POTENTIAL RESOURCES OF LITHIUM FROM OILFIELD BRINES IN NEUQUEN BASIN, ARGENTINA

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RESUMEN

Acciones inmediatas contra el impacto del cambio climático están acelerando la incorporación de fuentes de energía renovable a la matriz energética mundial. En este sentido, la explotación e industrialización del litio ha tenido una demanda creciente en los últimos 5 años, dado que su eficiencia energética se ha vuelto determinante en el ciclo de vida de las baterías de vehículos eléctricos, la fabricación de cualquier tipo de tecnología inalámbrica (Smartphones, Tablets, etc.), y en el almacenamiento de energía a gran escala. En la actualidad, la capacidad de producción de litio está limitada geográficamente, siendo necesario incorporar nuevas fuentes para garantizar la creciente demanda mundial.

El suministro mundial de litio proviene principalmente de pegmatitas (26%) y depósitos de salmuera (58%). En Argentina, su producción está restringida a los depósitos de salmuera de la región de la Puna, ubicados en cuencas endorreicas asociadas a salares, donde el método extractivo más común es la evaporación solar en piletas. Los costos de procesamiento son bajos y eficientes gracias a las condiciones climáticas áridas.

Se estima que alrededor del 3% de los recursos de litio mundial están contenidos en salmueras de campos petrolíferos, algunos ya documentados en diferentes lugares de los Estados Unidos (Dakota del Norte, Wyoming, Oklahoma, Arkansas y el Este de Texas). La Formación Smackover, en la cuenca del Golfo de los Estados Unidos, alberga un millón de toneladas métricas de recursos de litio. Esto permite que el Litio se recupere rápidamente mediante la aplicación de tecnologías avanzadas de separación primaria, tales como membranas, tamices moleculares y electrólisis.

Este trabajo expone posibles fuentes de litio para la cuenca Neuquina de acuerdo al denominado Lithium System, incluyendo distintos plays de potencial evaluación. Dada la ausencia de datos públicos que confirmen la presencia de litio en aguas de producción de yacimientos en operación, se proponen los mejores sitios para su caracterización. Estos análisis podrían permitir la incorporación de una nueva fuente de litio en Argentina, cuya extracción no requeriría la perforación de nuevos pozos, pudiendo las Compañías beneficiarse de los tratamientos de las aguas de producción. Además, la recuperación de litio reduciría costos operativos (OPEX) y aumentaría ingresos, brindando un nuevo panorama en la transición hacia una economía circular.

INTRODUCTION

Lithium is the third chemical element of the periodic table. It is included in the Group 1 and represented with the Li symbol (Fig. 1). It is a soft, silvery-white alkali metal that under standard conditions becomes the lightest solid element. Like all alkali metals, lithium is highly reactive and flammable (BGS 2016).

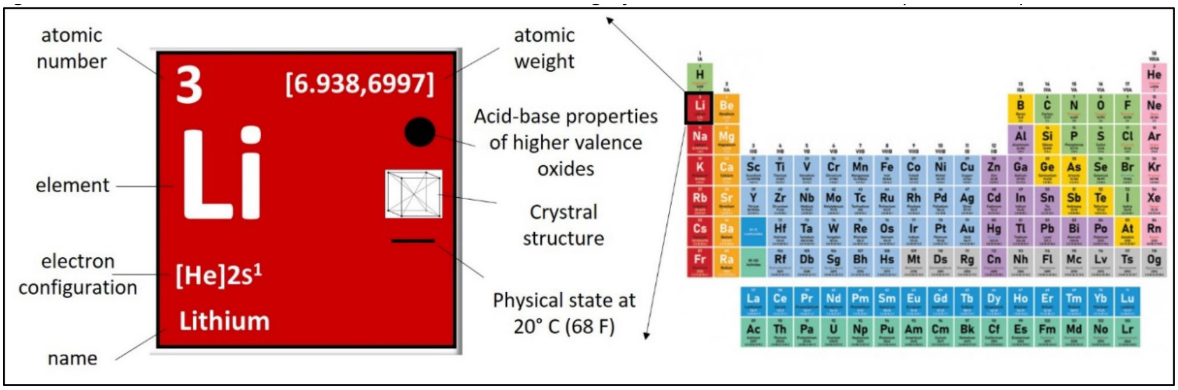

Figure 1. Lithium representation and properties as shown in the in Mendeleyev´s periodic table.

Lithium is enriched in the continental crust over the mantle. It does not occur free in nature since in igneous rocks appears as an element forming minerals such as spodumene, lepidolite, petalite and amblygonite (in pegmatites and some alkali granitoids), and also under solution in springs and seawaters (BGS 2016). Weathering of rocks containing lithium-rich minerals releases lithium to the environment. As a result, lithium is found in varying amounts in all soils. Carbonates and calcareous shales precipitated from evaporated seawater or lagoons can have high lithium concentrations (Aral and Vecchio – Sadus 2011). According to Goldschmidt (1937), lithium abundance on Earth´s crust was 65 ppm. However, an actual accepted average concerning igneous and sedimentary rocks is around 17 to 20 ppm (BGS 2016).

Concentrations of lithium are found mostly in closed basin brines and ore deposits (pegmatites), being less descripted in clays, geothermal brines and oilfield brines (Fig. 2A). Two-thirds of the world's lithium reserves are found in Argentina, Chile and Bolivia in what is known as the "Lithium Triangle", with 68% of the world's lithium resources (Fig. 2B). Even though, the actual leading productions comes from ore deposits, with Australia and Chile with almost the 80% of the market (Fig. 2C). This work focuses on reviewing the oilfield brines lithium potential resources linked with the conventional and unconventional plays in the Neuquén basin, a probably long-overlooked lithium-rich resource.

In hydrocarbon production fields, there is always a large volume of ion-enriched produced water (brines) which comes out with abnormal amounts of many prized commodities such as: magnesium, bromine, potassium and lithium, among others (Ecclestone 2020). This type of

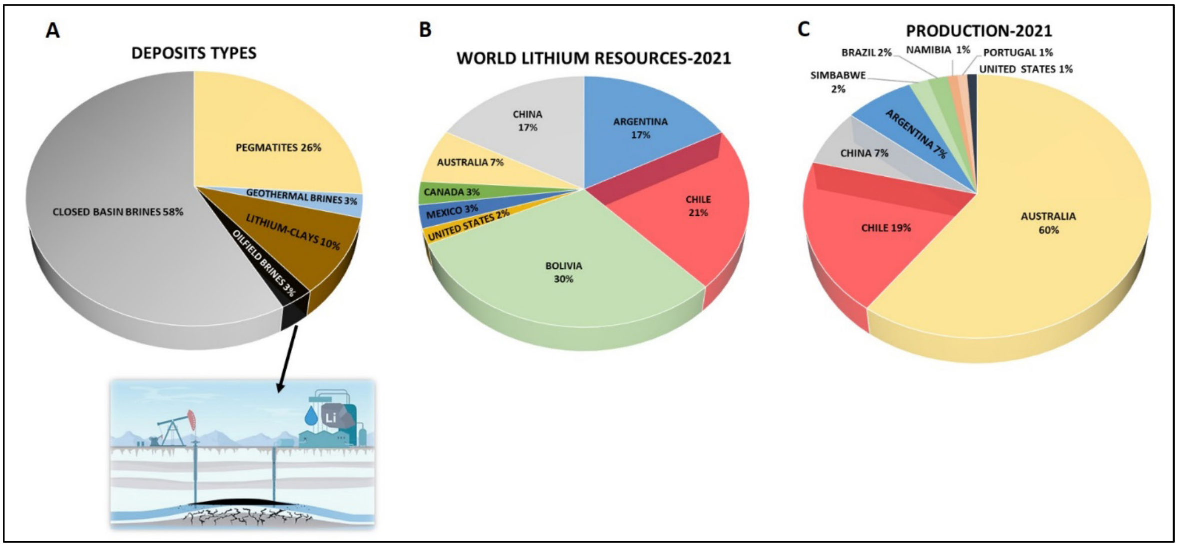

Figure 2. A) Lithium Deposit Types, B) Lithium Global Resources and C) 2021 Production (USGS Mineral Commodity Summaries, January 2022).

deposit, never considered before as a lithium source in the Neuquén basin, becomes of special interest considering the existing of surface infrastructure (industrial facilities, technical and operational management knowledge, roads and piping, already drilled wells, etc.) and the deep geological knowledge of the basin history which can rapidly include the lithium genesis and transport conceptualization within the different stratigraphic formations. The presence of lithium from oilfield brines has been demonstrated in recent years, mainly in those countries where other types of deposits absent:

- Arkansas, USA. Standard Lithium Company. Smackover Formation. Bromine produced from Smackover brine supplied over 40% of the world's bromine from 1986 to 1990 (Arkansas Geological Survey 2015). Standard Lithium announced the positive results of a preliminary economic assessment (PEA) for the Arkansas Smackover Lithium project in June, 2019. https://www.standardlithium.com/projects/arkansas-smackover
- Alberta, Canada. E3 Metals Corporation. Leduc Formation. The Leduc was a prolific light oil producer formation, and is mostly depleted now. It is still used for disposal wells and recently as a source of oilfield brines used in lithium extraction. http://www.e3metalscorp.com/our-assets/technical-reports/
- Formosa, Argentina. Palmar Largo Field. Cretaceous Basin, existence of lithium from volcanic reservoirs oilfield brines (100 ppm Li) was confirmed. Perfectibility stage (Vergani unpublished data, 2019).

Prices and demand for lithium production are expected to keep rising considering the growth driven by electric vehicles batteries, and the continuously evolving technology optimizations. All this coming evolution supports the idea of accounting for lower concentration lithium production sources, like those from oilfield brines.

Geological research allows a better understanding of the lithium genesis and contribution to the endorreic basins in the in the Puna region, Argentina (Catalano *et al.* 1964; Viramonte *et al.* 1984; Igarzábal and Alonso 1990, Alonso *et al.* 1991, 2020; Coira *et al.* 1993; López Steinmetz *et al.* 2018; Meixner *et al.* 2021). All actual lithium brine deposits share distinctive characteristics that enable the containment and concentration of lithium: closed basin, arid climate, suitable lithium source rocks, geothermal activity, time to concentrate a brine and one or more adequate aquifers. Figure 3 depicts the genesis of lithium brines in the Puna region according to De la Hoz (2021). This conceptual deposit model was considered an analogous for the Neuquén basin in order to identify potential sources of lithium along the stratigraphic column.

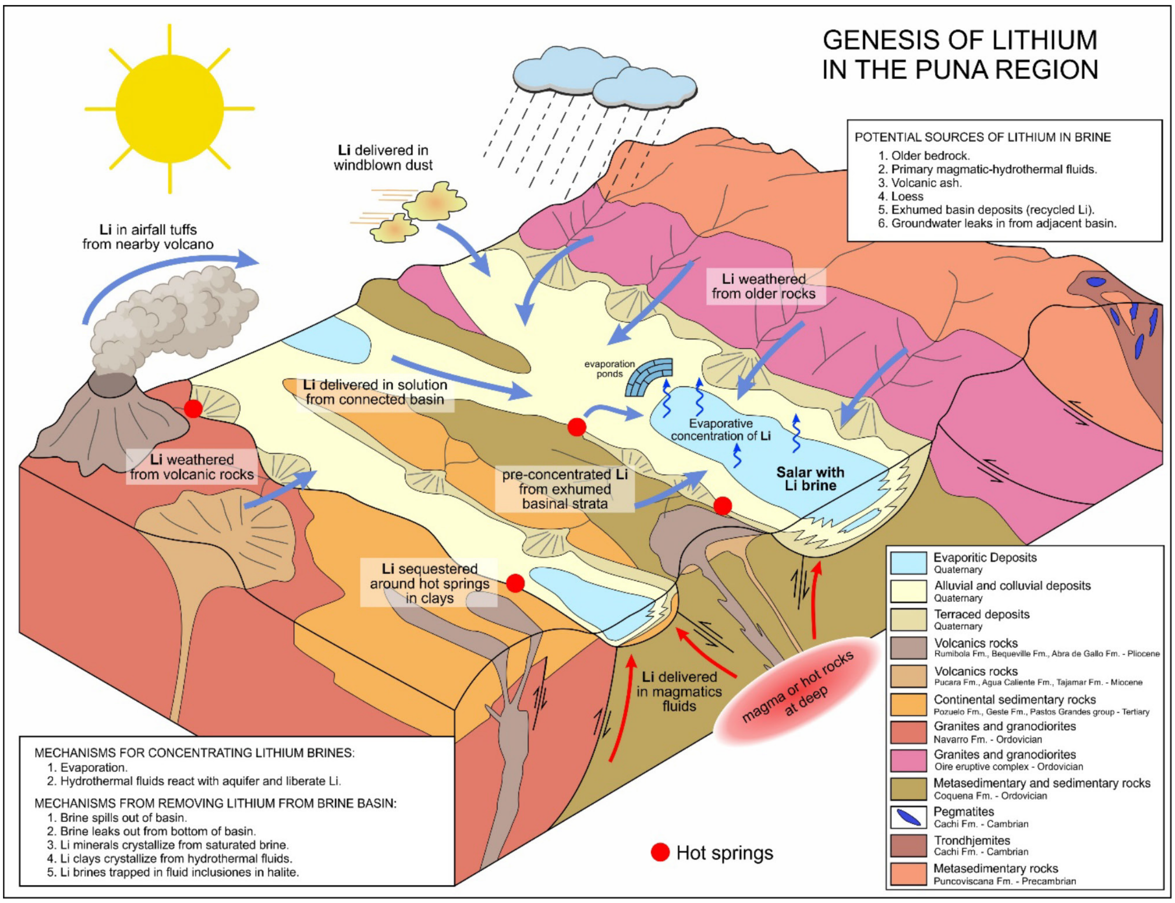

Figure 3. Genesis of Lithium in the Puna region (De la Hoz 2021).

NEUQUEN BASIN

The Neuquén basin is located on the eastern side of the Andes in Argentina and central Chile. It contains a near-continuous Late Triassic-Early Cenozoic succession deposited on the eastern side of the evolving Andean mountain chain (Fig. 4). The basin covers an area of approximately

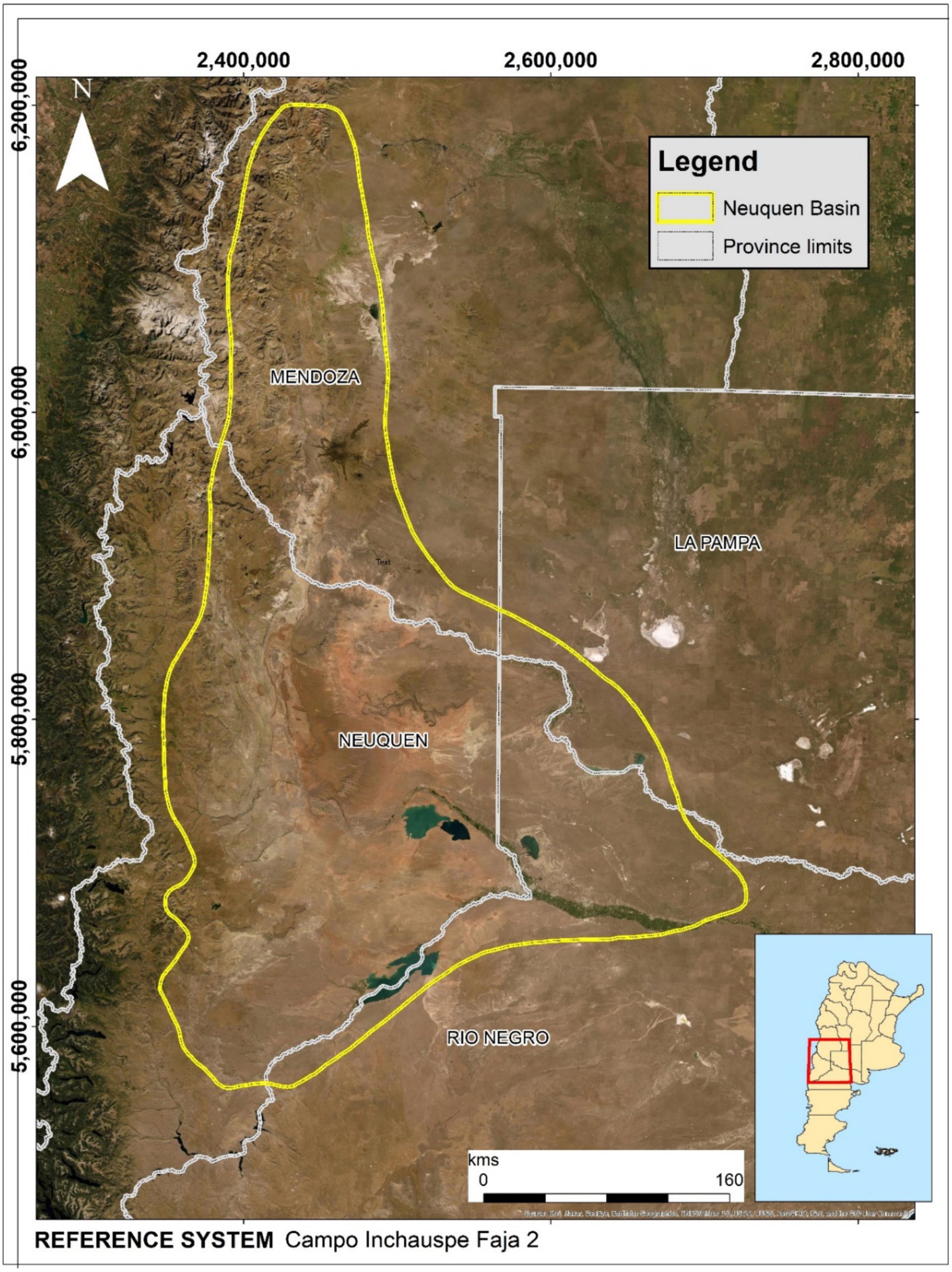

Figure 4. Neuquén basin sketch map. Limits taken from IHSMarkit database.

120.000 km^2 being originated in the Jurassic, later developed through alternating continental and marine conditions into the Tertiary (Veiga *et al.* 2020). Infilled sediments exceed 7000 m of marine and continental sedimentary successions (epiclastics, carbonates, evaporites and pyroclastics) from

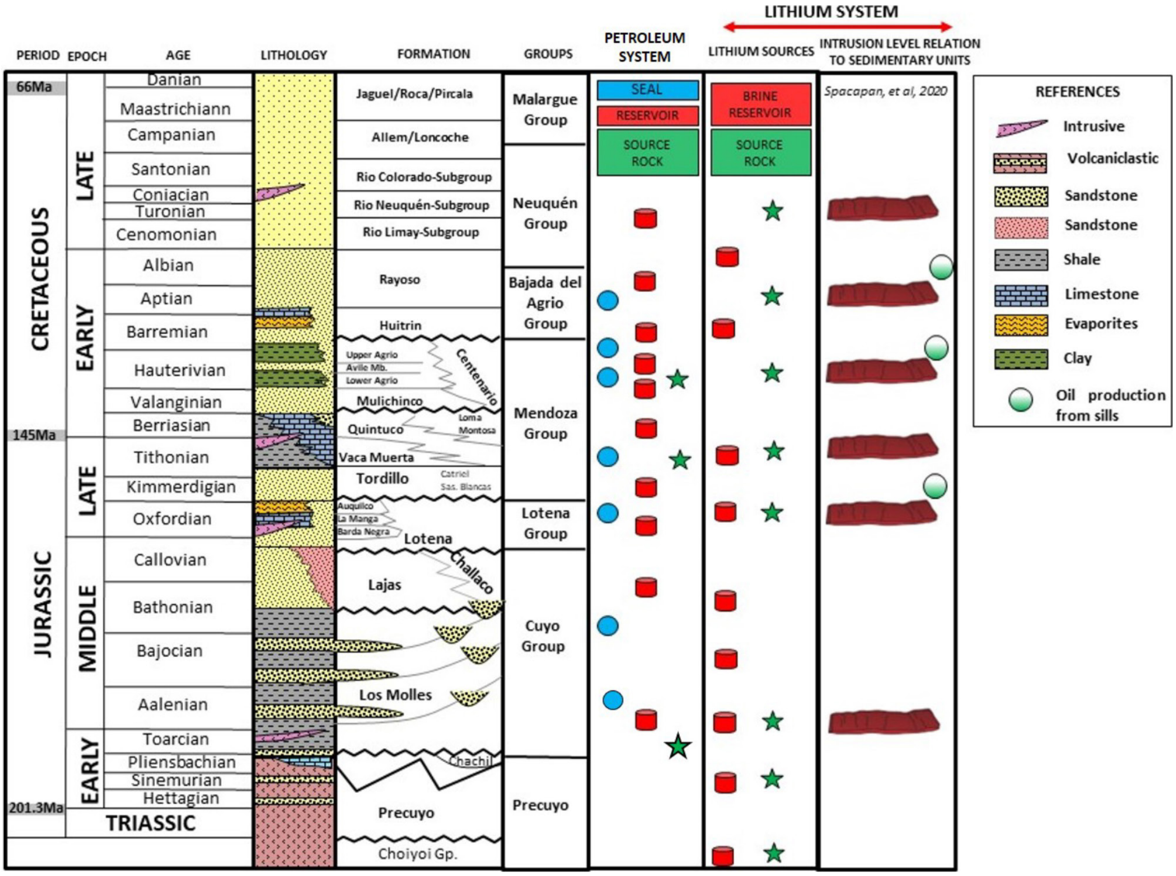

Figure 5. Neuquén basin chronostratigraphic column modified from Gutierrez *et al*. (2020). Note the lithium sources and Miocene intrusion levels (after Spacapan *et al*. 2020) which features the Lithium System.

Triassic to Cenozoic age, interrupted by several unconformities explained by different episodes of inversion and intermittent subsidence (Fig. 5). The structural inversion (Huincul High) was an important component on the hydrocarbon trapping (Aguada Baguales, Puesto Touquet, La Chilca, Cutral Co and Cupén Mahuida hydrocarbon fields). The Neuquén basin sedimentation has been historically divided in 7 different Groups: Precuyo, Cuyo, Lotena, Mendoza, Rayoso, Neuquén and Malargüe (Veiga *et al*. 2005). Two main eruptive cycles were described by Combina and Nullo (2011) in the northern part of the Neuquén basin: The Late Oligocene – Middle Miocene Molle Eruptive Cycle (MEC), and the Late Miocene – Pliocene Huincán Eruptive Cycle (HEC). These eruptive cycles resulted in thick lava flow sequences, as well as numerous andesitic to basaltic sills dominantly emplaced in the source rock formations of the Mendoza Group (Fig. 5).

The new Lithium System proposed in this contribution is represented in figure 5 considering the local sources of lithium and the potential lithium brine reservoirs. Green stars depict potential sources in basement and intrusive rocks like granites, ignimbrites, tuffs, and evaporites, similar as those observed in the Puna region. Lithium could have been leached from their source rocks and carried away by continental drainage and geothermal/hydrothermal fluids through major fault systems and associated fractures, into adjacent sediments feeding the lithium brine reservoirs. The extensive igneous intrusions embedded in sedimentary strata (Spacapan *et al.* 2020) provides an additional source of lithium for the system.

Geodynamic evolution

The complex history of the basin was controlled by changes in the Gondwana western margin tectonics during the Mesozoic. The evolution of the Neuquén basin is linked to the development of the Central Andes (sensu Ramos, 1999) and the geometry of the subducting proto-Pacific slab. Volcanic activity along a North-South trend magmatic arc, combined with long-term tectonics, and local uplifts and depressions, are essential to understand the basin evolution and identify potential resources of lithium along the stratigraphic column.

Howell *et al.* (2005) defined three stages for the Neuquén basin evolution (Fig. 6):

1. Initial Rift Stage (Late Triassic – Early Jurassic): Prior to the subduction onset, the Gondwana western margin was shaped by large transcurrent fault systems. This led to extensional tectonics and the evolution of narrow and isolated clastic and volcaniclastic filled depocentres within the basin (Vergani *et al.* 1995).
2. Subduction – Related Thermal Sag Stage (Early Jurassic – Early Cretaceous): Development of a steep and active subduction zone, associated with the evolution of a magmatic arc along the western margin of Gondwana, and back arc subsidence within the Neuquina basin. These movements promoted a retraction of the marine shoreface, flooding the whole basin with successive sets of transgressive and regressive cycles. Their magnitudes became dependant on the back arc subsidence rate, the localized uplift, and the eustatic oscillations, widely descripted in Cuyo, Lotena and Mendoza Groups (Veiga *et al.* 2005). This basin Post – Rift stage, which included thermal subsidence and back arc extension, can account in some locations with more than 4000 m of sediments (Vergani *et al.* 1995).
3. Foreland Stage (Late Cretaceous – Cenozoic): Transition to a low angle subduction zone resulted in the development of a compressional tectonic regime that caused inversion of previous extensional structures (Vergani *et al.* 1995) and deposition of more than 2000 m of synorogenic deposits (Rayoso and Neuquén Groups; Legarreta and Uliana 1999). At this stage, the Neuquén basin region became a foreland basin, with a continuous migration of the depocentres to the east (Legarreta and Uliana 1991), changing the basin size and shape. Global sea level hights in the late Cretaceous produced the first Atlantic transgression, recognized in almost all Argentina from their shallow marine facies and fosils.

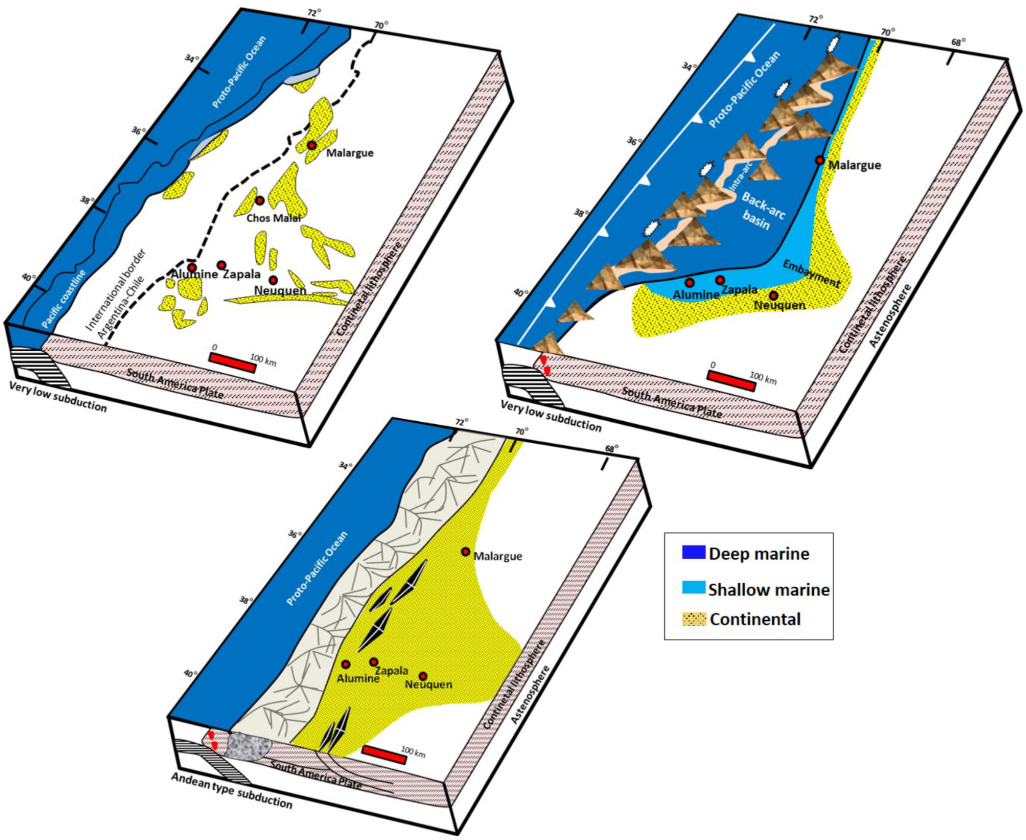

Figure 6. Geodynamic evolution of the Neuquén basin. Modified from Howell *et al.* (2005).

Several thin- and thick-skinned fold and thrust belts developed as a result of the foreland basin stage (Ramos, 1999) constitutes the major control of the present-day geomorphology of Neuquén basin.

Potential resources of lithium from oilfield brines

Ion-leaching by weathering and hydro or geo-thermal systems are considered the two main lithium extraction mechanisms from the source rocks (Meixner *et al.* 2022). This section focuses in the evaluation of potential lithium resources in the Neuquén basin by tracing the lithium pathway from source to sink (USGS 2016). Our proposed Lithium System (Fig. 7), is in order to the required geological elements and processes that are essential for getting high lithium concentration in brines. Those features are:

Elements: Lithium source rocks and potential storage in reservoirs brines.

Processes: Chemical weathering, lithium leaching, and migration pathways.

In geology, a petroleum play, is a group of hydrocarbon fields or prospects in the same region that are controlled by the same set of geological circumstances. The term is widely used in the realm of exploitation of hydrocarbon-based resources (Magoon and Dow 1994). According with

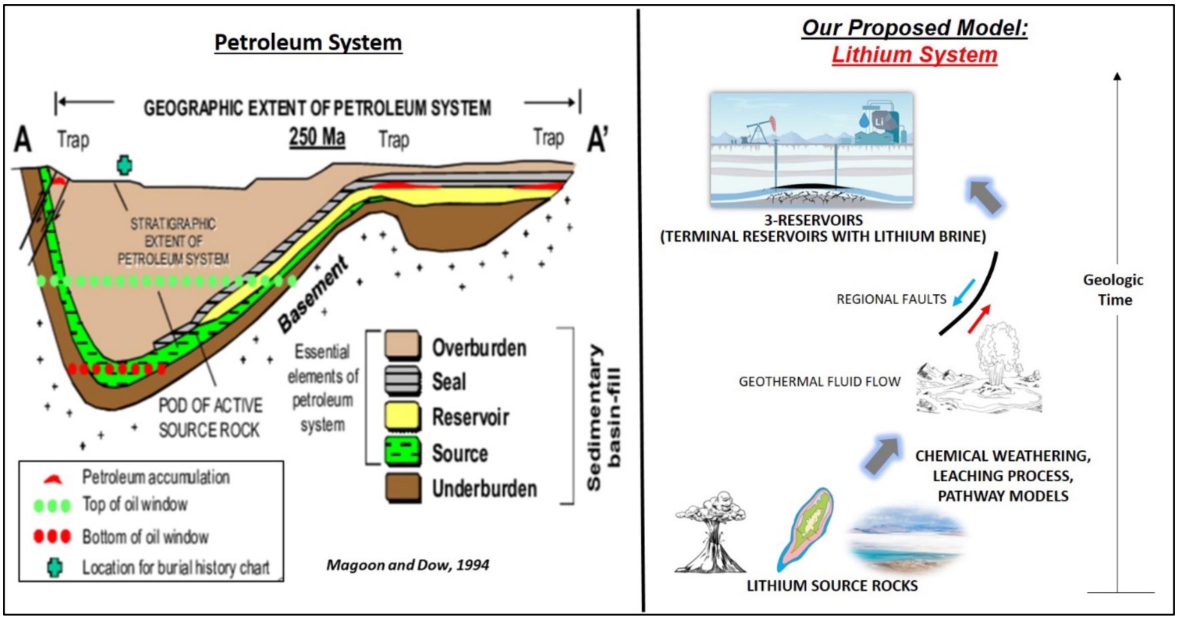

Figure 7. Our proposed model for the Neuquén basin - Lithium System. Since source, rocks are variable, spilling, migration and accumulation processes are diverse.

our Lithium System model and based on geological studies from underproduction and non-already exploited salt flats as analogous, we introduce this concept to the oilfield brines recognized four different lithium plays in the Neuquén basin (Fig. 8):

1. Basement Play: Choiyoi Group (granites, riolitic ignimbrites and tuffs).
2. Volcanoclastic + Intrusives Play: Precuyo Cicle (acid tuffs and sills in sedimentary units)
3. Evaporites Play: Auquilco and Huitrin formations (Marine Evaporites)
4. Unconventional Play: Vaca Muerta and Molles formations (Iong-leaching by high intensity pumping, hydraulic fractures on shales/tights)

A review of each of them will be done, considering some productive oil and gas fields, where analysis could be carried out to get an empirical test of these plays. Additionally, geothermal activity considerations will be exposed as to get a full idea of the best prospective zones within the Neuquén basin.

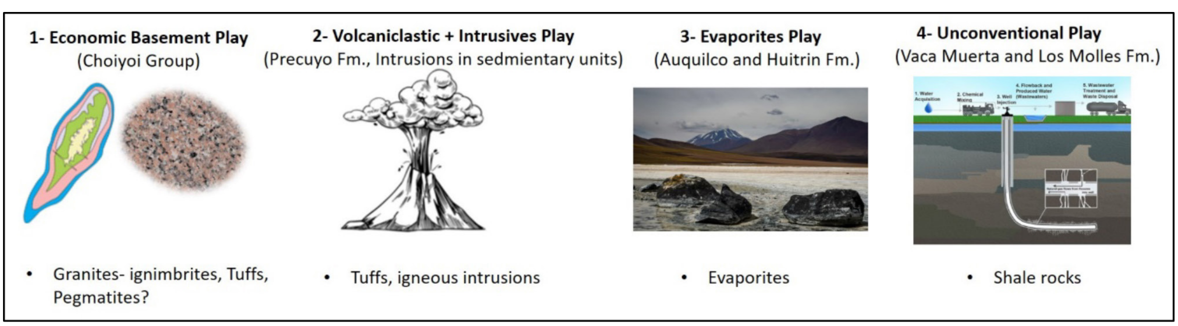

Figure 8. Lithium Plays proposed in this work for the Neuquén basin.

Basement Play

Weathering, erosion and leaching of volcanic (silicic ignimbrites and tuffs) and calc-alkaline plutonic rocks (Llambías *et al.* 2008) could be potential sources of lithium in oilfield brines. The Neuquén basin basement volcanism is widely represented with the Choiyoi Magmatic Province, spread out on central and western zones of Argentina during the Permo-Triassic (Fig. 9). Except for its long duration (~30Ma), it could be considered a LIP (Large Igneous Province) (Llambías 2008). These deposits provide an extensive dacitic to riolitic plateau that act as the basement of the Neuquén basin (Llambías and Aragón 2011) and could be a potential source of lithium for the oilfield brines. Two hydrocarbon fields are proposed to test the Basement Play based on their main lithology (Barrionuevo 2015; Velo *et al.* 2014), (Figs. 9 and 10):

- Rhyolitic ignimbrites from 25 de Mayo field. (Operator: Medanito)
- Granitoids from fractured Huechulafquen Formation in Octógono Fiscal field (Operator: YPF).

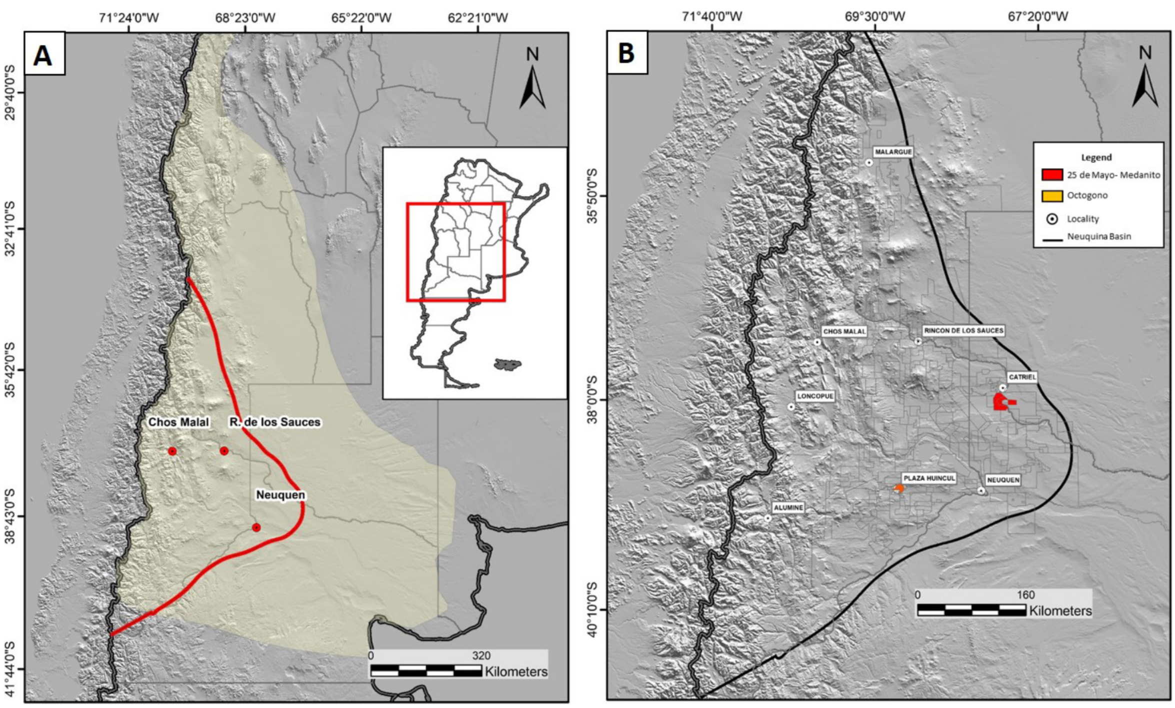

Figure 9. A) Maximum extension of the Choiyoi Group (Adapted from Llambías and Aragón 2011). B) Choiyoi hydrocarbon fields.

Volcanoclastic + Intrusive Play

Widespread volcanism has characterized the Neuquén Basin during the syn-rift evolution. As a result of the extensional tectonics, a series of rift troughs were distributed over the surface.

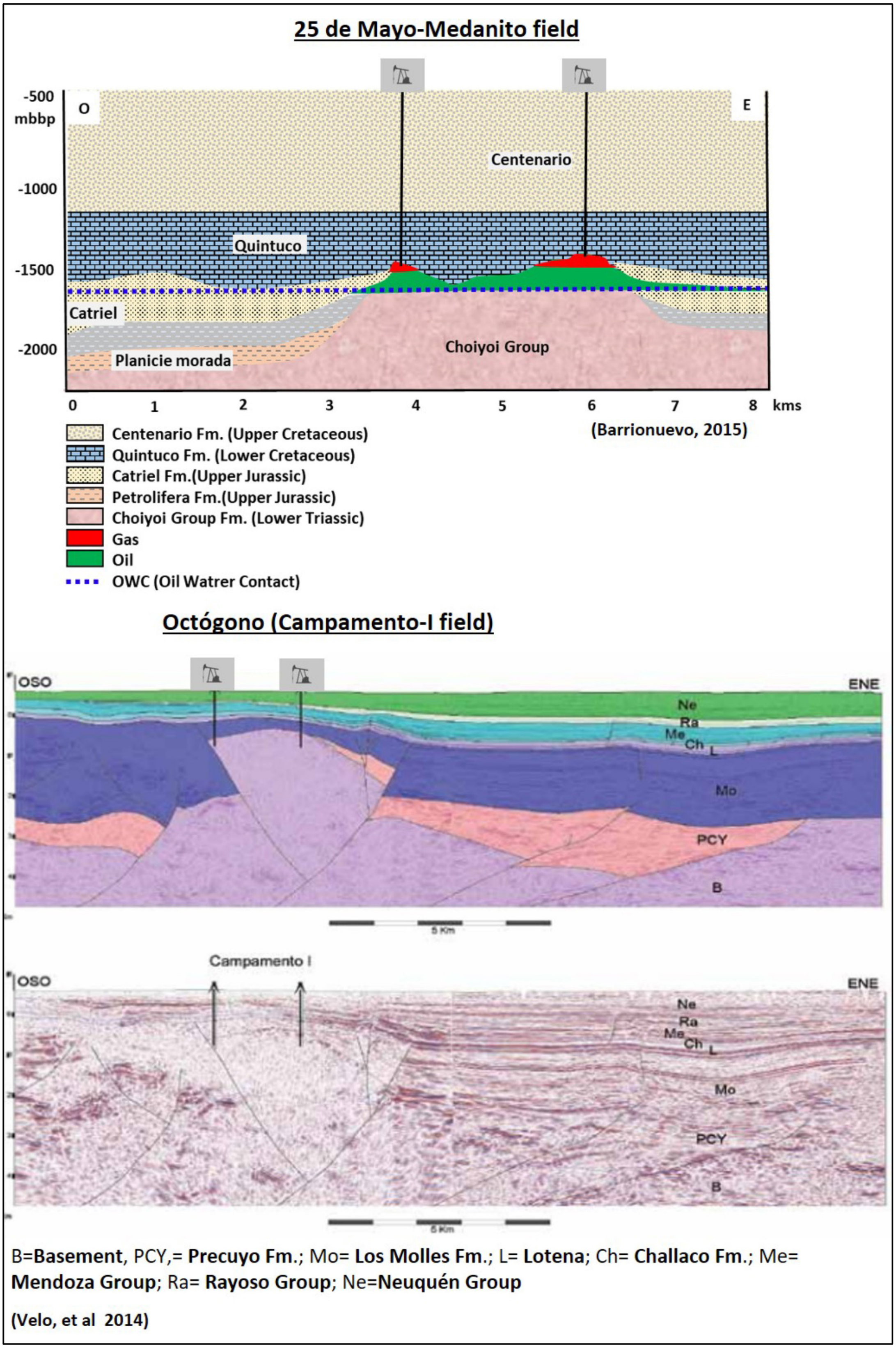

Figure 10. Sketch section from 25 de Mayo-Medanito and Octógono Fiscal fields.

Volcanic feeding systems promoted multiple-injection magmatic chambers and, in shallower crust, they became linked to major faults that delimited the depocenters. The rifting phase was accompanied by shallow intrusives, lava flows and pyroclastic deposits (D'Elía *et al.* 2020). Lithium brines enrichment is supposed to be originated from weathering and leaching of volcanic rocks, especially in the Huincul High region of the Neuquén basin, where several oil and gas fields could test the play (Fig. 11).

Regarding the instrusives, emplacement of Miocene sills has been descripted by Spacapan *et al.* (2020) along the stratigraphic column of Chihuido de la Sierra Negra and San Roque oilfield blocks (Fig. 13). The thermal effects of these intrusions on surrounding sedimentary strata are of huge interest considering hydrothermal alterations in the potential lithium source rocks, being one of the main factors for lithium concentration in oilfield brines (Fig. 5).

Upwelling magmas preferably intrudes low competence rocks and fractures. The tabular morphology of these igneous intrusions, mostly concordant, and the hydrothermal effects produced are easily distinguished in seismic and well logs due to their distinctive behavior (Schiuma and Llambías 2014) (Fig. 13).

Intrusives lithium play could be tested in four oil and gas fields: Cajon de los Caballos, Valle de Rio Grande, Chihuido de la Sierra Negra, and San Roque.

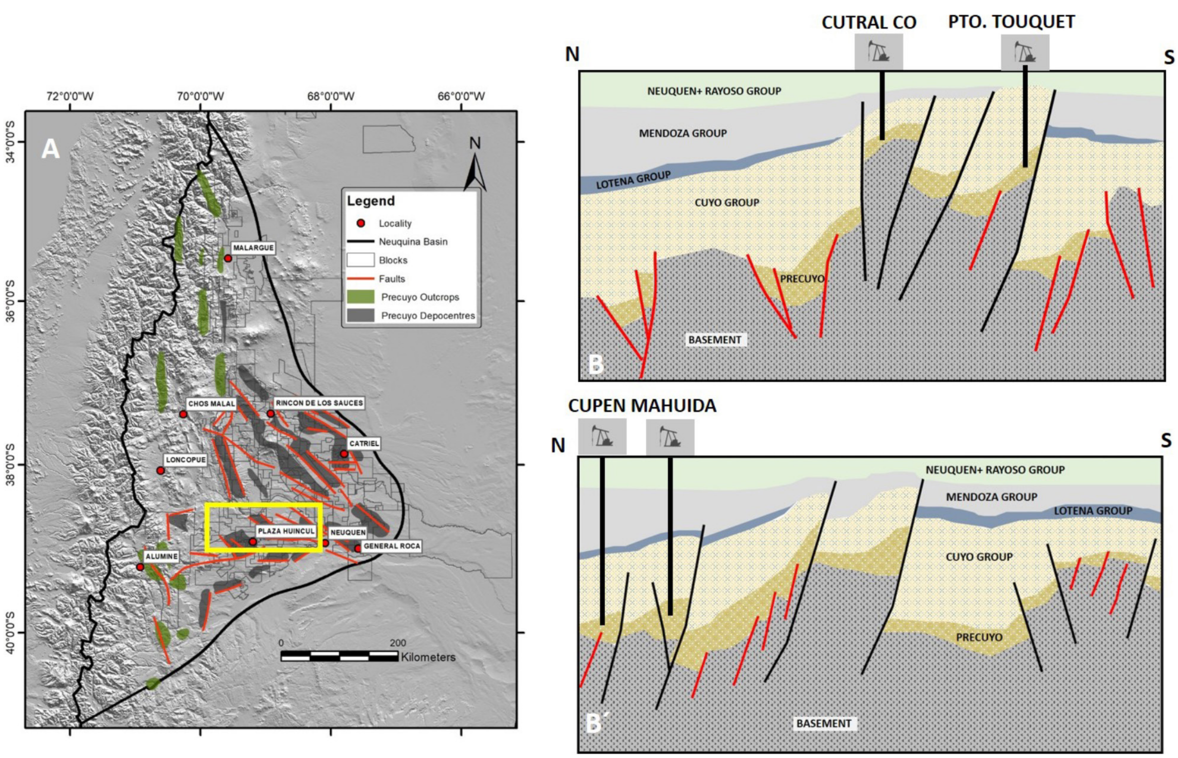

Figure 11. A) Distribution of the extensional depocenters in the surface and subsurface of the Neuquén basin (Adapted from D'Elía *et al.* 2020). B and B') Sketch section from the main Precuyo Formation productive fields (Adapted from Carbone *et al.* 2011).

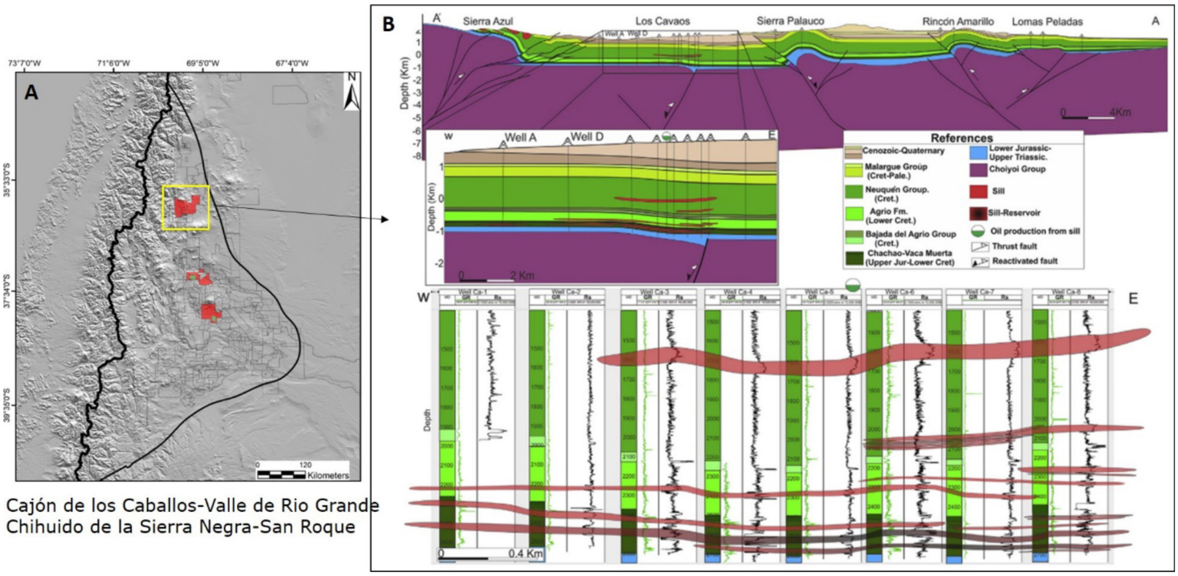

Figure 12. A) Main fractured intrusive Neuquina basin oil and gas fields, and B) sills intruding the basin sequence in Chihuido de la Sierra Negra and San Roque fields according to Spacapan *et al.* (2020).

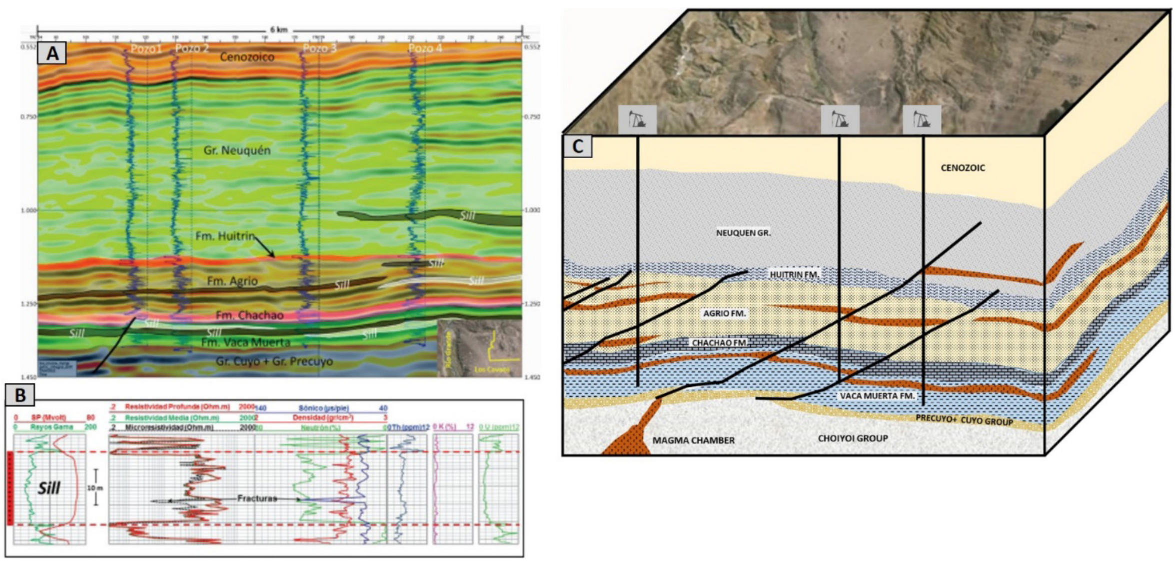

Figure 13. Sills identified on A) seismic and B) electric logs. C) Sketch showing the relationship between sills and faults, main migration pathways of the igneous intrusions (Adapted from Schiuma *et al.* 2014).

Evaporites Play

Local basins with restricted circulation may develop hypersaline environments. In warm dry regions, these waters will become concentrated enough to precipitate evaporite minerals, including gypsum, halite and different potassium and magnesium salts. The high solubility of lithium prevents its precipitation before most of the sodium, potassium, and magnesium salts have been crystalized. Lithium precipitation is so rare that a natural in situ lithium salt has never been observed (Vine 1975).

In the Neuquén basin there are no producing oilfields from this type of rocks. Nevertheless, they play a key role as a potential source rock of lithium, lithostatically squeezed during the geologic time (Fig. 14).

A combined effect of a relative base level fall and a pervasive volcanic activity along the magmatic-arc explain the isolation of the Neuquén depocenters from the Pacific Ocean. This volcanic arc could have provided the sources of lithium to the evaporites depocenters. With the continuous deposition and burial of the formations, the originally permeable salts would

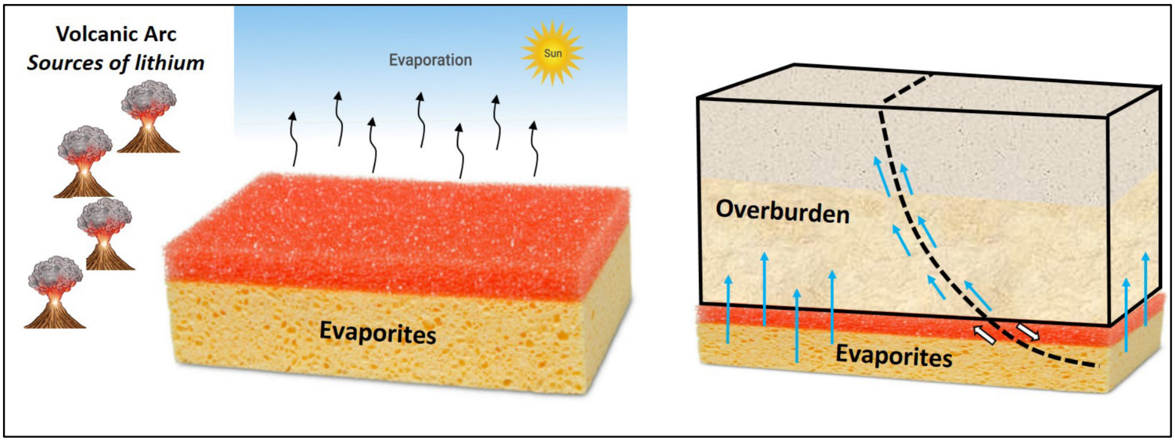

Figure 14. Brine migration pathway model for the Evaporites Play.

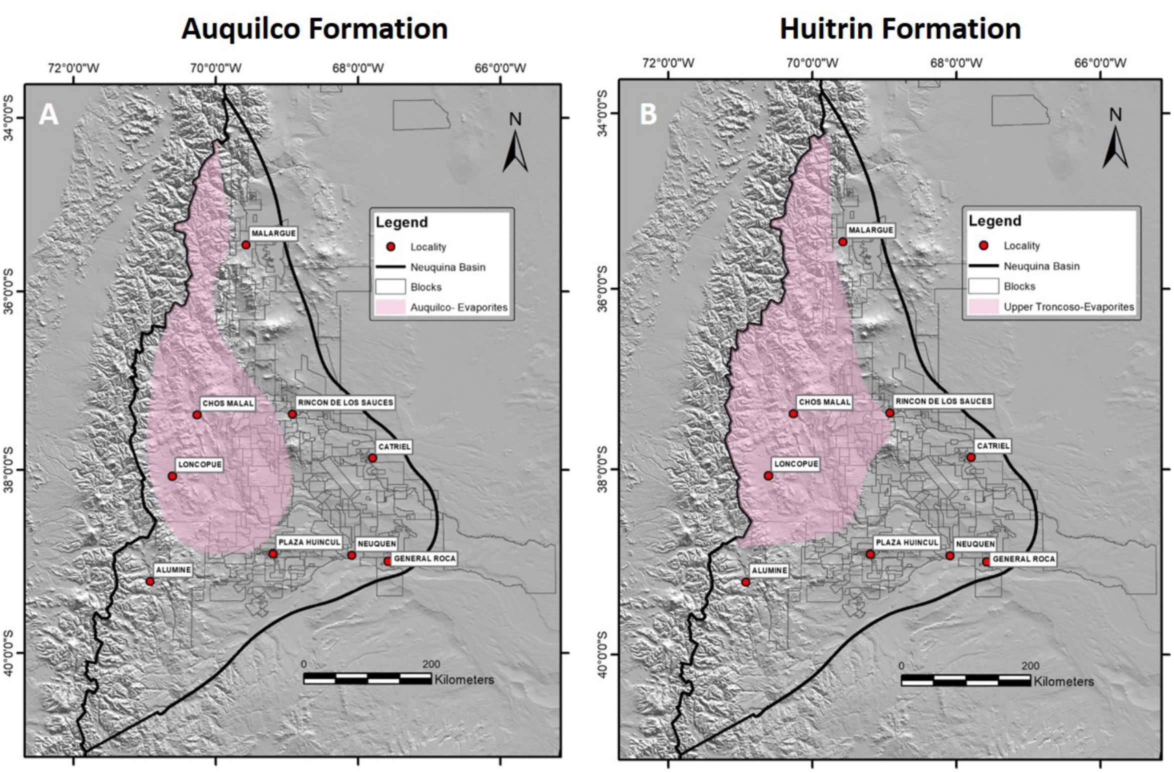

Figure 15. Maximum extension of A) Auquilco Formation and B) Huitrín Formation (Adapted from Legarreta 2002).

recrystallize into a solid dry mass causing the brine to be expelled away. Most of the lithium bearing fluids could have been carried away from its source rocks to their neighboring reservoir rocks along the main fault systems. This pathway model is similar as those proposed for the origin of lithium coming from the underlying evaporates units (Loaum Salts) in the Smackover Formation (NI-43–101 Rep. 2019).

There are two widely dispersed sedimentary units to test the play in the Neuquén basin, Auquilco and Huitrín Formations (Fig. 15).

Unconventional Play

The most common used method for exploration and exploitation of unconventional resources is hydraulic fracturing of reservoirs through horizontal wells. Hydraulic fracturing increases reservoir permeability by creating a complex fracture network that provides an artificial carrier for the hydrocarbons. This process is known in the oil and gas industry as the wellbore completion phase. During those operations, large volumes of pressurized and high flowrate water is pumped in as to beat the formation's minimum stress, and to act as a carrier for the also pumped sand inside the created fracture network (Zoback and Kohli 2019). Once the completion is over, the well is controlled open for testing and production. At this point, large volumes of injected and formation water return out to surface, as the so-called flowback brines.

In Neuquén basin, the Early Tithonian–Early Valanginian Vaca Muerta Formation is composed of organic-rich calcareous mudstones representing the main self-sourced unconventional play (Kietzmann *et al.* 2020, Minisini *et al.* 2020). Many authors described the presence of volcaniclastic deposits along the Vaca Muerta interval. The volcaniclastic deposits are composed of lithic fragments, plagioclase, pumiceous particles with common calcite replacement or alteration to clay minerals (González *et al.* 2016, and references therein). Figure 16 shows the huge ash bed density along Vaca Muerta Formation. These volcanoclastic intervals could be acting as potential sources of lithium, that could be obtained from the oil and gas wells flowback productions, as those described in the Appalachian basin Marcellus Shale by Fajfer *et al.* (2021). Enrichment genesis is thought to be the high pressure and flowrate used for the wells completions that leach the formation adsorbed ions (including lithium) and increase their concentration in the flowback brines.

Lithium lab tests have already been carried out in some manifold sampled flowback brines from unconventional assets around the embayment sector of the Neuquén basin. Obtained results showed lithium concentrations between 150 and 250 ppm that provide proved evidence about the unconventional play Lithium System.

Geothermal Activity

Several epithermal deposits have been worldwide recognized depending on their mineral assemblages, sulfur oxidation state and their former geotectonic environment (Hedenquist *et al.* 2000). Along the Andean fold and trust belt, geothermal fluids have been mentioned as the reason for the significant economic metallic deposits (Mas *et al.* 2011, Zanettini 2018). This process is consequence of naturally heated deep waters that flow throughout the sedimentary units and faults, forming near-surface (epithermal) deposits and geothermal springs, like those in the Puna region (Peralta Arnold *et al.* 2017).

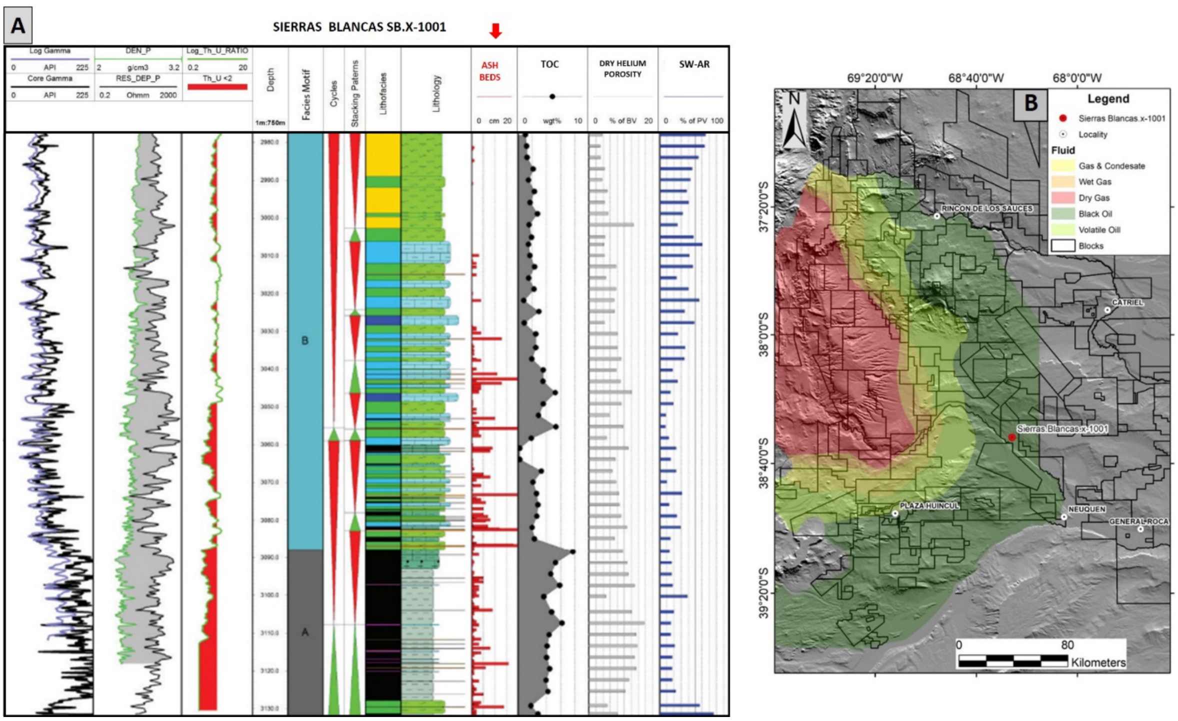

Figure 16. A) Sierras Blancas.x-1001 well logplot. Note the large quantity of identified ash beds. B) Fluid map for the Vaca Muerta Formation and Sierras Blancas x-1001 position (Adapted from Minisini *et al.* 2020).

Around the Neuquén basin, volcanic activity has been described from its initial rift stage (Howell *et al.* 2005) (Fig. 6). Actual heat flow is confirmed from the active volcanoes (Fig. 17a), some of them active since the Eocene (Galland *et al.* 2007), and from the Sigismondi and Ramos (2011) basin heat map (Fig. 17b), which preserves a close relation with the geothermal anomalies fields.

Considering all these conditions, fluids over 70 °C flowing around volcanic hotspots or basin geothermal gradient anomalies (Fig. 17b) could act as solvent solutions dissolving formations' minerals and leaching elements into the hot water. The exploration and study of oilfield brines from hydrocarbons fields that are close to geothermal systems, can be a reasonable starting point for the prospective analysis of the oilfield brines Lithium System.

CONCLUSIONS

The Lithium System model proposed for the Neuquén basin is consistent with lithium oilfield brines deposition criteria mentioned by different authors in the Puna region, on Northwest Argentina.

Geologic and climate factors may have converged to trap lithium in brines in the Puna from the Pliocene to Present. If we consider the evolution time of the Neuquén basin, millions of years of volcanic activity and changing depositional environments could have produced different elements and process interactions that would have concentrated lithium within the different oilfield brines lithium plays.

Potential contribution of lithium to the Neuquén basin could be related to volcanic rocks weathering and erosion processes, highly amplified by geothermal activity. As in Smackover Formation (Arkansas, USA), source to sink pathway models becomes necessary for understanding the Lithium System and the recognition of the different oilfield brines Plays.

Several oil and gas fields and geological formations were proposed for analyzing the lithium concentrations in oilfield brines of Neuquén basin. Some manifold sampled flowback brines from

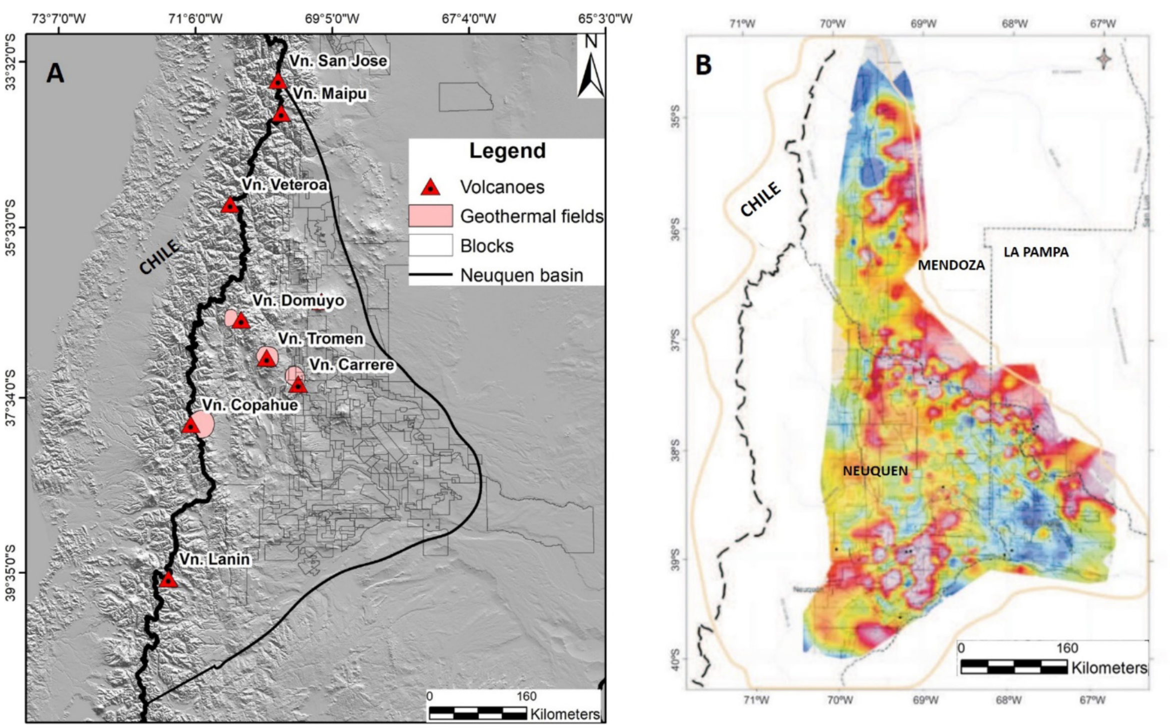

Figure 17. Volcanic activity and geothermal fields within the Neuquén basin. B) Geothermal gradient map (Sigismondi and Ramos 2011), warm and cold colors represent positive negative anomalies respectiviely (range of variation: 17 °C/km to 133 °C/km).

unconventional assets obtained optimistic results proving evidence about the unconventional lithium play development. Moreover, thousands of drilled wells have already intersected our proposed lithium plays. Regional seismic datasets, production records and a huge infrastructure resource are available, reducing prospective, and potentially productive, costs and risks. At present, public data regarding the lithium concentration in the Neuquén basin oilfield brines is scarce. The only documented lithium analysis didn't show abnormal results (Pons *et al.* 2015). We encourage its measure since may let the incorporation of a new lithium source, which extraction would not require the drilling of new wells, benefiting companies by reducing their oil and gas fields operative costs, helping in the transition towards a circular economy.

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