Supplementary Information

A Data-Driven Approach for Exploring Unconventional Lithium Resources in Devonian Sedimentary Brines, Alberta, Canada

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Contents

Section 1. Previous studies on investigating lithium anomalies in sedimentary brines

Supplementary Table S1 Studies investigating [Li] anomalies in sedimentary brines.

Region	Geologic properties				Geochemical properties																						Method	Objective	Reference
	Location	Geologic age	Sampling depth	Formation temperature	[Li]	[Na]	[K]	[Rb]	[Ca]	[Mg]	[Ba]	[Pb]	[Sr]	[Mn]	[B]	[Si]	[Cl]	[Br]	[I]	[HCO_3]	[SO_4]	[CO_3]	$\delta^{18}O$	δ^2H	TDS	pH			
Alberta, Canada	☑	☑	O		☑	☑	☑		☑	☑	O		O	O	O		O	O	O	O	O	O			☑	O	Neural-network model	Identify enriched environments with Li concentrations surpassing a specific cutoff level within the Devonian formations of Alberta.	This study
British Columbia, Canada	O	☑		☑	☑, *	☑	☑		☑	☑	O	O	☑, *		O		☑	☑	O		☑				☑		correlative analysis	Identify the in situ lithium concentrations and resource potential of deep saline brines in northeast British Columbia (NEBC).	Kaush et al, 2024
Alberta, Canada	O	☑			☑	☑							*				☑	☑					☑	☑	☑		correlative analysis	Hypothesis of Li origins for deep brines	Huff, 2016
Alberta, Canada	☑	O	O		☑, *	☑	☑		☑	☑	O	*	*	O	O		☑	☑		O	O				O	O	correlative analysis	Hypothesis of Li origins for deep brines	Eccles and Berhane, 2011
Alberta, Canada	O	☑			☑	☑											☑	☑									correlative analysis	Hypothesis of Li origins for deep brines	Bishop and Robbins, 2023
Alberta, Canada	O	☑			☑	☑	☑						*				☑	☑							☑		correlative analysis	Present the understandings of the characteristics and orgin of Li-rich brines	Lopez et al., 2018
Alberta, Canada	O		O		☑	O	O		O	O	O		O	O	O	O				O	O					O	Simple Kriging	Conduct preliminary economic assessment commerical lithium projects	Spanjers et al, 2017; MacMillan et al, 2021
Alberta, Canada	☑	☑	O	O	☑	O	O	O	O	O	O	O	O	O	O		O	O	O	O	O				O	O		Explore industrial mineral potential of Alberta deep brines	Brian et al, 1995
Jianghan Basin, China	☑	O			☑	☑			☑	☑			O		☑		☑	☑		☑	☑	☑			☑	O	correlative analysis	Investigate brine chemistry characteristics and determine Li enrichment mechanism.	Yu et al, 2023
Qaidam Basin, China	O	O	O		☑, *	☑	O	☑	O	☑							O	☑		O	O				☑		correlative analysis	Identify the areas with the highest [Li]	Li et al, 2021
Paris Basin, France	O	O	O	O	☑, *	O							☑, *		☑, *		O	O					O			O	correlative analysis	Explore the characteristics of Triassic formation waters	Millot et al, 2011
North German Basin, Germany	O	O			☑	O	O	O	O				O	O			O	O					O	O	O			Provide insights into the origin and fate of Paleozoic waters	Lüders et al, 2010
Heletz-Kokhav field, Israel	O	O	O	O	☑, *	☑	☑		☑	☑			☑			☑	☑	☑			☑						correlative analysis	Investigate the source of Li, the orgin and evolution of the brines	Chan et al, 2002
Appalachian Basin, USA		O			☑, *								*				☑	☑							☑		correlative analysis	Examine [Li] and its isotrope in formation waters of Marcellus Shale and discuss the Li resources and isotopic fractonation resulting from water-rock interation	Phan et al., 2016
Appalachian and Gulf Coast basin, USA	O	O	O	☑	☑, *												☑										correlative analysis	Understand the [Li] distribuition and explore characteristics of the lithium isotope ratio systematics	Macpherson et al, 2015
Southern Arkansas, USA	☑	☑	☑	☑	☑																						Random forest model	Predict [Li] distribution in Smackover formation brines in the gulf coast region for a regional resource esitmation for Li.	Knierim et al, 2024
Various regions	O	☑	☑	☑	☑					☑															☑		correlative analysis	Study geochemistry and global resources estimation in sedimentary formation waters with Li	Dugamin et al., 2021

NOTE:

Cations: Li = Lithium, Na=Sodium, K=Potassium, Rb=Rubidium , Ca=Calcium , Mg=Magnesium , Ba=Barium , Pb=Lead , Sr=Strontium , Mn=Manganese , B=Boron , Si=Silicon.

Anions: Cl=Chloride, Br=Bromine, I=Iodide, HCO_3=Bicarbonate ion, SO_4=Sulphate ion, CO_3=Carbonate ion.

Isotopes: $\delta^{18}O$ is the ratio of isotopes oxygen-18 (18 O) and oxygen-16 (16 O); δ^2H is the stable isotope ratio of hydrogen.

TDS stands for Total Dissolved Solids.

PH stands for potential of hydrogen.

[X] stands for the concentration of X ion.

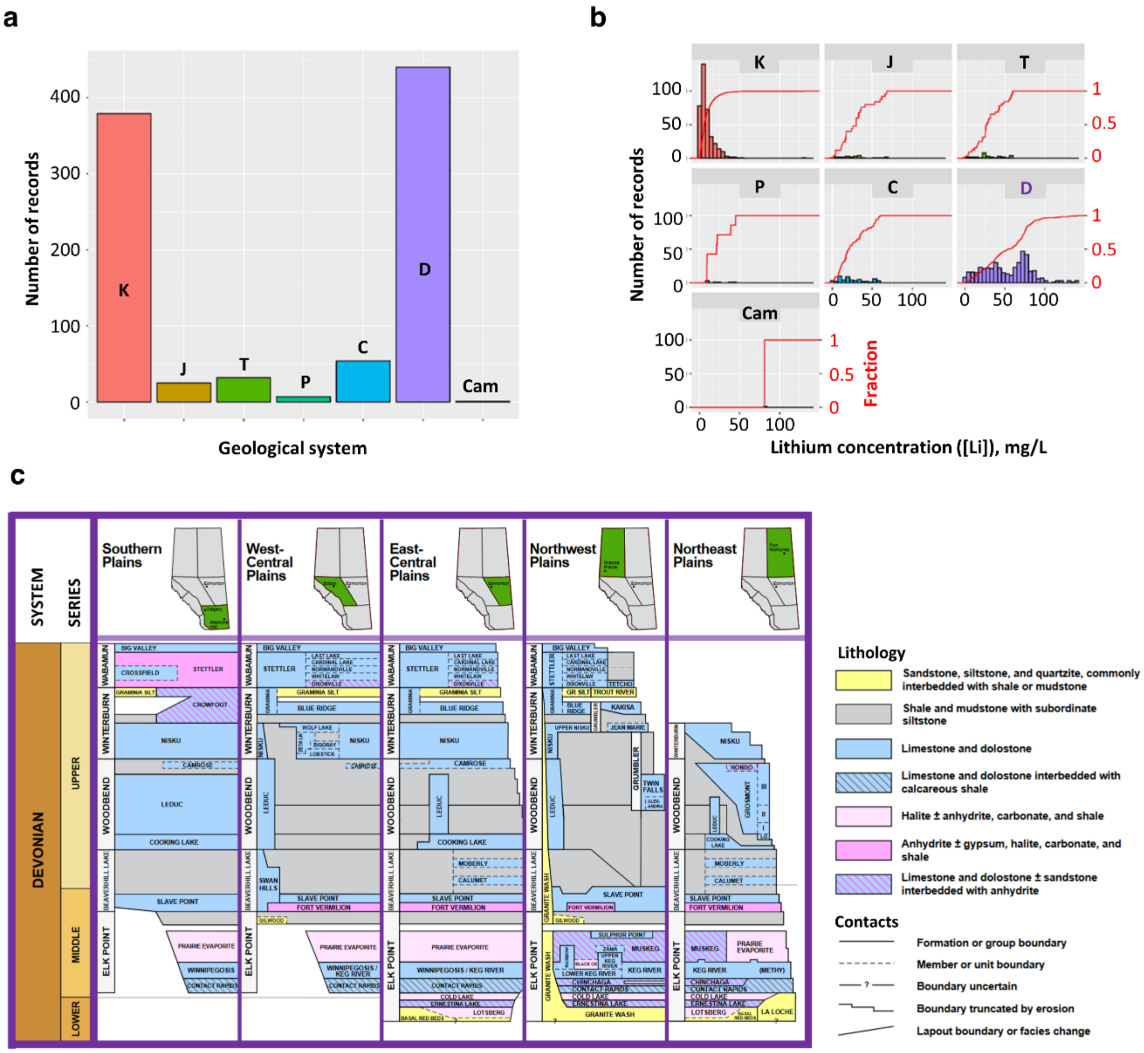
O represents the primary or secondary data were in the reported dataset but were not discussed in terms of [Li].

X represents the primary or secondary data were included and analyzed in terms of [Li].

***** represents the isotrope ratios of the coresponding element were analyzed in terms of [Li].

No filled cell the data is not available or explictly identified in literature.

Section 2. Background information of dissolved lithium in Alberta sedimentary brines



Supplementary Figure S1 (**a**) Distribution of Alberta water samples with lithium (Li) measurements, grouped by geological systems; (**b**) Histograms of lithium concentrations ([Li]) categorized by geological systems for data shown in (**a**); and (**c**) Stratigraphic columns of Devonian geological units across five Alberta regions, modified from Alberta Geology Survey (2019). Notes: K for Cretaceous; J for Jurassic; T for Triassic; P for Permian; C for Carboniferous; D for Devonian; and Cam for Cambrian.

The exploration of dissolved lithium in sedimentary brines began in Alberta as early as 1990 (Connolly et al., 1990a, 1990b). Supplementary Figure S1a shows the histogram of brine samples with [Li] measurements from various geological systems in Alberta based on water chemistry results available in public domain (Connolly et al., 1990b, 1990a; Dufresne, 2011; Eccles, 2018a, 2018b, 2018c; Eccles and Berhane, 2011b; Eccles and Dufresne, 2018; Eccles and Jean, 2013;

Huff et al., 2019, 2013, 2011; Lappin, 2019; Reimert et al., 2023, 2022), in which the Cretaceous and Devonian strata comprise 87% of the data.

In terms of [Li] distribution by strata, Supplementary Figure S1b shows the following observations. 1) The interquartile range of [Li] in the Cretaceous system spans from 3.00 mg/L to 12.00 mg/L, with an average of 9.05 mg/L and a median of 6.00 mg/L. 2) The [Li] in Devonian Strata range from 29.70 mg/L (the first quartile) to 74.53 mg/L (the third quartile), with an average of 52.88 mg/L and a median of 54.50 mg/L. 3) The other strata have much smaller data size and higher statistical dispersion.

Supplementary Figure S1c presents stratigraphic columns of Devonian geological units across five regions in Alberta, which are the focus of this study for exploring lithium resources. The Devonian strata in the Western Canada Sedimentary Basin (WCSB) exhibit a broad distribution and a diverse range of rock types, including carbonates, shales, sandstones, siltstones and evaporites. Carbonates, such as limestones and dolomites, dominate the upper and middle parts of the Devonian sequence. These carbonates form extensive reef complexes, such as those in the Leduc Formation, which not only host significant oil and gas accumulations (Wendte et al., 1992) but also represent potential unconventional lithium resources, as shown in Supplementary Figure S1b. In contrast, sandstones and siltstones are less prevalent, primarily occurring in the lower parts of the Devonian sequence (Meijer Drees et al., 1994). Current observations suggest that these lower units have a lower likelihood of hosting lithium-rich brines compared to the upper and middle Devonian units (Figure 6 of the main text).

Section 3. Newly acquired water chemistry data of Li brine from Devonian formations

Supplementary Table S2 Properties of Devonian produced water samples tested by Geological Survey of Canada-Calgary (GSC-Calgary).

UWI	GeoUnit	Lat_NAD83	Long_NAD83	Depth, m	Sampling Date	[Li][1,a], mg/L	[Ca][1,b], mg/L	[K][1, b], mg/L	[Mg][1,b], mg/L	[Na][1, b], mg/L	[B][2, c], mg/L	[Mn][2, d], mg/L	[Sr][1,e], mg/L	[Ba][1,f], mg/L	[Cl][4,g], mg/L	[Br][4,g], mg/L	[SO4][4,g], mg/L	[HCO3][5,h], mg/L	pH[i]	TDS[3], mg/L
100012105822W400	Leduc Formation	54.023529	-113.201993	1004.3	2023-02-20	19.70	4880	1050	1560	34300	50.6	0.0464	208.0	0.120	62600	254	1210	247	6.9	82600
102010505821W400	Leduc Formation	53.979817	-113.076344	980.5	2023-02-20	17.20	4280	935	1510	33700	44.7	0.0414	254.0	0.312	60900	243	845	255	7.1	78600
100091505721W400	Leduc Formation	53.929024	-113.028777	970.2	2023-02-20	16.20	4130	865	1490	28100	42.7	0.0341	261.0	0.393	60400	213	809	263	7.2	75900
100151905620W400	Leduc Formation	53.859793	-112.960245	958.3	2023-02-20	14.00	4500	694	1480	30500	36.5	0.0365	115.0	0.068	55900	208	1900	257	7.3	70900
100051405621W400	Leduc Formation	53.838093	-113.022019	988.2	2023-02-20	14.90	4770	755	1460	27300	37.1	0.0372	92.4	0.080	58500	224	2170	254	7.2	75000
100121306312W500	Swan Hills Formation	54.452331	-115.686358	2688.9	2023-02-21	14.50	2280	923	339	41300	82.3	0.0482	151.0	1.630	67400	197	890	249	7.3	93400
100080606411W500	Swan Hills Formation	54.506927	-115.643523	2762.4	2023-02-21	12.20	1930	749	308	29300	62.8	0.0409	117.0	0.847	54500	140	1210	180	7.5	80500
100042206311W500	Swan Hills Formation	54.459219	-115.585148	2770.9	2023-02-21	10.20	1830	523	289	24100	43.3	0.0578	99.5	0.617	44500	132	1360	189	7.4	67400
102040906410W500	Swan Hills Formation	54.515501	-115.440798	2559.1	2023-02-21	8.87	1560	478	263	21000	36.3	0.022	88.4	0.621	38600	102	1200	183	7.5	61000
100060706410W500	Swan Hills Formation	54.521482	-115.502489	2640	2023-02-21	8.92	1590	472	270	21000	36.2	0.0222	88.6	0.607	38600	110	1260	171	7.5	59300
100102906119W500	Swan Hills Formation	54.30667	-116.810814	3285.7	2023-02-21	79.90	21500	4130	2070	57500	141.0	0.342	1190.0	32.300	142000	451	168	< 1	7.1	109000
100162006119W502	Swan Hills Formation	54.295837	-116.804657	3263.6	2023-02-21	72.20	20100	3840	1880	52100	130.0	0.312	1050.0	32.400	130000	401	131	< 1	7.3	105000
100080306119W500	Swan Hills Formation	54.244895	-116.754742	3409.8	2023-02-21	61.70	21600	4150	1780	55600	137.0	0.293	1080.0	31.100	140000	432	146	< 1	7.4	112000
100012306220W500	Swan Hills Formation	54.370673	-116.881053	3282	2023-02-21	67.90	25700	4880	1900	49400	135.0	0.478	1290.0	31.500	141000	374	164	No data	7.1	265000
UWI	**GeoUnit**	**Lat_NAD83**	**Long_NAD83**	**Depth_m**	**Sample_Date**	**[Li]*, mg/L**	**[Ca]*, mg/L**	**[K]*, mg/L**	**[Mg]*, mg/L**	**[Na]*, mg/L**	**[B]*, mg/L**	**[Mn]*, mg/L**	**[Sr]*, mg/L**	**[Ba]*, mg/L**	**[Cl]*, mg/L**	**[Br]*, mg/L**	**[SO4]*, mg/L**	**[HCO3]*, mg/L**	**pH***	**TDS[0], mg/L**
100102407506W503	Gilwood Member	55.520168	-114.791375	0	2023-03-30	14.0	17800	571	2820	63300	Not tested	Not tested	Not tested	Not tested	113000	Not data	223	Not tested	5.5	195223
100140104123W400	Nisku Formation	52.504669	-113.185187	1800	2023-05-30	64.4	17400	4680	2930	55400	Not tested	Not tested	Not tested	Not tested	135060	750	398	Not tested	7.6	216275
100143604023W400	Nisku Formation	52.488619	-113.18939	1800	2023-05-30	66.7	17900	4840	3010	57300	Not tested	Not tested	Not tested	Not tested	138300	755	266	Not tested	6.7	221624
100161404123W400	Nisku Formation	52.53477	-113.196566	1800	2023-05-31	57.8	15500	4170	2660	54600	Not tested	Not tested	Not tested	Not tested	128110	670	944	Not tested	6.8	205989
100020904123W400	Nisku Formation	52.508925	-113.254965	1800	2023-05-31	67.7	17900	4770	3090	58600	Not tested	Not tested	Not tested	Not tested	139400	750	274	Not tested	6.8	224054

PARAMETERS:
UWI: Unique Well Identifiers.
GeoUnit: Geological stratigraphic Unit.
Lat_NAD83 and Long_NAD83: Latitude and longitude in the North American Datum of 1983 (NAD83)
Cations: Li for Lithium, Ca for Calcium, K for Potassium, Mg for Magnesium, Na for Sodium, B for Boron, Mn for Manganese, Sr for Strontium, and Ba for Barium.
Anions: Cl for Chloride, Br for Bromine, SO4 for Sulfate, and HCO3 for Bicarbonate.
TDS: Total Dissolved Solids.

MEASUREMENT METHODS:
[0] calculated from the sum of all measured ions.
[1] measured by ICP-OES (Inductively Coupled Plasma Optical Emission spectroscopy).
[2] measured by ICP-MS (Inductively coupled plasma mass spectrometry).
[3] measured by ISE (Ion selective electrode).
[4] measured by IC (Ion chromatography).
[5] measured by TITR (Titration).
* not reported in the original lab report.

DECTION LIMITS:
[a] 0.05 mg/L.
[b] 0.1 mg/L.
[c] 3 μg/L.
[d] 0.1 μg/L.
[e] 10 μg/L.
[f] 20 μg/L.
[g] 0.03 mg/L.
[h] 1 mg/L $CaCO_3$.
[i] 0.1.

OTHER COMMENTS:
Initially, the analysis included 36 elements tested using ICP-OES, 60 elements using ICP-MS, 7 elements using IC, and 4 elements using titration (TITR) for brines from nine formations: Cardium Formation, Duvernay Formation, Ellerslie Member, Leduc Formation, Mannville Formation, Rock Creek Member, Sunburst Sandstone Member, Swan Hills Formation, and Viking Formation. However, due to the limited table size and the scope of this study, only data relevant to the research are presented here.

Section 4. Data preprocessing

Supplementary Table S3 Criteria of data culling.

Culling level	Reject criteria	Reason for removing	Dataset: Exploration[1]	Dataset: Validation[2]	Dataset: Targeting[3]
0	Water sample is not in Alberta	Out of scope	☑	☑	☑
0	Water sample is not from Devonian formation		☑	☑	☑
0	Sampling geographic coordinates are missed	Missing information	☑	☑	☑
0	Content of Li is missed		☑	☑	NA
1	Weak constrain: 0 < Density (kg/L) < 0.95	Alcohols contamination (Kaush et al., 2024)	☑	NA	NA
1	TDS (mg/L) < 1000	Belong to fresh water (Wikipedia contributors, 2024)	☑	☑	☑
1	Duplicated well Identifiers and geographic location[4]	Duplication	☑	☑	☑
1	Weak constrain: Ion balance[5] < 0.8 or Ion balance[5] > 1.2	Ion charge inblance (Brian et al., 1995, alsglobal.com, 2022)	☑	☑	☑
1	Weak constrain: Calculated TDS / Reported TDS < 0.8	Misleading information	NA	☑	☑
1	Contents of TDS, [Ca], [K], [Mg], and [Na] are missed	Missing information	☑	☑	☑
1	Sampling formation name is missed		☑	☑	☑
1	Beyond the geographic and chemical data ranges in the **Culling Level1 of exploration dataset**		NA	☑	☑
2	Contents of [Cl], [Br] and [SO_4], pH, and density are missed	Missing information	☑	☑	-
2	Beyond the geographic and chemical data ranges in the **Culling Level2 of exploration dataset**		NA	-	-
3	Contents of [B], [Ba], and [Sr] are missed	Missing information	☑	☑	-
3	Beyond the geographic and chemical data ranges in the **Culling Level3 of exploration dataset**		NA	-	-

NOTE

[1] The water chemistry data with lithium measurements in Alberta and British Columbia reported by multiple sources (before May 2022).

[2] The water chemistry data with lithium measurements released by AER (2022) and collected by GSC-Calgary (May 2022 to June 2023)

[3] The historical water chemistry data without lithium measurements were archived in GSC-Calgary in-house database.

[4] Based on the characteristics of duplicated data, the record with max. TDS is selected to represent the rest of the records with duplicated well indentifers.

[5] Ion balance = Cation[a] sum (meq/L) / Anion[b] sum (meq/L), where

[a] Cations include Li^+, Ca^{2+}, K^+, Mg^{2+}, and Na^+

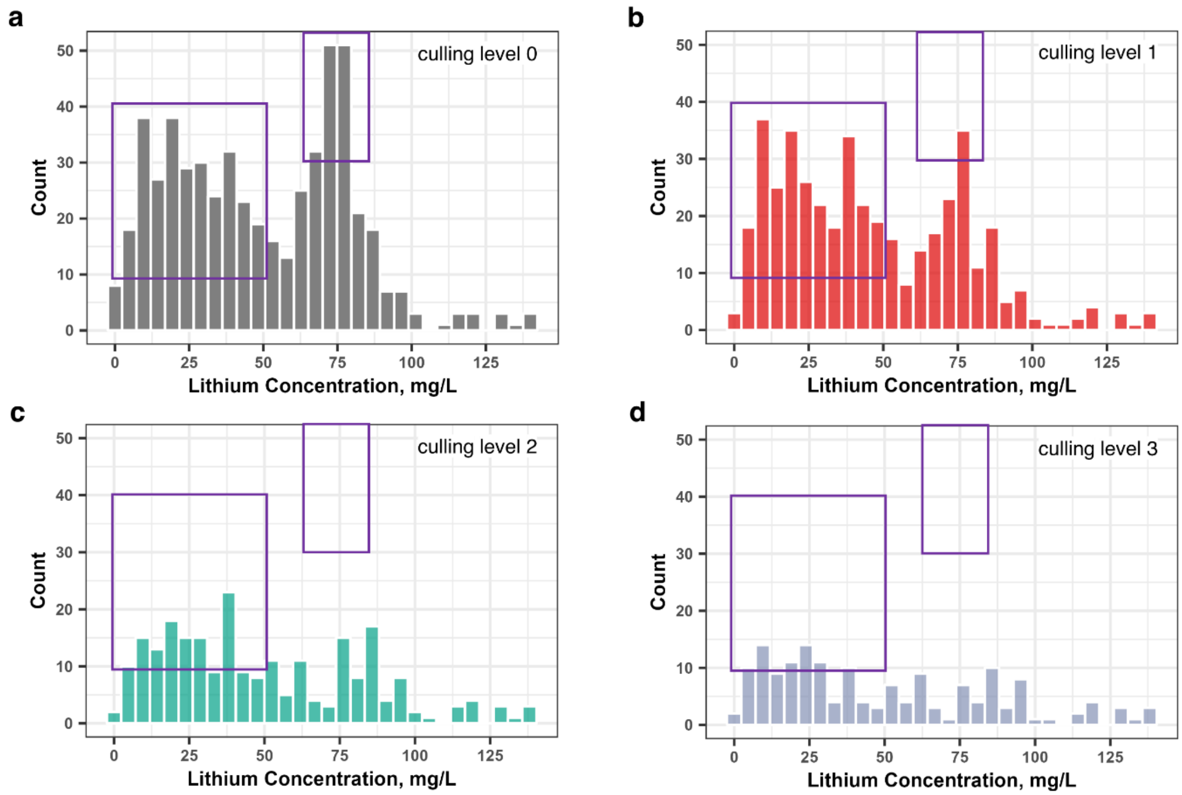
[b] Anions include Cl^-, Br^- and SO_4^{2-}

Weak constrain: If the relevant data available, then constrain will be calculated for data cleaning. Otherwise, the data will not be removed.

NA: The relevant data needed is not available.

-   : The data culling is not conducted.

Supplementary Table S3 lists the rules of data culling that have been applied for each dataset. The higher the culling level is, the more features in the dataset will go through data quality check.



Supplementary Figure S2 Histograms of [Li] distribution in the exploration dataset at: a, culling level 0; b, culling level 1; c, culling level 2; d, culling level 3. Notes: 1) The two purple boxes are positioned at the same locations in each sub-figure to facilitate observation and comparison of the changes in [Li] distribution. 2) Details of culling levels can be found in Supplementary Table S3.

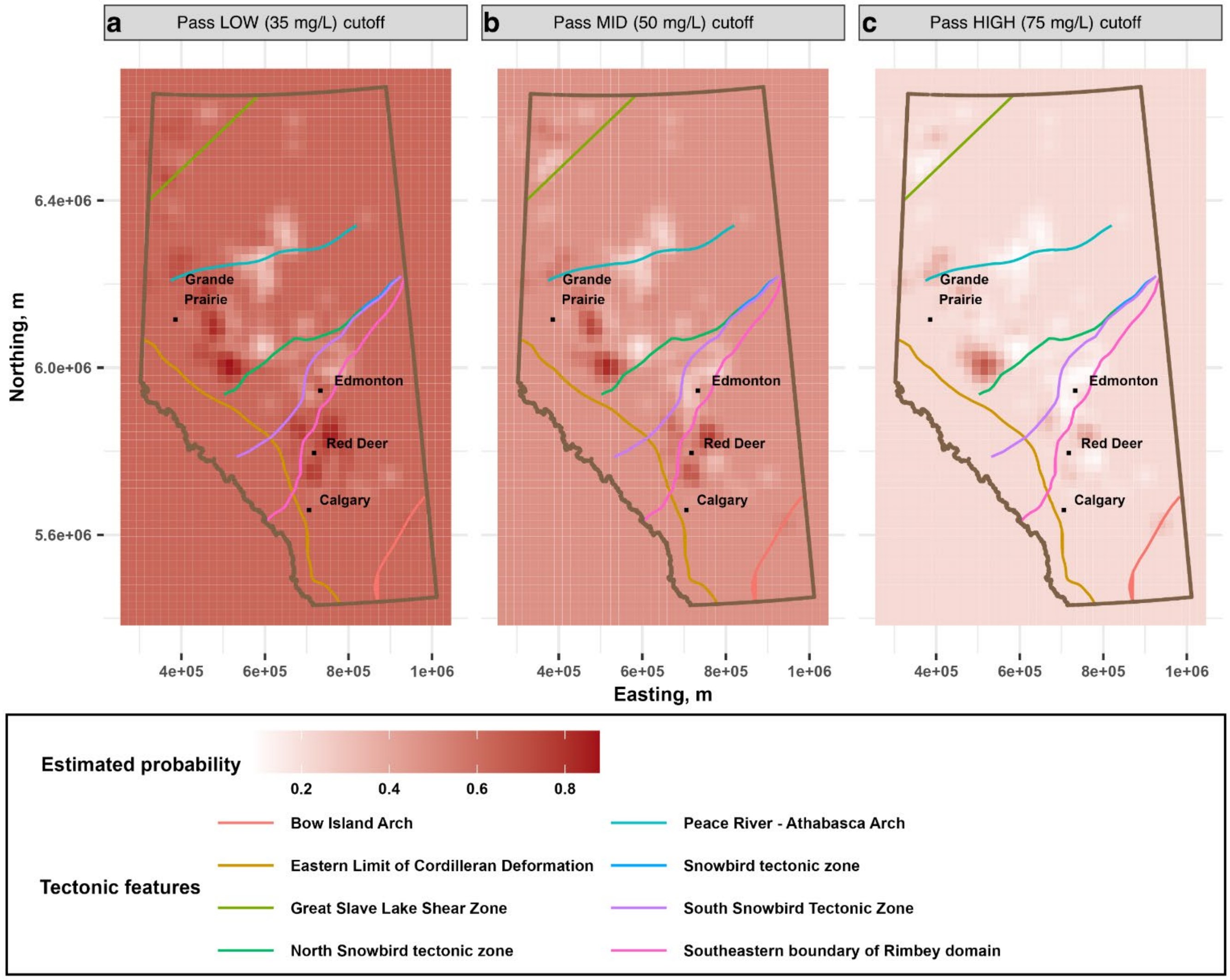
Supplementary Figure S2 displays how the [Li] distribution changed as culling level increases. It indicates that culling levels 2 and 3 can significantly reduce the dataset size and alter the characteristics of data distribution. Numerous observations with [Li] below 50 mg/L and around 75 mg/L are lost, resulting in a narrowed value range of other spatial and geochemical features for training. To retain as much information as possible, all datasets (exploration, validation, and targeting) are culled at level 1, as described in main text section of "Data compilation and preprocessing".

Supplementary Table S4 Properties of geochemical data in the culled datasets.

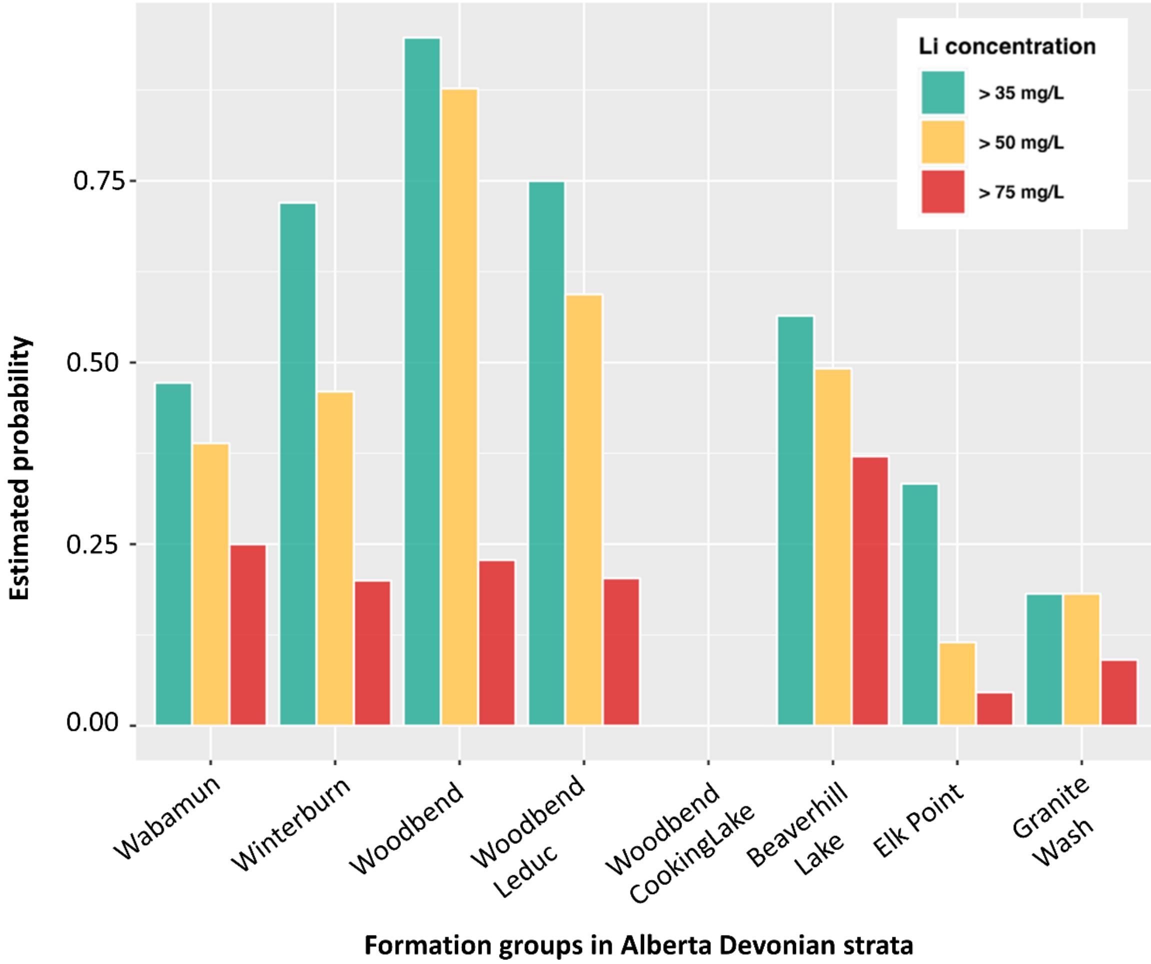
Selected water chemistry under the culling level 1	**Exploration dataset** n = 433		**Validation dataset** n = 102		**Targeting dataset** n = 897	
	Min.	Max.	Min.	Max.	Min.	Max.
[Li] (mg/L)	0.7	140	4.4	82.8	NA	NA
[Ca] (mg/L)	110	38700	400	35640	189	37398
[K] (mg/L)	65	8600	118	8620	67	8600
[Mg] (mg/L)	74	7800	71	3730	76.7	7160
[Na] (mg/L)	5380	100800	12000	72400	5490	91700
TDS (mg/L)	14062	335485	38931	296974	19000	323835

The geochemical parameters in both validation and targeting datasets fall within the corresponding ranges of the exploration dataset (knowledge pool).

Section 5. Data transformation

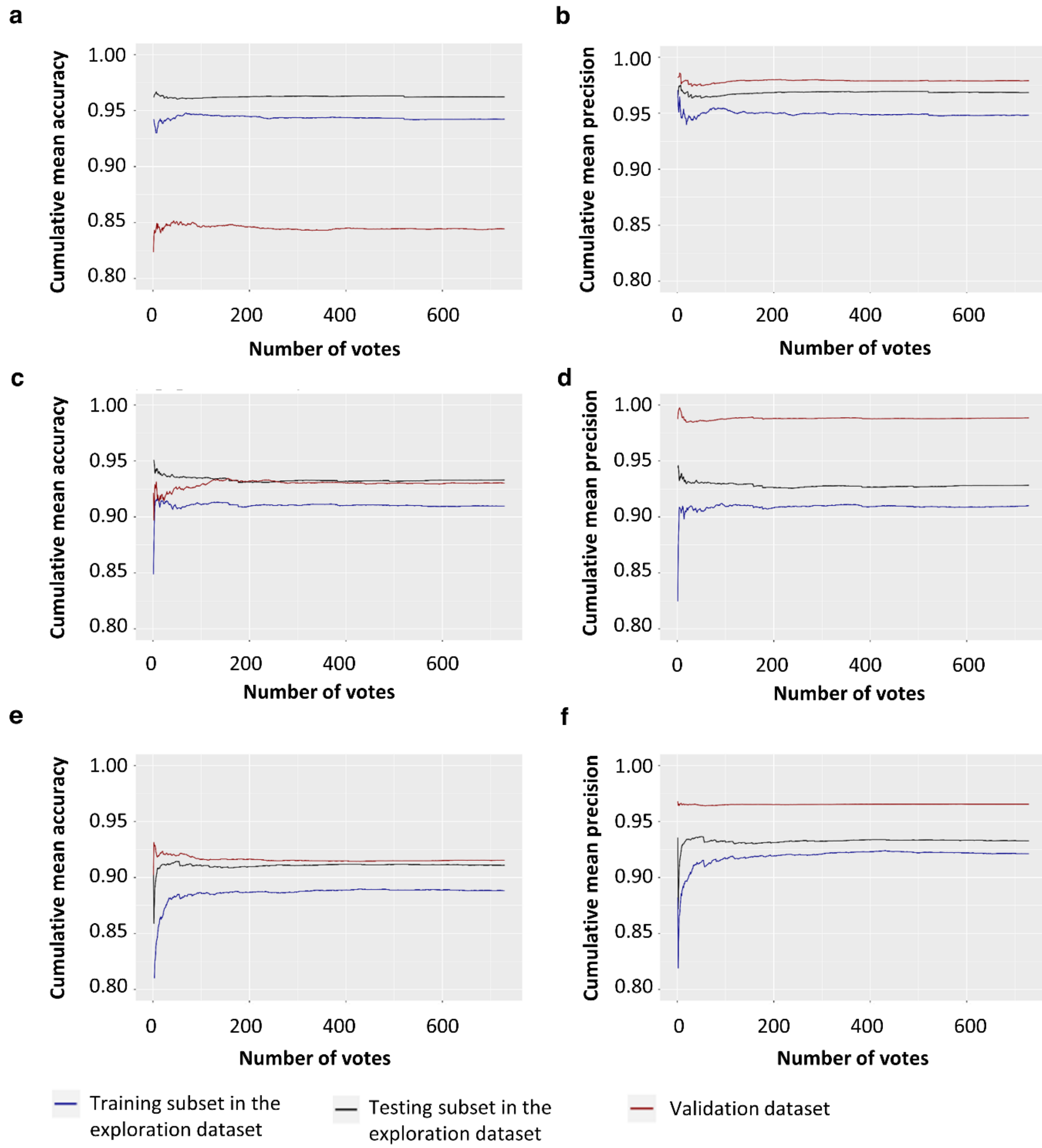


Supplementary Figure S3 Maps of estimated probability of Li brine distribution by three cutoff levels. The grid dimensions are 19,312 m by 19,312 m, which equals an area of four townships. Calculations are derived from the exploration dataset and applied for estimations in both the validation and targeting datasets.

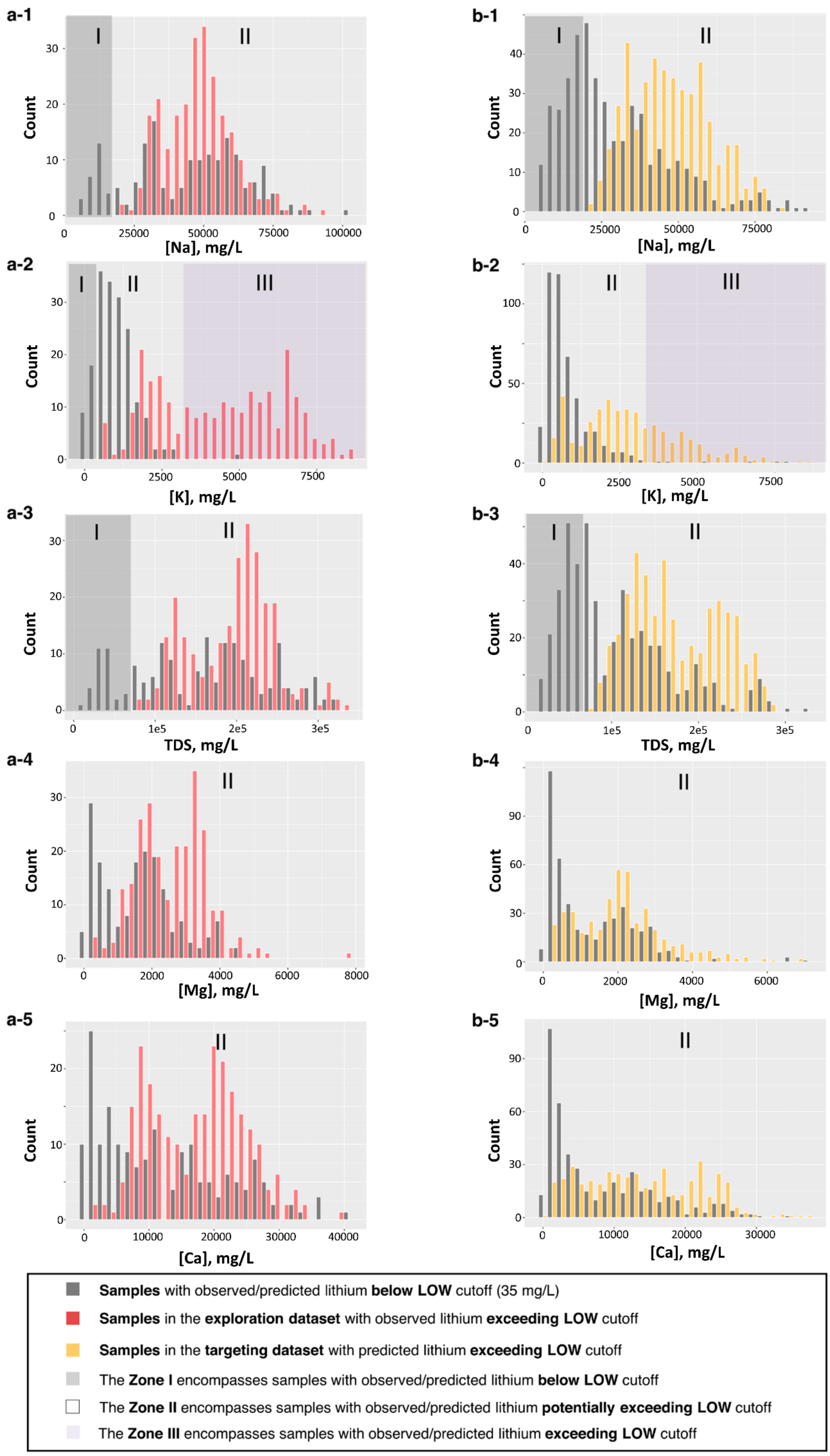


Supplementary Figure S4 Estimated probability of [Li] meeting predetermined cutoffs by formation groups. Calculations are derived from the exploration dataset and applied for estimations in both the validation and targeting datasets. Generally, the geological groups are ordered from newer to older periods.

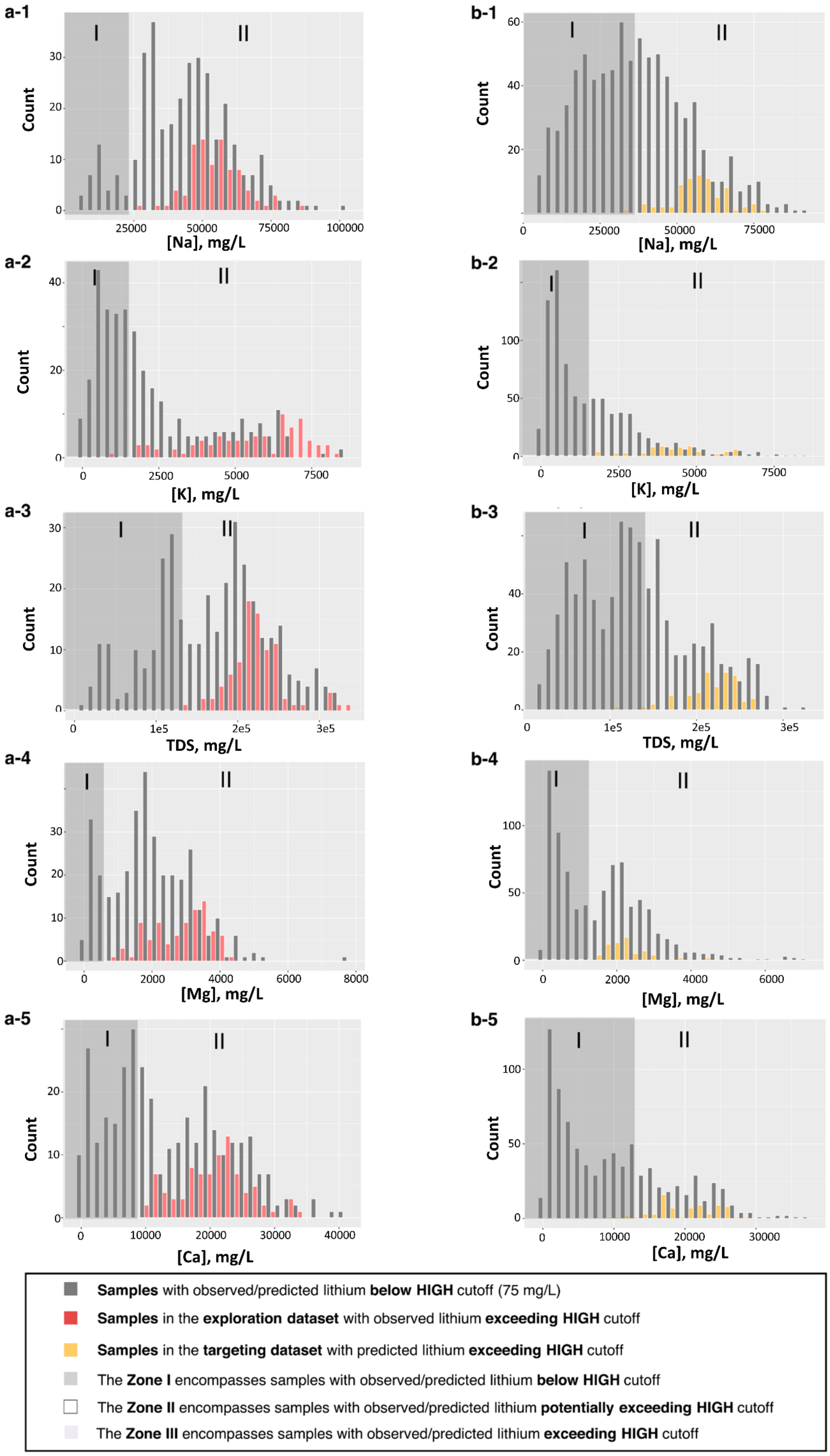
Seciont 6. Additional model results



Supplementary Figure S5 Cumulative mean accuracy and precision. (a) and (b) for passing the low cutoff case; (c) and (d) for passing the mid cut-off case; (e) and (f) for passing the high cut-off case.



Supplementary Figure S6 Comparison of selected geochemical distributions for LOW cutoff (35 mg/L). a-1 to a-5, Distributions of observed results in the exploration dataset (with [Li] measurements). b-1 to b-5, Distributions of predicted results in the targeting dataset (without [Li] measurements).



Supplementary Figure S7 Comparison of selected geochemical distributions for HIGH cutoff (75 mg/L). a-1 to a-5, Distributions of observed results in the exploration dataset (with [Li] measurements). b-1 to b-5, Distributions of predicted results in the targeting dataset (without [Li] measurements).

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