural brines, forming an isolated hypersaline plume rather than completely mixing with the natural aquifer brine, resulting in a mixing zone at the plume edges. Such mixing zones have been observed at the SDA between dense brines and fresh waters,[50] and naturally in other salt pans, where dense evaporated surface brines drive circulation with deeper, less dense brines.[51−54] Therefore, the mixing scenarios and porosity changes described above would likely impact the hypersaline plume edges.

For reinjection, blending wastewaters with spent brines may offer a path to preventing some of these issues. For example, mixing the saline wastewater with the spent brine from pond 8 in a 3:2 ratio would result in a brine at halite saturation, a pH ∼6.6, and a density of 1.24 g/mL, only ∼0.016 g/mL (1.3%) greater than the natural brines, potentially alleviating some of the issues described above. The multiple waste streams with varied salinities and pH values allow for several blending options to mitigate the negative effects of the spent brines and wastewaters. Nonetheless, in the event of leaking storage ponds, the issues described here would still apply. This is especially true since the pond liners at the SDU have degraded.[55,56]

3.3. Environmental Implications. The potential leaking of brines from the evaporation ponds and artificial discharge of the spent brines and wastewaters to the surface brines in the basin could result in adverse environmental impacts. In particular, the elevated As concentration and low pH in the brines from the evaporation ponds (Figure 2) could have ecological effects upon their release to the environment. For example, brine shrimp are adversely affected by As concentrations above 8 mg/L, and in turn, flamingos that inhabit the salars of the Andes and consume brine shrimp bioaccumulate As, resulting in negative health outcomes.[57−60] Therefore, release of spent brine with ∼6-fold higher As than this threshold could have a negative ecological impact. Since As is found abundantly in sulfide deposits, groundwater, surface water, and geothermal waters in the LT, other Li-brines and evaporation ponds are also likely to be enriched in As, similar to the results presented here.[34,35,61−64] Similarly, high As concentrations have been reported in natural brines of some Chilean salars.[36] As far as we are aware, the As issue has never been presented in the context of waste solutions from Li-brine extraction. The data from the SDU presented in this study indicate a potential environmental hazard of high As and low pH in the spent brine from the evaporation ponds, in contrast to the high pH and low As of the wastewaters from the Li carbonate plant (Figure 2). Furthermore, the high pH values of both wastewaters exceed the CCC threshold (pH 8.5) for potential ecological impacts of chronic discharge of effluents.[31] While As and B are known toxins, Li toxicity remains understudied, yet some studies suggest that it could also have toxic effects.[65,66] Therefore, the introduction of high Li, B, and As concentrations to the aquatic environment from wastewaters or spent brines could have the potential to adversely impact aquatic life in the salar environment.

While evaporation ponds are commonly located on the salar surface, overlying brine aquifers and within a closed basin, processing plants are located either adjacent to the evaporation ponds, within the basin (as is the case at the SDU), or off-site (as is the case with operations at the SDA).[8] Consequently, wastewaters from processing plants located away from the basins might have a greater potential to contaminate freshwater resources in the vicinity of these plants. In contrast, the salar environments are dominated by brines, with freshwater resources typically only found at the mixing zones of the salar margins.[50,67,68]

Overall, the data on brines from evaporation ponds and wastewaters from the processing plant at the SDU suggest that injection of spent brines and wastewater could induce unfavorable impacts on the chemistry of natural brines and consequently the underlying brine aquifer porosity. This study highlights the occurrence of elevated concentrations of As, a highly toxic element, and its conservative accumulation in evaporation pond brines to levels that are orders of magnitude higher than ecological standards. Consistently, we show that the pH of the evaporated brines is progressively decreasing to acidic conditions. Future studies should further evaluate the mechanisms that control the pH decline and the enrichment of As in evaporated brines as well as the environmental implications of Li-rich and spent brines from other basins. Since injection of spent brines could be an important tool to compensate for brine extraction, future studies should also investigate possible management strategies (e.g., blending different wastewater streams) to alleviate the potential negative impacts.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.4c01124.

Supplemental Figures S1−S4, experimental details, and Tables S1 and S2 (PDF)

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Notes

The authors declare no competing financial interest.

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