



Geological controls on lithium production from basinal brines across North America

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ABSTRACT

With increasing lithium (Li) demands for electric car batteries and grid storage, additional Li beyond traditional continental brines and hard-rock sources might be necessary. Elevated Li concentrations have been reported in select oil field brines. Li could be extracted from the large volumes of saline fluids produced by oil and gas wells or from new wells dedicated to Li production. However, the spatial distribution of Li⁺ concentrations, the rates at which fluids can be produced in areas with elevated Li⁺ concentrations, and potential sources of Li enrichment are not well characterized. To help identify additional sources of Li, this study investigates the concentration, origin, and potential production rates of Li from sedimentary basin brines across North America. New Li data from brines in various stratigraphic units in the Paradox Basin are combined with existing datasets from the basin and others across North America. New Li analyses of organic-rich shales in the Paradox Basin are also examined to provide insights into the origins of Li. Results from this study show the median Li⁺ concentration in oil and gas produced waters across North America is ~5 mg/L; these generally low concentrations are unlikely to support Li production. However, higher Li⁺ concentrations (>65 mg/L) are found in select basins and strata containing deep saline fluids, associated with evaporation of paleo-seawater and precipitation of halite and potash salts, that have not been flushed by meteoric recharge. High Li content of organic-rich shales (range: 20–440 ppm; median: 60 ppm) interbedded with evaporites in the Paradox Basin suggest fluid-rock interactions with diagenetically-altered shales and organic matter are one potential source of Li-enrichment in basinal brines. Observed fluid production and injection rates indicate that Li production will likely be highest in sandstone and carbonate formations with relatively high permeability. Potential Li production rates can vary by orders of magnitude even within these strata due to heterogeneity in both Li concentrations and permeability, as well as recovery efficiencies between 40 and 70 % with current technology. Targeted Li production wells, rather than relying on existing oil and gas wells, would likely be necessary to produce brines at sufficient volumes to support Li production at these low concentrations. Co-recovery of other critical elements or combination with other subsurface developments, such as geothermal energy production, may make Li production from sedimentary basin fluids more viable.

1. Introduction

Lithium (Li) is critical to achieving a sustainable energy transition (Greim et al., 2020). Demand for Li for electric car batteries and grid

storage has increased dramatically with the need to reduce the use of fossil fuels to decrease carbon dioxide emissions (Barbot et al., 2013; Bradley et al., 2013; Can Sener et al., 2021; Daitch, 2018; Martin et al., 2017; Seip et al., 2021; Tabelin et al., 2021; Verma et al., 2016). Most of

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the Li production today comes from pegmatites (e.g., Greenbushes pegmatite in Western Australia) and continental brines (e.g., associated with salars in the “Li triangle” in Argentina, Chile, and Bolivia) (Benson et al., 2017). Li-rich clays in Nevada are currently being explored as another potential major Li source (Cough, 2021). Brines co-produced with oil and gas in sedimentary basins (“oil field brines”) have also been proposed as a potential Li source (Can Sener et al., 2021; Seip et al., 2021; Tahil, 2008; Verma et al., 2016); however, there have been few systematic studies to date on Li^+ concentrations and potential production rates from oil-field and other, non-hydrocarbon associated sedimentary basin brines (Dugamin et al., 2021).

Large volumes of brines ($2.8 \times 10^9 \text{ m}^3$ in 2017) are produced from sedimentary basins during oil and gas production (Clark and Veil, 2009; Veil, 2015, 2020). Although recovery from Li brines are well under 70 % with existing technologies (Tran and Luong, 2015), there is considerable interest in extracting Li from these produced waters. Previous studies of Li in sedimentary basin brines have focused on the Smackover Formation in the Gulf Coast region (including Gulf Coast, East Texas and Arkla basins) where Li^+ concentrations reach 1700 mg/L, although average values are much lower (174–187 mg/L; Bradley et al., 2013; Collins, 1979; Daitch, 2018; Tahil, 2008).

Maximum potential Li production rates at the wellhead from Smackover Formation brines have been estimated to range from 2.0×10^4 to 5.5×10^4 tons per year (tpy) lithium carbonate equivalent (LCE), which is approximately equal to 4.0×10^3 to 1.1×10^4 tpy of Li (Tahil, 2008; Verma et al., 2016). Potential Li production rates from the Permian Basin in Texas and New Mexico and oil fields in California has been estimated to reach as high as 4.7×10^4 tpy (Can Sener et al., 2021). Whether these Li production rates are possible in a broad range of sedimentary basins is unclear because of the wide range of Li^+ concentrations (Blondes et al., 2018) and ability to produce sufficient volumes of brines (Ferguson and Ufodu, 2017).

Here, we provide an overview of the potential to produce Li from sedimentary basin brines across the United States and the Williston Basin in Canada. We identify sedimentary basins and specific geologic units within the basins that contain high Li^+ concentrations from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018). New geochemical analyses of Paradox Basin formation waters, and recently published analyses of Williston Basin brines (Mowat, 2023) are used to supplement these existing data. We compare major ion chemistry and Br^- to seawater evaporation trajectories (McCaffrey et al., 1987) to identify processes that lead to elevated Li^+ concentrations. New geochemical analyses of organic-rich shales within the Paradox Basin help constrain Li sources. Potential Li production rates are estimated from using solute concentrations and observed rates of fluid production and injection.

2. Background

2.1. Lithium geochemistry of basinal brines

Paleozoic sedimentary basins often contain highly saline fluids (brines; $>35,000 \text{ mg/L}$ salinity; Hanor, 1989) that came from evaporation of paleo-seawater and/or the dissolution of marine evaporite deposits. Brines derived from evaporation of seawater tend to be enriched in Na^+ , Cl^- , and Br^- with relatively low $\text{Cl}^-:\text{Br}^-$ ratios. These brines are often enriched in Ca^{2+} and depleted in Mg^{2+} and K^+ from fluid-rock reactions, such as albitization, dolomitization and illitization, respectively (Kharaka and Hanor, 2003). In contrast, brines derived from dissolution of evaporites tend to have low Br^- concentrations and high $\text{Cl}^-:\text{Br}^-$ and $\text{Na}^+:\text{Br}^-$ ratios (Hanor, 1994, 2001).

Lithium in basinal brines is thought to have initially come from seawater when marine-derived brines were formed at the surface under highly evaporative conditions, such as in restricted basins (Daitch, 2018; Holser, 1979; Huff, 2019; Phan et al., 2016; Vengosh et al., 1992; Yu et al., 2013). Li^+ could have been added to seawater via rivers or diffuse

groundwater discharge or dust deposition from continental weathering (Bradley et al., 2013; Daitch, 2018; Yu et al., 2013). Some Li^+ was likely removed via adsorption onto organic matter and clays near the surface (Williams et al., 2015). As seawater evaporated, carbonate and evaporite minerals were deposited and incompatible elements, such as Li^+ and Br^- were concentrated in solution (Bradley et al., 2013; Holser, 1979; Huff, 2019).

As evaporated seawater-derived brines infiltrated into the subsurface and/or were buried with basin sediments, Li^+ may have been added by fluid-rock reactions with Li-bearing rocks and minerals, such as volcanic ash (e.g., rhyolitic tuff), detrital sediments, and clays, dependent on the fluid chemistry and temperature (Kesler et al., 2012; Moldovanyi and Walter, 1992). Increased salinity typically drives greater diagenetic reactions (Hanor, 2001). At depth in sedimentary basins (e.g., Denver Basin), as clay minerals are altered to illite and organic matter is converted to oil and gas, Li^+ can be released from adsorption sites into solution (Williams et al., 2015). Thus, Li^+ generally increases with depth and total dissolved solids (TDS) in sedimentary basin fluids.

Fluid-rock reactions which added or removed Li^+ to solution may have taken place in situ or by circulation of basinal brines through adjacent formations or underlying crystalline basement rocks (Garrett, 2004; Huff, 2019; Kesler et al., 2012; Yu et al., 2013). For example, brines in the Smackover Formation in the Gulf Coast Basin may have obtained Li^+ from in-situ dissolution of detrital feldspar minerals, exchange with clays, and/or reaction with volcanic materials (Collins, 1978; Moldovanyi and Walter, 1992). Brines in the Western Canada Sedimentary Basin, including the Alberta and Williston basins, may have elevated Li^+ from hydrothermal inputs from underlying basement rocks (Eccles and Berhane, 2011; Garrett, 2004; Huff, 2019). In the Marcellus Shale in the Appalachian Basin, brines have variable Li^+ concentrations depending on their spatial location that reflects differences in the Li content of the organic-rich shale across the basin (Phan et al., 2016). Genetic models of Li sources are generally lacking, in part because of a paucity of formation water and associated rock Li data. The new data on the Li content of organic-rich lithologies from the Paradox Basin, provided in this paper, present an opportunity for further evaluation of the relationship between Li brine-concentration and Li-rock concentration.

2.2. Fluid production from sedimentary basins

Fluid production rates are an important consideration in the recovery of Li^+ from sedimentary basin brines. Overall produced fluid volumes are large (Clark and Veil, 2009) but it is unclear how and whether these volumes correspond to Li^+ concentrations. Rates of water production by the oil industry vary substantially between (McIntosh and Ferguson, 2019) and within basins (Ferguson and Ufodu, 2017; Jellicoe et al., 2022).

Variations in fluid production rates can partly be attributed to geological factors. Permeability values in sedimentary rocks varies over several orders of magnitude in sedimentary basins, with some shales having values $<10^{-20} \text{ m}^2$ (Neuzil, 2019), while sandstones and carbonates commonly have permeabilities of 10^{-16} to 10^{-14} m^2 (Gleeson et al., 2011). Reservoir thickness (B) also exerts an important control on fluid production. The pressure response of a reservoir to pumping is a function of transmissivity (T), which is defined as:

$$T = k \rho g B / \mu \quad (1)$$

where k is permeability, ρ is fluid density, g is acceleration due to gravity and μ is viscosity.

Historical production rates from oil wells provide a basis for estimating how much water could be produced to extract Li. However, the petroleum industry does not operate wells in a manner that maximizes water production, suggesting that wells designed and operated to produce Li could produce more water from the same strata. Comparison of injection rates in disposal wells to production wells in the same strata in

the Western Canada Sedimentary Basin indicates that wells operated to optimize water production should be able to produce more water than existing oil wells (Ferguson and Ufodu, 2017). We expect that wells designed for brine extraction will produce greater volumes of fluid than existing oil and gas wells.

2.3. Lithium extraction from basinal brines

The minimum Li^+ concentration in brines for economic production ranges from >65 mg/L (Tahil, 2008) to >100 mg/L (Kumar et al., 2019; Ventura et al., 2020), assuming that brines can be produced in sufficient quantities. We use the minimum value (>65 mg/L) to identify basins and strata with elevated Li contents that may be viable for production. The actual economical Li^+ concentration threshold will depend on the cost of drilling and brine production, extraction technology, and market price, which are beyond the scope of this study.

Traditional methods of Li extraction from brines begin with spreading the pumped brine out into an evaporation pond, preferably large, wide, and shallow. Solar evaporation pre-concentrates the brine until halite and sylvite precipitate, by which time the Li^+ concentration has typically increased from 100 s of ppm to 4–6 % (Tran and Luong, 2015). At this point, processes diverge. For brines with high Mg and/or B contents, the next step is to remove Mg or B, which interfere with Li^+ extraction. Magnesium stabilizes carnallite, which absorbs Li^+ and causes loss; B interferes with electrolytic Li separation in later stages. High Ca contents may require Ca precipitation during this step as well. These are removed by adding a precipitant, such as lime, dolime, or NaOH; oxalates; or Na_2SO_4 . Once most major contaminants have been precipitated, the remaining brine is decanted away from these solids. If they can be purified cheaply, the precipitates (halite, sylvite, bischofite, borates, gypsum, Mg-hydroxide, potash) can be sold as by-products (Tran and Luong, 2015).

From this brine, Li^+ may be directly precipitated as Li_2CO_3 via carbonation or further purified by electrodialysis, producing LiOH that can precipitate from solution with further evaporation or by chilling (Tran and Luong, 2015). These suffice for low-purity applications, such as glassmaking, but can also be put through cycles of redissolution and recrystallization to produce LiCl, LiOH, or Li_2CO_3 at >99.5 % purity. For ultra-pure Li_2CO_3 , such as used in batteries, the purification to >99.9 % usually includes another redissolution step, followed by ion exchange, before final precipitation.

This traditional process takes months to years because of its reliance on evaporation. Combined with increasing Li demand, this has led to considerable recent research into faster alternatives relying on direct separation of Li^+ from other ions in the brine. One proposed technique is electrolysis, with Li^+ separated across an anion-selective membrane between two electrodes (Tran and Luong, 2015; Iyer and Kelly, 2022). Depending on the electrode, this can offer efficient separation, but electrode performance degrades rapidly with use, diminishing loading capacity over time. The expense of the electrodes and current also leads to separation rates too low for the industrial scale with existing systems.

Selective adsorption of Li^+ has also been studied using synthetic oxides of Mn, Ti, Al, and other metals that can load 1 to 30 mg Li/g adsorbent from solutions with as little as 5 ppm Li. The Li is then eluted from the adsorbent with acid for refining and precipitation. Reported recoveries can approach 100 %, particularly for the more complex synthetic adsorbents, but Li^+ tends not to be the only solute recovered. There is an inverse relationship between recovery and selectivity, and even ion sieves accumulate significant Na^+ , K^+ , Ca^{2+} , and Mg^{2+} , as well as Li^+ . These desorb with the Li^+ during elution, and then must be precipitated anyway. These initial attempts at direct Li^+ extraction (DLE) have tended to meet with little success (Tran and Luong, 2015). More recently, several companies are testing ion-exchange and selective adsorption processes that show promise of recoveries of 90 % or higher, from brines of moderate Li^+ concentration (Anson Resources, 2023, <https://www.ansonresources.com/>; Volt 2023, <https://voltlithium.com/>;

E3 Lithium, <https://www.e3lithium.ca/our-assets/technology/>; International Battery Metals, <https://www.ibatterymetals.com/direct-lithium-extraction>; accessed October 24, 2023). As an example, a pilot facility is currently testing DLE from Salton Sea geothermal brine, which averages >200 ppm Li^+ (McKibben et al., 2021). The extractive processes begin after electricity generation and the filtration or precipitation of interfering brine components. A selective adsorbent then extracts Li^+ , which desorbs to a LiCl solution. When precipitated, LiCl can be readily converted into saleable LiOH (Iyer and Kelly, 2022; McKibben et al., 2021), while spent brine is reinjected into the ground for standard geothermal cycling (<https://www.esminerals.com/>). These processes are still in the testing phase, so their overall economic viability and minimum Li concentration thresholds remain unknown.

3. Methods

3.1. Formation water analysis and data compilation

To evaluate the concentration of Li^+ in sedimentary basin brines across North America, formation water data were compiled from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018), coupled with additional published sources (Blondes et al., 2020; Gallegos et al., 2021; Hite, 1964; Mayhew and Heylmun, 1965; Mowat et al., 2021; Mowat, 2023; Peterman et al., 2017; Phan et al., 2016; Rowan et al., 2011; Tasker et al., 2020), as summarized in Supplemental Table S1. To augment the existing Li dataset, we also collected and analyzed select formation water samples from the Paradox Basin (Table S2) where we had access to oil and gas wells and exploratory Li mining wells. The location of formation water samples with Li data, collected as part of this study and compiled from existing literature are shown in Fig. S1.

Thirty formation water samples were collected from the Paradox Basin, in the Colorado Plateau, USA, and analyzed for Li^+ (Table S2; Fig. S1) in addition to previously published major ion and isotope chemistry (Kim et al., 2022a). The samples spanned from near the surface in fresh to brackish water aquifers to saline formation waters at depth in oil and gas reservoirs. One formation water sample was collected from a Li exploration well completed in the Pennsylvanian Cane Creek Shale member of the Paradox Formation, northwest of Moab, Utah. Eight formation water samples were collected from oil and gas wells located in the Lisbon Valley near the Utah and Colorado borders. These samples include the Pennsylvanian Honaker Trail, Mississippian Leadville Limestone, and the Devonian McCracken (sandstone) formations. Six ground water samples in the Lisbon Valley were collected from the Jurassic Navajo Sandstone and Burro Canyon formations. In the Greater Aneth oil field, south of Blanding, Utah, 11 samples from oil and gas wells were collected from Ismay – Desert Creek members of the Paradox Formation. Saline formation waters were also collected from four brine extraction wells associated with the salt diapir in the Paradox Valley, Colorado. For more detailed information on sampling location and collection methods, auxiliary geochemical and isotopic data, and fluid history see Kim et al. (2022a, 2022b). New Paradox Basin data were combined with previously published data (Table S1).

Paradox Basin formation water aliquots for Li^+ were filtered with $0.45 \mu\text{m}$ nylon membrane filters, placed into 30 mL pre-acid-washed HDPE sample bottles, and acidified by adding two drops of concentrated Optima-grade nitric acid. Prior to analysis, the samples were stored on ice in the field, and at $\sim 4^\circ\text{C}$ in a refrigerator in the laboratory. Li^+ concentrations were determined using Inductively Coupled Plasma - Mass Spectrometry in the Department of Geological Sciences at the University of Texas El Paso in the range of 3 % to 24 % analytical precision (1σ), using a standard reference sample (USGS T217). Methods for previous collection and analysis of Cl^- , Br^- , Mg^{2+} , and K^+ are reported in Kim et al. (2022a).

3.2. Rock sample lithium measurements

Cores, chips, and cuttings of shale interbeds from the Paradox Formation were collected from the United States Geological Survey, Core Research Center (Table S3). Core materials came from boreholes drilled in various locations throughout the Paradox Basin. Rock samples were analyzed by ALS Geochemistry Labs in Vancouver, British Columbia, Canada. Each sample was weighed, crushed to 70 % passing 2 mm, then split. Analytical splits were pulverized to 85 % passing 75 μm . Lithium content was analyzed as part of a base metal suite determined by 4-acid digestion followed by ICP-AES with analytical range for Li from 10 ppm to 1 %. New analyses (Table S4) were combined with previously published Li data for Paradox Basin black shales (Tuttle et al., 1996).

3.3. Lithium production rate estimates

To estimate potential Li production rates for formations with the highest Li^+ concentrations ($>65 \text{ mg/L}$), measured and estimated flow rates for produced waters were compiled for the Jurassic Smackover Carbonate in the Gulf Coast, East Texas and Arkla regions (ADEE, 2021), Devonian Marcellus Shale in the Appalachian Basin (Kondash and Vengosh, 2015), Mississippian Charles/Midale Carbonate (Jellicoe, 2021) and Devonian Birdbear and Duperow limestones in the Williston Basin (IHS, 2023), and the Pennsylvanian Paradox Formation in the Paradox Basin (<https://dataexplorer.ogm.utah.gov/>) (Mayhew and Heylmun, 1965).

Initial potential Li production rates (Q_{Li}) were estimated by multiplying the average fluid production (Q_{water}) rate by the Li^+ concentration (C) (Table 1).

$$Q_{\text{Li}} = Q_{\text{water}} C \tag{2}$$

Observed fluid production and injection rates were used to estimate potential Li production rates (Table 1). The median and 95th percentiles of Li^+ concentrations are used in our estimates to account for both typical settings and select locations of high Li^+ concentrations.

4. Results

4.1. Distribution of lithium and geochemical relationships

The median Li^+ concentration of formation waters in sedimentary basins across the United States and the Williston Basin in Canada is $\sim 5 \text{ mg/L}$, with 5th and 95th percentiles of 0.2 mg/L and 97 mg/L , respectively. There are numerous sampling points (Fig. S1) and basins with average Li^+ concentrations exceeding 65 mg/L (Fig. 1), but the concentrations often vary over five orders of magnitude. In general, Li^+ concentrations increase with total dissolved solids (TDS; Fig. 2). Li^+

values $>100 \text{ mg/L}$ tend to be only found in saline fluids with $>100,000 \text{ mg/L TDS}$; however, not all saline fluids are enriched in Li^+ .

Many sedimentary basins, in particular those in the western United States without extensive marine evaporites (Fig. 3), have median Li^+ concentrations $<10 \text{ mg/L}$ and all basins examined have median concentrations $<50 \text{ mg/L}$ (Fig. 3a). The highest median Li^+ concentration ($40\text{--}50 \text{ mg/L}$) is found in the Appalachian Basin, followed by $20\text{--}30 \text{ mg/L}$ in the Arkoma Basin-Ouachita Thrust Belt, and $10\text{--}20 \text{ mg/L}$ in the Michigan, Illinois, and Palo Duro basins (Fig. 3a). The 95th percentile of Li^+ concentrations in sedimentary basins is $>50 \text{ mg/L}$ in the Gulf Coast, East Texas, Arkla, Arkoma, Appalachian, Michigan, Paradox and Palo Duro basins with samples with concentrations $>65 \text{ mg/L}$ in each of these basins (Fig. 3b). Note that the USGS National Produced Waters Geochemical Database (Blondes et al., 2018) uses a slightly different basin classification than the USGS Sedimentary Basin Catalog (Coleman Jr and Cahan, 2012), where the East Texas and Arkla basins are included as subbasins within the Gulf Coast Basin.

We focus more detailed investigation on the Paradox and Williston basins, with new data in this study and recently published data (Mowat, 2023); the Gulf Coast Basin that has the highest Li^+ content reported to date (Smackover Formation brines); and the Appalachian Basin, another petroliferous, salt basin with remnant saline fluids and elevated Li^+ . Within these sedimentary basins, Li^+ concentrations vary substantially by geologic formation and lithology, and generally increase with depth (Fig. 4). In the Gulf Coast Basin, the highest Li^+ concentrations ($>65 \text{ mg/L}$ median values) are found in the Jurassic Smackover Formation and Cretaceous Edwards Group, both carbonates (Fig. 4a). In the Appalachian Basin, the Devonian Oriskany Sandstone and Cambrian-Lower Ordovician Theresa Formation (primarily carbonates) have median values $>65 \text{ mg/L}$ (Fig. 4b). Brines in several other geologic formations have median Li^+ values approaching 65 mg/L , including the Middle Devonian Marcellus Shale, Silurian Clinton and Medina Groups (mostly sandstones), and Ordovician Utica Shale. The Heidelberg, Tuscarora and Gatesbury formations contain one measured sample each with Li^+ concentrations $>65 \text{ mg/L}$. In the Williston Basin, median Li^+ concentrations of the Devonian Three Forks Formation (dolomite, mudstone and bituminous shale) are above 65 mg/L , while several other geologic units contain $>10 \text{ mg/L}$, including the Mississippian Madison, Devonian Bakken, Birdbear, Duperow, Souris River, Dawson Bay, and Prairie, Silurian Interlake, Ordovician Red River and Winnipeg, and Cambrian-Ordovician Deadwood formations (Fig. 4c). The median Li^+ concentration of formation waters in the Paradox Basin are all $<65 \text{ mg/L}$ (Fig. 4d). Li^+ is noticeably low in the Pennsylvanian through Cretaceous siliciclastic formations overlying the Pennsylvanian Paradox Formation evaporites. There are several samples with $>65 \text{ mg/L Li}^+$ in the Paradox Formation, and median Li^+ values in the underlying Mississippian Leadville Limestone and McCracken (sandstone) Formation are 15 and

Table 1
Production rate estimation for Li enriched brines using the average and maximum Li^+ concentration in the Gulf Coast (including East Texas and Arkla), Appalachian, Williston, and Paradox basins.

Basin	Formation	Permeability (m^2)	Thickness (m)	Median Li (mg/L)	95th % Li (mg/L)	Flow rate (L/yr)	Median Li Flux (tpy/well)	95th Percentile Li Flux (tpy/well)
Gulf Coast/East Texas/ Arkla	Smackover	$3 \times 10_i^{-14}$	180 _{vi}	84	396	$4.9 \times 10_x^8$	41	190
	Marcellus	$1 \times 10_{ii}^{-19}$	53 _{vii}	65	214	$1.7 \times 10_{xi}^5$	0.01	0.04
Appalachian	Midale	$2 \times 10_{iii}^{-14}$	24 _{viii}	25	70	$2.3 \times 10_{iii}^7$	0.58	1.6
	Bakken	$4 \times 10_{iv}^{-17}$	60 _{viii}	45	82	$1.2 \times 10_{iii}^9$	0.75	1.4
	Birdbear	$1 \times 10_{iii}^{-14}$	35 _{viii}	33	72	$4.2 \times 10_{iii}^7$	1.4	3.0
Williston	Duperow	$2 \times 10_{iii}^{-15}$	150 _{viii}	10	70	$1.3 \times 10_{iii}^8$	1.3	9.2
	Cane Creek	N/A		110	N/A	$2.4 \times 10_{iii}^9$	0.26	N/A
Paradox		1×10^{-15} to						
	Leadville	$8 \times 10_v^{-15}$	150 _{ix}	13	78	$4.2 \times 10_v^8$	5.5	33

References: i. Melas and Friedman, 1992; ii. Elsaig et al., 2016; iii. IHS, 2023; iv. Kuhn et al., 2012; v. King et al., 2016; vi. Vestal, 1950; vii. Coleman et al., 2011; viii. Williston Basin TGI (<https://www.manitoba.ca/iem/geo/williston/tgi/abouttgi.html>); ix. Nuccio and Condon, 1996; x. Arkansas Oil and Gas Commission (<https://www.aogc.state.ar.us/data/default.aspx>); xi. Kondash et al., 2017; xii. Utah Department of Natural Resources (<https://oilgas.ogm.utah.gov/oilgasweb/live-data-search/lds-prod/prod-lu.xhtml>).

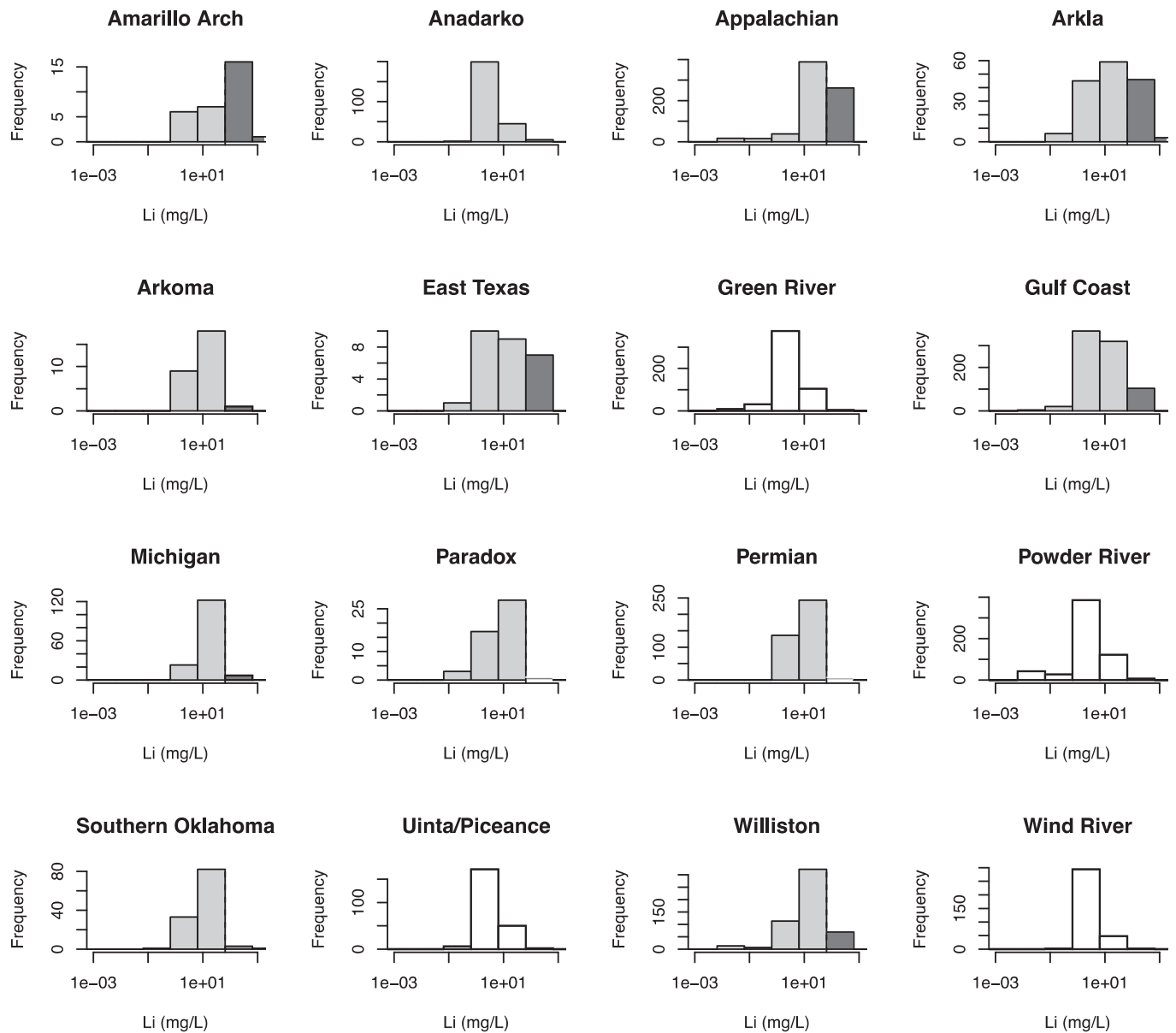


Fig. 1. Li^+ concentration of produced waters from sedimentary basins across the United States and the Williston Basin in Canada, compiled from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018). Basins in grey indicate those with extensive evaporite deposits and brines derived from evaporation of paleoseawater. Basins in white do not contain extensive marine evaporite deposits. Dark grey bars represent samples containing $>65 \text{ mg/L Li}^+$.

24 mg/L, respectively.

Formation waters within the Gulf of Mexico, Appalachian, Paradox and Williston basins, associated with elevated Li ($>65 \text{ mg/L}$) and TDS ($>100,000 \text{ mg/L}$) are enriched in Cl^- and Br^- , relative to seawater (Fig. 5a). Many of the brine samples plot along the seawater evaporation curve, past halite saturation ($> \sim 1.8 \times 10^5 \text{ mg/L Cl}^-$; $> \sim 630 \text{ mg/L Br}^-$), with a few brines plotting past potash salt saturation ($> \sim 2.2 \times 10^5 \text{ mg/L Cl}^-$; $> \sim 4680 \text{ mg/L Br}^-$) (McCaffrey et al., 1987). Some samples in the Williston, Appalachian and Paradox basins plot far above the seawater evaporation trend, enriched in Cl^- relative to Br^- . In several cases, brine samples plot below the seawater evaporation curve, along a mixing line with a dilute meteoric water end-member (plotting near the origin in the ion versus Br^- plots; Fig. 5). The highest Li^+ concentrations are found in basinal brines with relatively low $\text{Cl}^-:\text{Br}^-$ mass ratios (Fig. 6a). Formation waters with high $\text{Cl}^-:\text{Br}^-$ mass ratios have relatively low Li^+ concentrations.

Formation waters in all four basins, except for a few samples, are substantially depleted in Mg^{2+} relative to evaporated seawater (Fig. 5b).

Formation waters in the Gulf Coast and Appalachian basins are also depleted in K^+ (Fig. 5c). Brines in the Williston and Paradox basins are more variable, with some samples plotting near the seawater evaporation trend for K^+ and Br^- , some enriched in K^+ relative to Br^- and some depleted. Formation waters with elevated Br^- concentrations, relative to seawater, are also significantly enriched in Li^+ , in some cases by over 2000 $\times$ seawater composition ($\sim 0.2 \text{ mg/L}$) (Fig. 5d). Samples that are depleted in K^+ relative to evaporated seawater, show a positive linear relationship between K^+ and Li^+ (Fig. 6b), with a R-squared value of 0.27 for $\log \text{K}^+$ versus Li^+ , which is significant ($p = 2.2 \times 10^{-6}$). There is no apparent relationship between Mg^{2+} and Li^+ , although the highest Li^+ samples ($>200 \text{ mg/L}$) all have low Mg^{2+} concentrations ($< \sim 10,000 \text{ mg/L}$).

New measurements of rock Li concentrations of Paradox Formation organic-rich shales in the Paradox Basin vary from 20 to 440 ppm (Table S4). These values are comparable to previously published Li data for the Paradox Formation (Tuttle et al., 1996). The median Li content for the Paradox Formation, including all samples measured to-date is 60

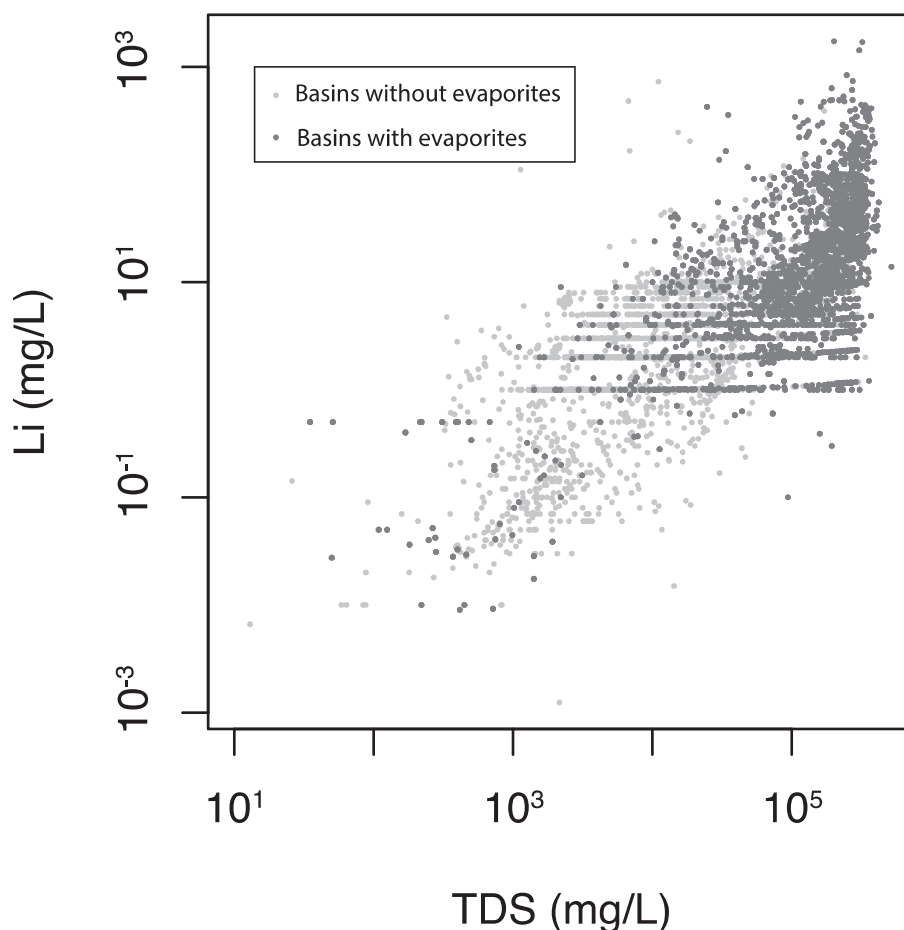


Fig. 2. Li^+ concentration versus total dissolved solids (TDS) of formation waters from basins with (darker grey) and without (lighter grey) extensive marine evaporite deposits. Data compiled from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018).

ppm (Fig. 7).

4.2. Estimated lithium production rates

Brines have been historically produced at a rate of 4.9×10^8 L/yr from brine production wells in the Smackover Formation (Arkansas Department of Energy and Environment, 2023). If these wells fluids had the median concentration of Li^+ of 84 mg/L, this would equate to 41 tpy (Table 1). At the 95th percentile of Li^+ concentration of 396 mg/L, 190 tpy per well could be produced.

In the Duperow, Birdbear and Midale carbonates of the Williston Basin, injection rates at disposal wells were used as a proxy for possible production rates (Table 1). Oil wells typically operate at lower rates and are not optimized to produce water. Approximately, 2.3×10^7 to 1.3×10^8 L/yr could be produced (IHS, 2023), which would result in 0.58 to 1.4 tpy/well of Li production at the median concentration of 10 to 33 mg/L and as much as 9.2 tpy/well of Li production at the 95th percentile of Li concentration of 70 mg/L in the Duperow Formation (Table 1). Production from the Bakken Formation would be more limited despite its higher median Li^+ concentration of 45 mg/L. Production of water from the Bakken Formation occurs at a median rate of 1.2×10^6 L/yr per well (IHS, 2023), which would result in Li production of 0.05 tpy/well.

Production of Li from the Marcellus Shale would be limited by low fluid production rates. Wells in the Marcellus Shale have historically produced 1.7×10^5 L/yr of water (Kondash et al., 2017). At the median concentration of Li^+ of 65 mg/L, this would result in 0.01 tpy/well (Table 1). At the 95th percentile of Li^+ concentrations of 214 mg/L, 0.04 tpy/well could be produced.

The Utah Department of Natural Resources (2023) reports a single Li

well in their database for the Paradox Basin. This well installed in the Paradox Formation has produced water at a rate of 2.4×10^6 L/yr over the course of its history as both an oil well and Li well (Table 1). If this production rate is typical of wells completed in the Paradox Formation, where a Li^+ concentration of 110 mg/L has been observed, 0.26 tpy/well of Li could be produced (Table 1). A brine disposal well operated by the Bureau of Reclamation installed in the Leadville Formation has operated at a rate of 4.2×10^8 L/yr (King et al., 2016) (Table 1). If production of fluid was possible at a similar rate, this would result in production of 5.5 tpy/well of Li at the median concentration of Li^+ (13 mg/L) found in the Leadville Formation. At the 95th percentile Li^+ concentration (78 mg/L), approximately 33 tpy/well of Li may be produced.

5. Discussion

5.1. Geochemical characteristics of Li-enriched brines

The highest Li^+ concentrations in formation waters come from sedimentary basins containing remnant saline fluids at depth, derived from evaporation of seawater in the geologic past, and extensive evaporite and organic-rich shale deposits (Figs. 1 and 3). High Cl^- and Br^- concentrations, and low $\text{Cl}^-:\text{Br}^-$ mass ratios of saline fluids at depth in the Gulf Coast, Appalachian, Williston, and Paradox basins (Fig. 5a) are consistent with their origin as evaporated paleo-seawater, as shown in previous studies (Carpenter, 1978; Dresel and Rose, 2010; Hanor, 1994; Moldovanyi and Walter, 1992; Sanders, 1991). These saline fluids have been substantially modified by fluid-rock diagenetic reactions, with carbonates, clays and organic matter, that removed Mg^{2+} and K^+ and added Li^+ to solution (Fig. 5b-d).

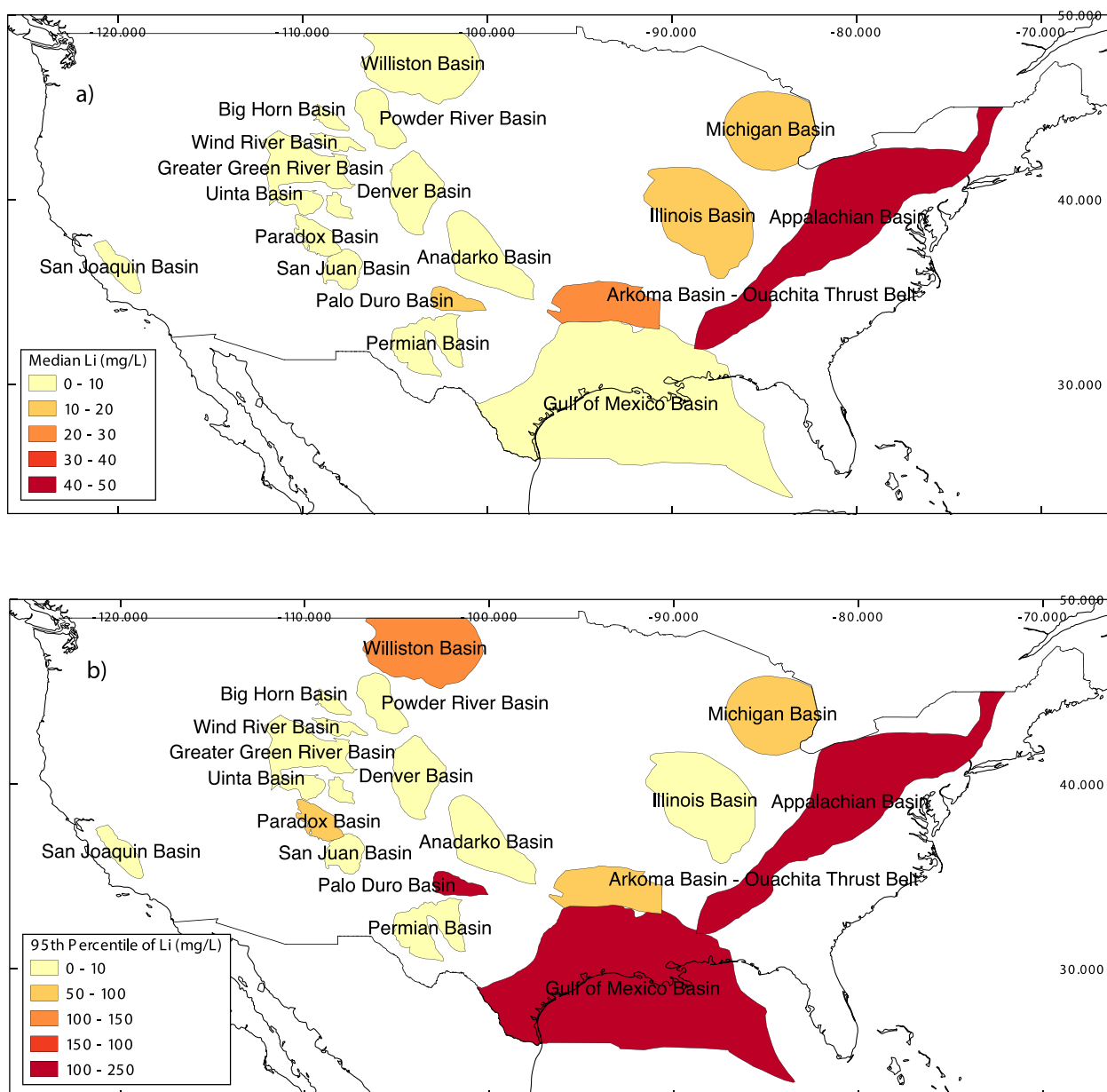


Fig. 3. Median (a) and 95th percentile (b) Li^+ concentrations in sedimentary basins across the United States. Data compiled from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018). Basins with higher Li^+ concentrations tend to contain evaporites and evaporated paleo-seawater with groundwater flow systems affected by the negative buoyancy of dense brines. The Gulf of Mexico basin includes the Arkla and East Texas basins.

Formation waters in the Gulf Coast, Appalachian, Williston, and Paradox basins with the highest Li^+ concentrations are depleted in Mg^{2+} and K^+ relative to evaporated seawater (Fig. 5b). Li-enriched brines in the Jiangnan Basin in central China have similar geochemical characteristics (Yu et al., 2023). Depletion of Mg^{2+} in basinal brines has been attributed to dolomitization of carbonates and precipitation of chlorite and ankerite minerals (Connolly et al., 1990), while depletion of K^+ is attributed to alteration of smectite to illite clay minerals, formation of authigenic K-feldspars, and precipitation of potash salts (Phan et al., 2016). Li^+ is known to adsorb onto smectite clays; conversion of smectite to illite at depth in sedimentary basins may release Li^+ to solution (Williams et al., 2015). Li^+ and K^+ are positively related (Fig. 5c), suggesting a common source or sink of these alkali elements in sedimentary basins. Dissolution of Mg-silicates at depth may also release Li^+ to solution (Milliken and Olson, 2017), although there was no apparent increase in Mg^{2+} for the highest Li^+ samples. Higher salinity, due to greater degrees of seawater evaporation, promotes greater diagenetic

reactions (Hanor, 2001), which may further enhance Li enrichment. This may partially explain why some of the most saline fluids within basins tend to be the most Li-enriched (Fig. 2).

Lithium also adsorbs onto organic matter, and as organic matter is converted to oil and gas, at depth under high temperatures and pressures, Li^+ may be concentrated in solution (Williams et al., 2015). The relatively high Li contents of organic-rich shales (Fig. 7) and associated brines (Fig. 4d) in the Paradox Formation of the Paradox Basin, comparable to other oil field brines (Collins, 1976), coals (Swaine, 2013), and shales (Clauer et al., 2014; Williams et al., 2013), supports this hypothesis. The organic-rich lithologies of the Paradox Basin, recognized as major petroleum source rocks, were deposited from algal blooms during cyclic flooding and desiccation of the basin (Whidden et al., 2014). They are accompanied by other clastic lithologies, dolomite, and gypsum as interbeds between halite intervals. These clastic units have been informally numbered from the top of the Paradox Formation downward, in similar fashion to the numbering of the halite

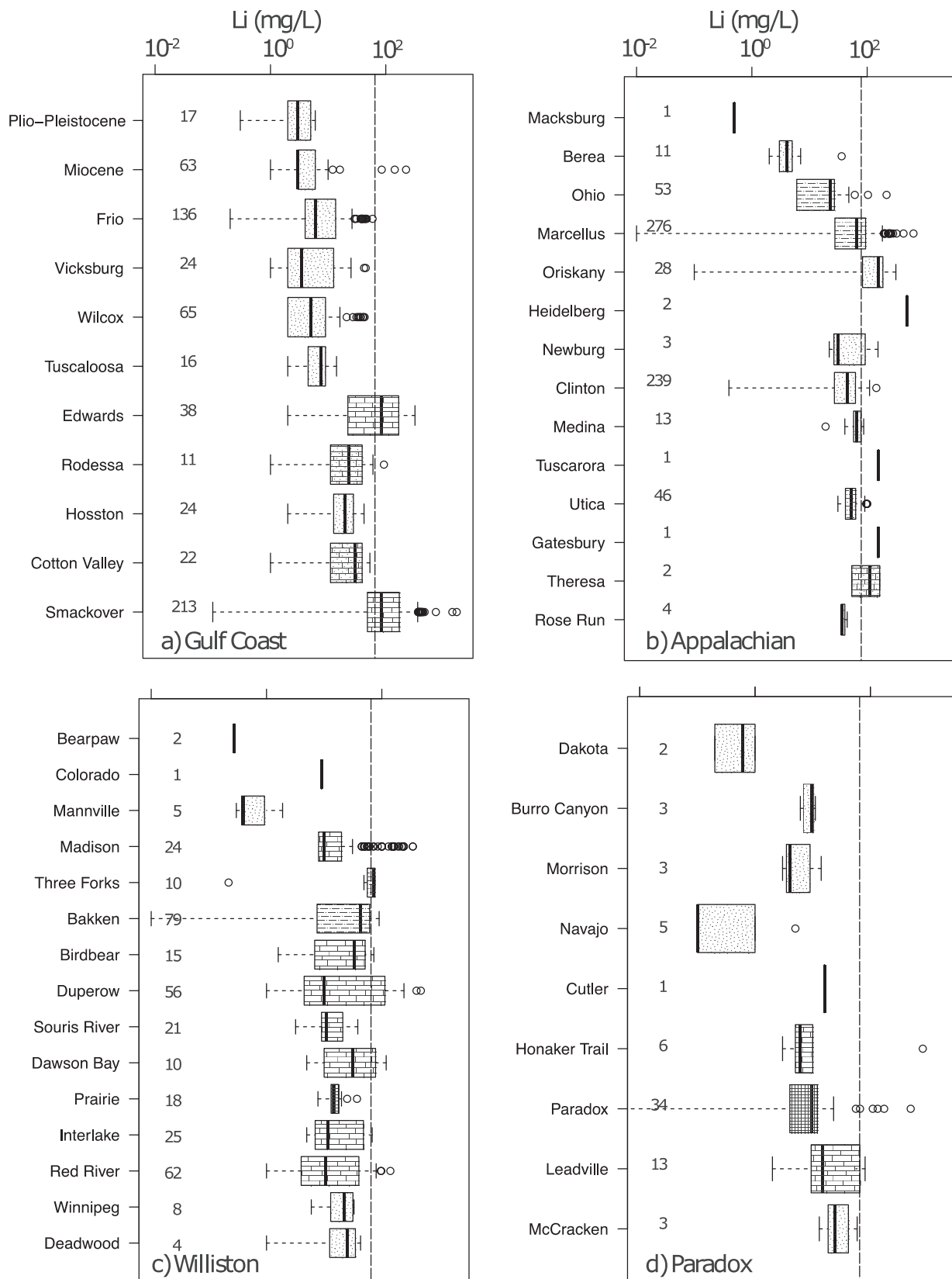


Fig. 4. Lithium concentrations for selected formations in the a) Gulf Coast (including Arkla and East Texas), b) Appalachian, c) Williston and d) Paradox basins. For the box and whisker plots, median and 1st and 3rd quartiles make up boxes, while 1.5× interquartile range make up the whiskers. The lithologic pattern (Federal Geographic Data Committee, 2006) within the boxes represents the dominant lithology of each geologic unit. The vertical dotted line represents 65 mg/L Li^+ , the lower potentially economic limit for Li production as suggested by prior studies (Tahil, 2008). Formation water data compiled from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018), as well as additional published sources (Blondes et al., 2020; Gallegos et al., 2021; Hiite, 1964; Mayhew and Heylmun, 1965; Mowat et al., 2021; Mowat, 2023; Peterman et al., 2017; Phan et al., 2016; Rowan et al., 2011; Tasker et al., 2020).

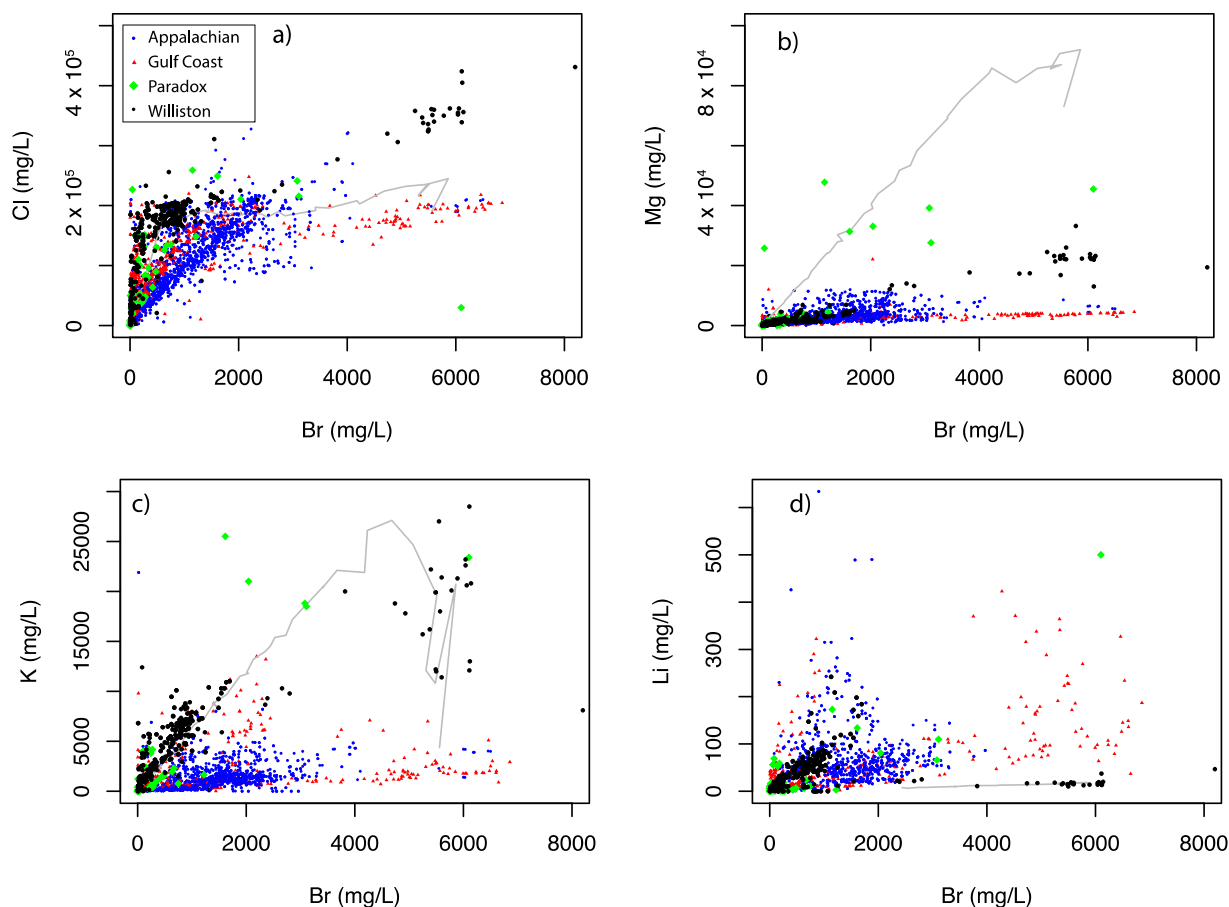


Fig. 5. Major ion (Cl^- , Mg^{2+} , K^+) and Li^+ versus Br^- in formation waters from the Gulf Coast (including Arkla and East Texas), Appalachian, Paradox and Williston basins. The solid grey line shows the evolution of solute compositions during seawater evaporation (McCaffrey et al., 1987). Data compiled from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018), as well as additional published sources (Blondes et al., 2020; Gallegos et al., 2021; Hite, 1964; Mayhew and Heylman, 1965; Mowat et al., 2021; Mowat, 2023; Peterman et al., 2017; Phan et al., 2016; Rowan et al., 2011; Tasker et al., 2020).

units (Hite et al., 1984). Some of the clastic units contain organic-rich lithologies (black shales) with elevated Li concentrations in the rock (>100 ppm; Table S4), probably corresponding to the elevated concentrations in the brines (Tables S1 and S2). Some of the clastic units have also been found to contain relatively permeable zones capable of producing Li-brines (e.g., Anson Resources, personal communication). Previous studies of the Smackover and Bakken formations in the Gulf Coast and Williston basins, respectively, have also suggested that brines became enriched in Li^+ via fluid-rock reaction with shales (Peterman et al., 2017; Stueber et al., 1984).

Li-rich brines in carbonate and sandstone reservoirs, adjacent to evaporite and shale units, indicates that brines migrated laterally or via faults (Hanor and McIntosh, 2007; Macpherson, 2015; Moldovanyi and Walter, 1992; Moldovanyi et al., 1993). Alternatively, fluid circulation through underlying basement rocks may have added Li^+ in some cases (Eccles and Berhane, 2011; Garrett, 2004; Huff, 2019). Future studies are needed to identify Li^+ concentrations and sources in sedimentary basin fluids. Li isotopes, in particular, show promise for fingerprinting Li sources and reactions controlling solute concentrations (Chan et al., 2002; He et al., 2020; Pfister et al., 2017; Phan et al., 2016; Williams et al., 2015; Yan et al., 2023).

Li-enriched saline fluids have been retained at depth in many sedimentary basins over geologic time by negative buoyancy (density trapping) and low permeability of shale and evaporite confining units (Ferguson et al., 2018). For example, Li-rich brines are retained in reservoirs within and adjacent to shale source rocks and evaporites in the Gulf Coast, Appalachian, Williston, and Paradox basins (Fig. 4a-d).

Along basin margins, in higher-permeability reservoirs, and in places

with high hydraulic gradients, saline fluids have been partially or completely flushed in many cases (McIntosh and Ferguson, 2021; McIntosh and Walter, 2005). This can be seen in formation waters in the Gulf Coast, Appalachian, Williston, and Paradox basins that have high $\text{Cl}^-:\text{Br}^-$ mass ratios (Fig. 6a) and plot along mixing trends between evaporated paleo-seawater derived brines and dilute meteoric recharge (Fig. 5a). Basinal brines derived from dissolution of evaporite minerals, with high $\text{Cl}^-:\text{Br}^-$, are not enriched in Li^+ , as Li^+ concentrations are relatively low in halite and potash salts (Bekele and Schmerold, 2020).

These results suggest that other basins and geologic formations that have had substantial meteoric influx are unlikely candidates for Li production. For example, siliciclastic reservoirs in the upper hydrostratigraphic units within the Paradox Basin have experienced substantial flushing over the past several million years with incision of the Colorado River, and regeneration of salinity via salt dissolution (Kim et al., 2022b; Tyne et al., 2022). Formation waters in these reservoirs are not enriched in Li^+ (Fig. 4d). The Permian Basin has had a similar dynamic history of incision and meteoric flushing of basinal brines (Stueber et al., 1998) and has relatively low Li^+ concentrations overall (Fig. 1). Shallow siliciclastic formations overlying the Edwards Aquifer in the Gulf Coast have also been flushed (Chowdhury et al., 2018; Land and Macpherson, 1992) and have low Li^+ contents (Fig. 4a).

5.2. Potential production rates of lithium from selected reservoirs

Potential Li production rates require estimation of fluid production rates along with Li^+ concentrations. Wells operated in sedimentary basins by the oil industry have been used to provide some insights into the

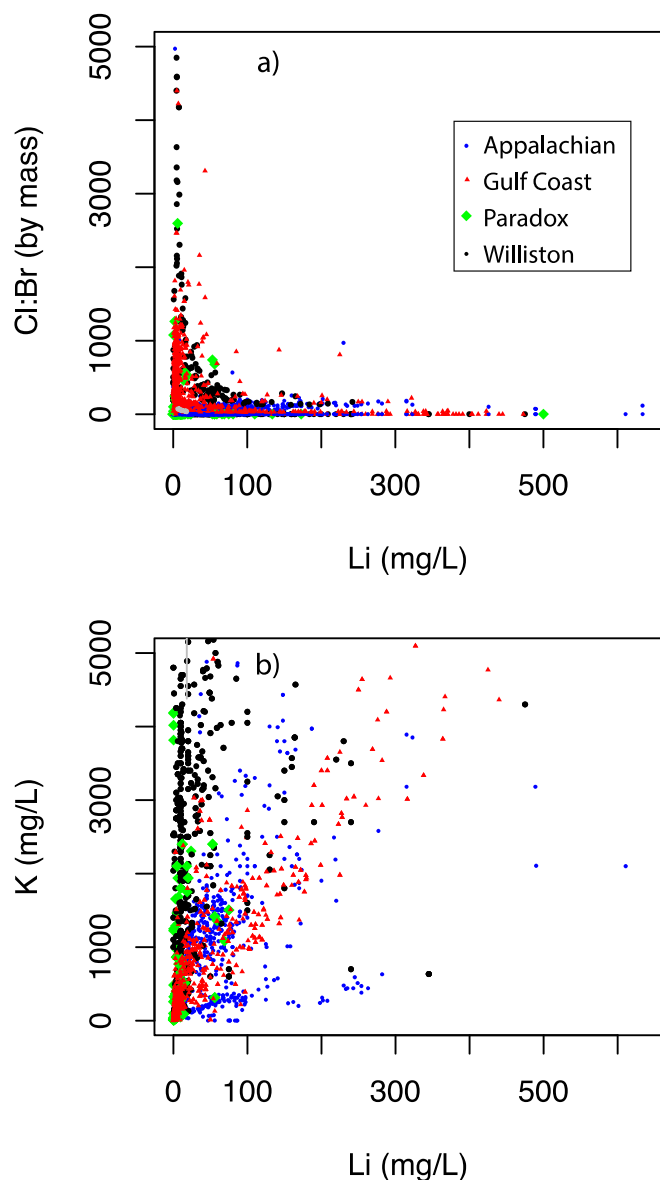


Fig. 6. Ratio of Cl^- to Br^- (both in mass units) (a) and K^+ (b) versus Li^+ concentration of formation waters from the Appalachian, Gulf Coast (including Arkla and East Texas), Paradox and Williston basins. Data compiled from the USGS National Produced Waters Geochemical Database (Blondes et al., 2018), as well as additional published sources (Blondes et al., 2020; Gallegos et al., 2021; Hite, 1964; Mayhew and Heylman, 1965; Mowat et al., 2021; Mowat, 2023; Peterman et al., 2017; Phan et al., 2016; Rowan et al., 2011; Tasker et al., 2020).

level of fluid production that is possible. Experience with dedicated brine production wells is more limited with wells in the Smackover Formation perhaps providing the best example.

The high flow rates in brine wells installed in the Smackover Formation can be partly attributed to the manner in which those wells are constructed and operated. The high permeability ($3 \times 10^{-14} \text{ m}^2$) of the Smackover Formation is also necessary to allow these flow rates. For strata where estimated Li production exceeded 0.5 tpy/well, permeabilities all exceed 10^{-15} m^2 (Table 1). Lower permeability strata, such as the Bakken Formation and Marcellus Shale, do not have the permeability to support high flow rates required to produce large amounts of Li, despite the high Li^+ concentrations present in these strata even following hydraulic fracturing. In some cases, thicker geologic units could produce greater fluid fluxes, offsetting lower permeabilities.

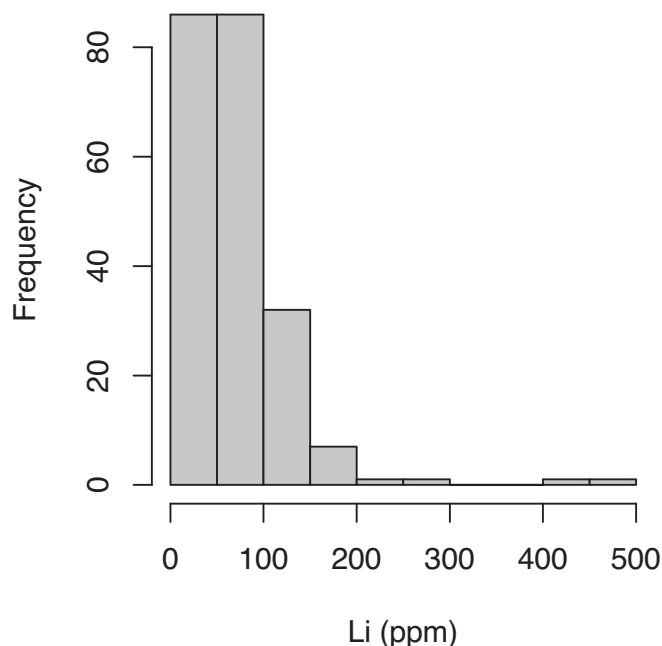


Fig. 7. Li^+ content of organic-rich shales in the Paradox Formation, Paradox Basin from new measurements, as part of this study (Tables S3-S4), and previously published data (Tuttle et al., 1996).

Low permeability may complicate Li production from the Paradox Formation, a relatively tight formation (Thackston et al., 1984). High flow rates (equivalent to $1.5 \times 10^7 \text{ L/yr}$) have been reported during hydraulic testing, but these have only been sustained for periods of hours to days (Mayhew and Heylman, 1965; Utah Department of Natural Resources, 2023). Production from the underlying Leadville Limestone could be an alternative in the Paradox Basin. Elevated Li^+ concentrations are present in some areas of the Leadville Limestone (95th percentile of 78 mg/L) and permeabilities as high as $8 \times 10^{-15} \text{ m}^2$ have been noted (King et al., 2016). This Li^+ may have originated via interaction with organic-rich shales in the Paradox Formation, suggesting that a source rock and reservoir model might be required to find appropriate targets for Li production.

Permeability is a key consideration in Li production from sedimentary basin brines (Fig. 8). High permeabilities are present in strata where high production rates have been estimated from observed fluid production and injection rates and concentrations. High Li^+ concentrations on their own are not sufficient as an exploration tool because they are often associated with low permeability strata. This can be explained by the variability in these two parameters: permeability can vary over ~ 13 orders of magnitude (Freeze and Cherry, 1977), while Li^+ concentrations vary over only ~ 3 orders of magnitude (Fig. 2).

Production of Li from sedimentary basin brines will need to look beyond co-produced waters from the oil and gas industry. For every L of oil produced, approximately 5 L of water is co-produced (Clark and Veil, 2009). For example, in 2021, $5.1 \times 10^{12} \text{ L}$ of oil was produced (BP, 2022), which suggests that approximately $2.6 \times 10^{13} \text{ L}$ of water was produced. At the median Li concentration of 5 mg/L, this would result in production of approximately 130,000 tpy, if all Li could be extracted from this fluid. Once extraction efficiencies are considered, this amount would be reduced to $\sim 50,000$ to 90,000 tpy. This suggests that recovery of Li from every oil and gas well in the world would equal ~ 40 to 70 % of the annual global production of 130,000 tpy (U.S. Geological Survey, 2023). Most of these oil and gas wells have Li^+ that is well below the 65 mg/L threshold for economic production, suggesting that Li production from sedimentary basin brines will not add substantially to production unless a large amount of development occurs. Environmental impacts from Li production will depend on the geologic setting, extraction

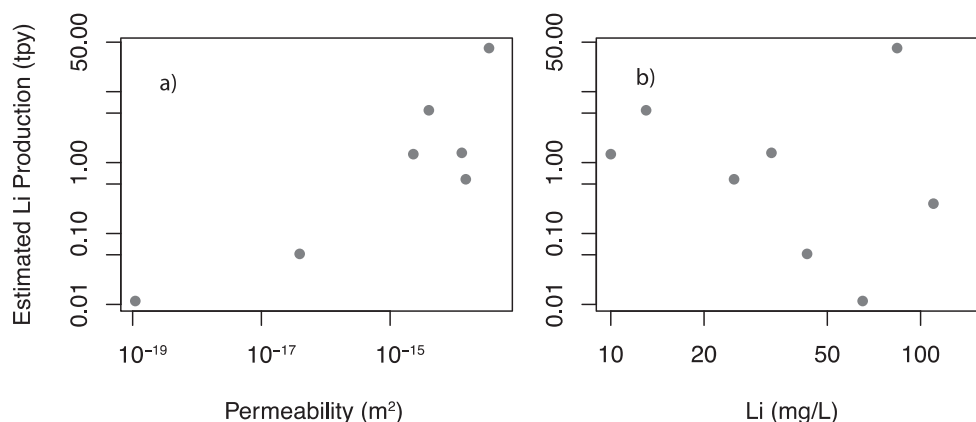


Fig. 8. Estimated lithium production (tons per year; tpy on a per well basis) versus reservoir permeability (a) and concentration of Li in formation waters (b). Data shown in black symbols correspond to various basins and geologic formations shown in Table 1.

techniques, and disposal of waste products (Flexer et al., 2018).

Opportunities to produce Li in conjunction with other types of development in sedimentary basins should also be explored. Co-production of other critical elements, such as Au, Ag, Co, Ga, Ge, Ln, Nb, Se, Te, Re, Br, platinum group elements, and rare earth elements, which are often enriched in sedimentary basin brines, may improve the economics of Li production (Alessi et al., 2021; Can Sener et al., 2021). Geothermal energy developments in sedimentary basins will require large fluid production rates (Ferguson and Ufodu, 2017), which could support Li production where Li^+ concentrations are sufficiently high. Potential Li production should also be considered in the broader context of pore space management. The potential to mitigate greenhouse gas emissions with carbon capture and sequestration (CCS) in sedimentary basins has been emphasized (Benson and Cole, 2008; Zoback and Smit, 2023). How and to what extent CCS might impact potential Li production is unclear.

6. Conclusions

Lithium concentrations in sedimentary basin formation waters are typically too low (median value of 5 mg/L) to support production of Li with current technology, indicating a need to better understand the factors leading to high Li^+ concentrations. Elevated Li^+ concentrations (>65 mg/L) are found in sedimentary basins containing remnant seawater associated with evaporite deposits and organic-rich shales. Brines with high Li^+ concentrations typically have low $Cl^-:Br^-$ ratios, indicating an evaporated seawater source of salinity. However, concentration of seawater is insufficient to explain the observed Li^+ concentrations. Diagenetic reactions that have removed Mg^{2+} and K^+ and added Li^+ , relative to evaporated seawater, appear to be necessary in many samples. Organic-rich shales, interbedded with evaporites, in the Paradox Basin are enriched in Li (median value of 60 ppm; range of 20–440 ppm). The presence of organic-rich shales may have acted as a source of Li^+ to formation waters in adjacent sandstones and carbonates in the Paradox and other basins. Brines with high Li^+ concentrations appear to occur in regions of sedimentary basins that have not been flushed with meteoric water (based on their chemical and isotopic composition and residence time), either due to the effects of negative buoyancy associated with dense brines, or due to the presence of low permeability strata.

Lithium production will be limited not only by the distribution of brines with elevated Li^+ concentrations, but also by the ability to produce these fluids. Permeability values in the strata examined here varied over five orders of magnitude, resulting in vastly different estimated fluid and Li production rates. The combination of high Li^+ concentrations and high permeabilities that would allow for production of large amounts of Li are present in only a subset of strata in sedimentary basins.

Sedimentary basins, at the current level of Li development and understanding, are unlikely to produce amounts of Li similar to salars or hard rock mining. However, as technology improves, basinal brines could be important in incrementally increasing global production and enhancing energy security. They may have certain advantages over salar production, which involves huge evaporation water losses and impacts on South American indigenous communities, and over difficult-to-permit conventional Li mines. Current research and experimentation on DLE may contribute to improved economics of Li recovered from sedimentary basin brines.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.gexplo.2023.107383>.

CRediT authorship contribution statement

Mohammad Marza: Formal analysis, Methodology, Writing – original draft. **Grant Ferguson:** Conceptualization, Formal analysis, Methodology, Supervision, Writing – review & editing. **Jon Thorson:** Formal analysis, Resources, Writing – review & editing. **Isabel Barton:** Investigation, Writing – original draft. **Ji-Hyun Kim:** Formal analysis, Investigation. **Lin Ma:** Formal analysis. **Jennifer McIntosh:** Conceptualization, Methodology, Supervision, Writing – review & editing.

Data availability

All data are shared in the supplemental tables

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