

### Lithium Water Sustainability Project Work Package 2 Executive Report

### Simulating hydrologic responses to pumping scenarios at Salar de Atacama & Salar del Hombre Muerto

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### **Executive Summary**

Lithium brine extraction in South American salars has varied potential impacts on water resources and dependent environmental ecosystems. The physical and geochemical relationships between the lithium brine and surrounding freshwater wetlands are key factors controlling the environmental impacts of brine extraction. Our modeling study presented below constrains the relative magnitude of the impacts using key metrics of the environmental characteristics of these systems. We find that fresh groundwater and transitional brine withdrawals have a larger impact on total GW discharge than halite brine extractions. This result strongly suggests that fresh groundwater extractions should be minimized. Additionally, producers should avoid developing lithium mining operations or projects in transitional brine environments. These are regions typically between mature halite regions and freshwater systems. The timescales of impacts in general are longer for halite brine withdrawals and shorter for fresh groundwater and transitional brine withdrawals. This result has important implications for the timing of impacts and the length and quality of the monitoring programs used to detect changes or deleterious impacts. Our specific recommendations from these findings are: 1) a focus on environmental monitoring using innovative ways to measure spring discharge through remote sensing and *in situ* monitoring, 2) monitoring of salinity (and chemical composition) is important and complimentary to monitoring of physical groundwater levels and 3) that regardless of pumping conditions, the total amount of inflow to wetlands from groundwater will decrease and undoubtedly impact the salinity distribution of wetlands. Monitoring of salinity is key to understanding the impacts and the timescales at which they will be apparent.



### Introduction and Motivation of Study

The extraction of lithium from brine-rich salt flats (salars) in South America and associated environmental impacts is the subject of much debate (1). The magnitude of impacts and environmental sustainability depends on specific characteristics of the hydrogeological system and hydrological stresses on water in the basins where extraction is taking place. Our team has defined the following questions to address this issue:

- 1. How much freshwater recharges the brine aquifer system?
- 2. How hydraulically connected are the brine aquifer systems and the freshwater aquifers that feed marginal lagoons?
- 3. What controls the dynamics and extents of freshwater lagoons?
- 4. Does the freshwater/brine interface hydraulically support the lagoons?
- 5. What is the role of climate variability on observed changes to lagoon and transitional pool extents?
- 6. What are the timescales of response for freshwater and brine aquifer systems to pumping?

Salars are dynamic environments where freshwater (surface water and groundwater) moves into the basin floor and interacts with lithium-bearing brine (**Figure 1**). These freshwater inflows are driven to the surface to form springs, which then feed fresh and brackish surface water wetlands and lagoons. The springs are the main source of inflow to the wetland complexes and thus are critical to their existence. Yet the relationships between freshwater extraction and brine extraction on environmental features remains obscure. While it is surmised that resource extraction in these environments could lead to negative Impacts, questions remain about the magnitude and timing of such impacts.



Figure 1. Conceptual illustration of groundwater flow in the brine-to-freshwater transition zone.

### Water Types and Interactions

It's important to clearly define three primary water groupings and their relationship to one another in these systems. Freshwaters consist of streams and groundwaters flowing towards the salars from the higher elevations and marginal alluvial aquifers. These waters range from 0.05 to 3 mS/cm of specific conductance. Brackish waters are saline groundwaters and surface waters (lagoons) that exist within the Transition Zone, the zone between freshwater inflow and the nucleus brine aquifers. These waters range from 10 to 100 mS/cm of specific conductance. Brines are highly saline waters that reside within the evaporite aquifers of the salar nucleus and in pools that form along the margins of the salar nucleus, herein referred to as Transitional Pools. These waters range from 200 to >250 mS/cm of specific conductance. The interactions between these waters are key to understanding responses and impacts in these environments. Due to large density differences between the freshwater and the brine, a sharp interface forms at the margin of these salars in the subsurface and is a major control on where fresh groundwater discharge and springs occur and brackish wetlands form (2, 3). This sharp density contrast also creates distinct flow regimes within the brine aquifers as compared to the freshwater inflow aquifers.

### Work Package Scope and Approach

The purpose of this study is to develop a physical basis for assessing the impacts of brine and freshwater withdrawals on water resources in salar systems. We perform this analysis with a parametric study of the relative impacts of these withdrawals on the quantity and quality of groundwater discharge as well as response times of these impacts using two-dimensional, numerical, density-dependent groundwater flow models. The models represent the primary groundwater flow pathway in each of three groundwater flow systems located at Salar de Atacama, Chile, and Salar del Hombre Muerto, Argentina (**Figure 1b**). These three systems have distinct hydrogeologic characteristics and climate regimes which provide separate endpoints to compare and isolate the effects of brine and fresh groundwater withdrawals.

These three models utilize the best available geologic, hydrogeologic, and climate data to represent groundwater flow conditions along a single groundwater flow pathway (**Figure 1a**). Using these inputs, the models simulate groundwater flow and discharge as well as dissolved salt concentrations, which show changes in the brine-freshwater interface. We then apply groundwater withdrawals at the upgradient boundary of the model domain and brine withdrawals either from the brine body in the salar nucleus (nucleus brine) or from the brine body beneath the brine-freshwater interface (transition zone brine). We individually test withdrawals at each of these locations using a range of groundwater pumping rates and record the changes in simulated groundwater discharge and the brine-freshwater interface. We then compare these changes and the response times over which these changes occur for each model to evaluate the relative impacts of different types of withdrawals. Finally, we compare the simulated changes between the three models to evaluate the role of hydrogeologic conditions in withdrawal impacts.

The purpose of this study is to develop a physical basis for assessing the impacts of brine and freshwater withdrawals on water resources in salar systems.

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**Figure 2.** Map of the Lithium Triangle region of the north-central Andes. Major lithium brine bearing salar basins are outlined in black and mean annual precipitation derived from the TerraClimate dataset is shown. The basins assessed in this report are outlined in red.



### Salar de Atacama

### Model simulations of groundwater and brine withdrawals and relative impact evaluation at Salar de Atacama

### Introduction and Model Objectives

The Salar de Atacama contains the world's largest lithium reserve (approximately 42% of the world's supply) and has one active lithium mine and multiple pilot plants as of 2021 (4). These lithium mines withdraw brine from the halite aquifer and fresh groundwater from the transition zone. Additional water users in the basin include members of a number of communities, including the indigenous Lickanantay and the city of San Pedro de Atacama, agriculture, a significant tourism industry, and a copper mining industry, all of which use fresh groundwater either directly or indirectly. The basin also contains wetlands of international importance as defined by the Ramsar Convention, and these and other wetlands in the basin are sustained by and dependent upon groundwater inflows. The 2dimensional groundwater flow and transport model for this basin evaluates the relative impact of the fresh groundwater withdrawals from multiple users as well as the brine withdrawals from lithium mining activities in the halite aquifer (halite brine withdrawals). The model also evaluates a hypothetical scenario in which brine withdrawals occur in the transition zone outside of the halite aquifer (transitional brine withdrawals). We then measure the simulated change in groundwater discharge and the brine-freshwater interface relative to a baseline scenario in which no withdrawals occur and compare the relative impacts of each withdrawal scenario.

### **Model Design**

The Salar de Atacama model is located along the southeastern margin of the salar where the Monturaqui-Negrillar-Tilopozo (MNT) Aquifer enters the salar Transition Zone, extending to near the center of the salar Nucleus (Figure 3). This domain follows the primary groundwater flow path into the southern Transition Zone, including freshwater wetlands such as Tilopozo, the brackish lagoons Punta and Brava, and brine-bearing Transitional Pools along the nucleus margin. We developed a geologic conceptual model of the subsurface in the area of the model domain. We subsequently established a hydrogeologic framework, wherein we ascribed hydraulic properties to each lithostratigraphic unit for the groundwater flow model.

Additional water users in the basin include members of a number of communities,... agriculture, a significant tourism industry, and a copper mining industry...





**Figure 3.** Map of the Salar de Atacama basin showing the location of the 2-D geologic and hydrogeologic models along with significant features.

### **Geologic Conceptual Model**

We developed a geologic conceptual model for the southeastern margin of Salar de Atacama using several sources of information including surface geological maps, drill hole core logs, geophysical surveys, and available literature, as well as the commonly accepted concepts of sedimentary geology to further inform the distribution of lithologies in this part of the basin (Figure 4). The source of the primary information used to develop this model is detailed in the supplemental material of McKnight et al. (2021) and in Munk et al. (2021). Generally, the geology can be described as ignimbrite overlying undifferentiated basement units, overlain by alluvial sediments along the basin edge interfingering with evaporite sediments towards the salar-ward end of the transect. Moving further into the salar, evaporite sequences transition into a very thick and extensive halite unit. Lithostratigraphic units identified here include alluvium, silt & fine sand w/clay, silt, halite, gypsum, carbonate, ash, ignimbrite, and undifferentiated basement (Figure 2b). While smaller units that may not necessarily fit into one of the categorizations were observed in cores, it is important to note that lithologic characterization occurred at the meter scale and therefore small-scale lithologic heterogeneity was not captured within the geologic conceptualization. We subsequently categorized the geologic model into distinct hydrostratigraphic units.



Figure 4. Salar de Atacama geologic conceptual model showing geological units, major faults and wells along the transect with core data. The general locations of important surface water features are labeled.

We used the hydrogeologic conceptual model to create a twodimensional, numerical, densitydependent groundwater flow model...

### Hydrogeologic Conceptual Model

We compiled hydraulic properties for each of the hydrostratigraphic units identified in the geologic conceptual model from available pumping test data. These hydraulic properties include hydraulic conductivity, specific storage, and specific yield. Where pumping test data were unavailable, we estimated these properties based on a range of published values from similar geologic units. In addition, the halite aquifer exhibits depth-dependent hydraulic conductivity due to compaction and a resulting reduction in permeability with depth. Therefore, we developed depth-dependent hydraulic conductivity values for the halite unit based on the available pumping test data. **Table S1** lists the hydrogeologic parameters used in the model.

### **Groundwater Flow Model**

We used the hydrogeologic conceptual model to create a twodimensional, numerical, density-dependent groundwater flow model using the USGS program SEAWAT (5). The Salar de Atacama groundwater flow model domain is 24 km long and 100 m wide. The bottom of the domain is fixed at elevation 1,950 m, and the top of the domain is set to a smoothed topographic digital elevation model (DEM). The model contains 64 layers, and each layer is discretized into 100 m wide by 100 m long cells (**Figure 5**). The bottom of the top layer is elevation 2,298 m with a top set to the DEM elevation. The remaining layers have thicknesses of 2 m from elevation 2,298 m to 2,250 m, 5 m thick from elevation 2,250 m to 2,160 m, and 10 m thick below elevation 2,160 m.

Boundary conditions describe how the model interacts with external factors, using reasonable assumptions rather than explicitly modeling these processes. Groundwater recharge enters the model domain along the upgradient (left) boundary in the upper 15 model layers at a rate of 500 cubic meters per day  $(m^3/d)$ . This value is derived from previous groundwater recharge estimates for the MNT aquifer (Munk et al., 2018). The right model boundary contains a general head boundary condition, which adds brine recharge into the model or brine flux out of the model to maintain a constant brine elevation of 2,302.33 meters. This brine elevation is a long-term average of measurements in nearby wells. A conductance term controls the flux on the general head boundary, limiting the magnitude of the flux to prevent unreasonably large brine recharge rates. An evapotranspiration boundary condition on the top of the model domain simulates groundwater discharge. This boundary condition causes groundwater flux out of the model domain when the water table is within one meter of the ground surface; it simulates evapoconcentration by not removing salt from the model domain. The evapotranspiration boundary simulates the combined effects of evaporation and spring discharge. All remaining model boundaries are a no-flow condition, which does not allow water or salt flux in or out of the model domain.

We ran the model until the salt concentrations in the model domain reached a dynamic steady state. We then used the final conditions of the pseudo-steady state model as the initial conditions for the model simulations. These initial conditions represent long-term equilibrium groundwater conditions without anthropogenic influence. We then used this model to run 13 simulations representing groundwater withdrawals from three different locations - fresh groundwater withdrawals, halite brine withdrawal, and transition zone brine withdrawal (**Figure 2c**). Withdrawal rates ranged between a minimum rate of 10% of fresh groundwater recharge and a maximum rate of 40% of fresh groundwater recharge. For reference, in 2014, actual freshwater withdrawals for lithium mining in the MNT aquifer were approximately 1.3% of fresh groundwater recharge and actual halite brine withdrawals were approximately 40% of fresh groundwater recharge (*1*). The simulations are summarized in **Table 1**.



Simulation Number	Fresh Groundwater Recharge (m <sup>3</sup> /d)	Fresh Groundwater Withdrawals (m <sup>3</sup> /d)	Halite Brine Withdrawals (m <sup>3</sup> /d)	Transitional Brine Withdrawals (m <sup>3</sup> /d)	Total Withdrawals (% of Recharge)	
Baseline						
0	500	0	0	0	0%	
Fresh Ground	water Withdrawals	S				
1	500	50	0	0	10%	
2	500	100	0	0	20%	
3	500	150	0	0	30%	
4	500	200	0	0	40%	
Halite Brine Withdrawals						
5	500	0	50	0	10%	
6	500	0	100	0	20%	
7	500	0	150	0	30%	
8	500	0	200	0	40%	
Transitional Brine Withdrawals						
9	500	0	0	50	10%	
10	500	0	0	100	20%	
11	500	0	0	150	30%	
12	500	0	0	200	40%	



**Figure 5.** Salar de Atacama hydrogeologic conceptual model showing hydrogeologic units, starting conditions for all simulations, withdrawal locations for the various simulations, and boundary conditions for the groundwater flow model.

### Results

We investigate the effects of fresh groundwater and brine withdrawals on the groundwater system at Salar de Atacama by comparing the simulated conditions for the fresh groundwater, halite brine, and transitional brine scenarios to the baseline scenario. We measure the simulated conditions primarily as change relative to the baseline simulation, which isolates the impacts of the withdrawals from other variables. The aspects of the system that we evaluate are the overall flow dynamics, groundwater discharge, and the location of the brine-freshwater interface.

### **Flow Dynamics**

Flow dynamics describe how fluid moves through the system as a whole. In the Salar de Atacama model, fresh groundwater flows into the model domain on the left (upgradient) boundary and exits the domain at the top model boundary as groundwater discharge. Salt does not exit the model domain, leading to evapoconcentration of salt and the formation of brine. When groundwater or brine withdrawals occur, both the amount of fluid flowing through the system changes and the relative proportion of fresh groundwater to brine changes. **Figure 6** shows the flow dynamics after 100 years of continuous withdrawals for Simulation 0 (baseline scenario), Simulation 4 (fresh groundwater withdrawal at the maximum rate), Simulation 8 (halite brine withdrawals at the maximum rate).

In the baseline scenario, the majority of groundwater discharge occurs as fresh discharge, with some brackish discharge and almost no brine discharge. Most of the groundwater flow occurs in the fresh groundwater, with very little flow in the brine body. In the fresh groundwater withdrawal scenario (Simulation 4), the upgradient fresh groundwater discharge decreases significantly, and brackish discharge increases proportionally. As the relative proportion of fresh groundwater in the system decreases, the elevation of the brine-freshwater interface increases; however, the location of the brine-freshwater interface at the water table remains relatively unchanged.

In the halite brine withdrawal scenario (Simulation 8), groundwater discharge remains relatively unchanged. As the relative proportion of brine in the system decreases, the elevation of the brine-freshwater interface decreases. At the water table, the freshwater boundary of the brine-freshwater interface remains relatively unchanged, but the brine boundary moves toward the salar, causing the brine-freshwater interface to grow wider. Brine flow velocities increase and the brine flow vectors change direction from towards the brine-freshwater interface to towards the nucleus, in the direction of the brine withdrawals.

In the transitional brine withdrawal scenario (Simulation 12), upgradient fresh groundwater discharge decreases and the downgradient discharge increases proportionately. Brine flow velocities increase significantly, and the flow directions point towards the location of the brine withdrawals. In the fresh groundwater, flow paths diverge near the brine withdrawal location, with deeper paths flowing downwards towards the brine withdrawal location and shallower paths continuing to flow upward towards the surface. The elevation of the brine-freshwater interface decreases and a cone of depression is visible in the shape of the interface where the brine withdrawals occur. At the water table, the brine-freshwater interface moves towards the nucleus and narrows. ...the majority of groundwater discharge occurs as fresh discharge...



**Figure 6.** Flow dynamics after 130 years of simulated flow for the Salar de Atacama model simulations. Fluid flux vectors show both the direction and relative magnitude of groundwater/brine flow. Groundwater discharge vectors at the water table show the location and relative magnitude of groundwater discharge. Image frames show a subset of the groundwater flow model focusing on the brine-freshwater interface. **A)** Baseline flow dynamics with no fluid withdrawals. **B)** Flow dynamics with fresh groundwater withdrawals on the left model boundary at a rate of 40% of the total fresh groundwater recharge. **C)** Flow dynamics with halite brine withdrawals from a well located to the right of the image frame at a rate of 40% of the total fresh groundwater recharge. **D)** Flow dynamics with transitional brine withdrawals from beneath the image frame at a rate of 40% of the total fresh groundwater recharge.

### Groundwater Discharge

Groundwater discharge is a combination of spring discharge and groundwater evapotranspiration, which is removed from the subsurface by evaporation or transpiration before the water reaches the ground surface. Both spring discharge and transpiration play a critical role in supporting wetland ecosystems. We measure the simulated groundwater discharge as the flux of groundwater out of the top of the model domain. We then calculate the change in groundwater discharge relative to the baseline scenario ( $\Delta O$ ) to evaluate the impacts of withdrawals on groundwater discharge. In addition, we separate out  $\Delta Q$  into three categories. Fresh discharge is groundwater discharge in the region of the model where the salt concentration of water at the discharge point is less than 50 g/L in the baseline simulation. Brackish discharge is groundwater discharge in the region of the model where the salt concentration of water at the discharge point is 50-150 g/L in the baseline simulation, and brine discharge is groundwater discharge in the region of the model where the salt concentration of water at the discharge point is greater than 150 g/L in the baseline simulation. Figure 7 shows the  $\Delta Q$  as a percentage of total groundwater recharge for each simulation across the whole model domain as well as for each of the three discharge categories.



**Figure 7.** Maximum change in groundwater discharge ( $\Delta Q$ ) within 100 years of constant rate pumping in Salar de Atacama simulations, reported as a percentage of freshwater recharge to the model. Simulations included fresh groundwater withdrawals, halite brine withdrawals, and transitional brine withdrawals at 10-40% of the model's groundwater recharge rate.  $\Delta Q$  is presented as total  $\Delta Q$  and is also subdivided into fresh, brackish, and brine discharge areas.

In all withdrawal scenarios, withdrawals result in a decrease in total groundwater discharge. Fresh groundwater withdrawals cause the greatest decrease in groundwater discharge, with a 1:1 relationship between fresh groundwater withdrawals and  $\Delta Q$ . Halite brine withdrawals have the least impact on  $\Delta Q$ , with fresh groundwater withdrawals resulting in 167% more loss of groundwater discharge than halite brine withdrawals at the highest withdrawal rate. The impacts of transitional brine withdrawals are similar to those of fresh groundwater withdrawals, with total groundwater discharge decreasing slightly less than the equivalent fresh groundwater withdrawal scenarios.

For all three withdrawal scenarios, the majority of the change in spring discharge occurs in the fresh discharge area. The impacts of fresh groundwater withdrawals and transitional brine withdrawals on the fresh discharge area are nearly identical, whereas the impacts from halite brine withdrawals are significantly less. Decreases in fresh discharge in the transitional brine withdrawal scenarios may be a result of the diverging fresh groundwater flow paths described above.

Simulations for all three withdrawal scenarios show a slight increase in groundwater discharge in the brackish discharge area. Brine withdrawal simulations result in a greater increase in groundwater discharge in the brackish discharge area than fresh groundwater withdrawals, with transitional brine withdrawals causing the greatest increase. These increases in brackish groundwater discharge are likely a result of the change in flow dynamics, which causes groundwater discharge to move downgradient towards the brackish discharge area. While some increase in brackish discharge may occur, it does not offset the loss of fresh discharge in any of the simulations.

There is very little brine discharge at Salar de Atacama owing to the position and elevation of the brine body. In the baseline simulation, total brine discharge is approximately 1% of fresh groundwater recharge. A 1% decrease in  $\Delta Q$  results in a total loss of all brine discharge. All three withdrawal scenarios meet this threshold, with halite brine withdrawals and transitional brine withdrawals meeting the threshold at all simulated withdrawal rates and the freshwater withdrawal scenario meeting this threshold at the highest withdrawal rate.

Fresh groundwater withdrawals cause the greatest decrease in groundwater discharge...



### **Response Time**

We evaluate response time by examining the rate of change in groundwater discharge for each of the simulations. When withdrawals occur,  $\Delta Q$  changes with time according to an exponential decay function as defined by:

$$\Delta Q = Ae^{-kt} + c_0$$

where t is time, A is the initial value,  $c_0$  is an offset term, and k describes the rate of change. Where k is higher, groundwater discharge changes at a faster rate than when k is lower. **Figure 8** shows total  $\Delta Q$  over time along with the exponential decay trendline for the highest withdrawal rate simulations (40% of fresh groundwater recharge) for the fresh groundwater, halite brine, and transitional brine withdrawal scenarios. The k value is highest for the transitional brine withdrawal (k=7.16x10<sup>-2</sup>) and fresh groundwater withdrawal scenario (k=6.86x10<sup>-2</sup>) and lowest for the halite brine withdrawal scenario (k=3.21x10<sup>-2</sup>), indicating that transitional brine and fresh groundwater withdrawals cause groundwater discharge to decrease at a rate more than twice as fast as halite brine withdrawals in the Salar de Atacama model.



**Figure 8.** Salar de Atacama model timeseries of  $\Delta Q$  over the first 200 years for the maximum withdrawal rate simulations (40% of groundwater recharge) for the fresh groundwater withdrawal, halite brine withdrawal, and transitional brine withdrawal scenarios. Dashed lines represent the exponential decay trendline for each simulation, along with the rate of change constant (k) for each trend.

...transitional brine and fresh groundwater withdrawals cause groundwater discharge to decrease at a rate more than twice as fast as halite brine withdrawals...

### **Brine-Freshwater Interface**

We evaluate changes in the water table surface expression of the brine-freshwater interface, which indicates the change in composition and therefore the quality of groundwater discharge. The brine-freshwater interface is a zone of mixing between fresh groundwater and brine and is bounded by a fresh interface limit, defined here as the 50 g/L salt concentration isocontour, and a brine interface limit, defined here as the 150 g/L salt concentration isocontour. We measure the change in the position of the interface limits relative to the baseline for each of the model simulations. **Figure 9** shows the changes in the water table surface expression of the brine-freshwater interface.



**Figure 9.** Salar de Atacama model brine-freshwater interface boundaries' maximum migration distance during the first 100 years of continuous withdrawals.

Both the fresh interface limit and the brine interface limit migrate towards the salar nucleus in all withdrawal scenarios. In the freshwater withdrawal scenario, interface migration is negligible. In the halite brine withdrawal scenarios, the fresh interface limit migration is also negligible, but at halite, brine withdrawal rates greater than 10% of fresh groundwater recharge, the brine interface limit migrates 860-905 meters towards the nucleus. In the transitional brine withdrawal scenario, significant interface migration occurs on both the fresh interface limit and the brine interface limit, with up to approximately 1 km of potential interface migration.



## Salar del Hombre Muerto Eastern Subbasin

Model simulations of groundwater and brine withdrawals and relative impact evaluation at the Eastern Subbasin of Salar del Hombre Muerto.



There are currently three lithium mining claims in the eastern subbasin...

### Introduction and Model Objectives

Salar del Hombre Muerto contains two subbasins with distinct geologic and hydrogeologic regimes. There are currently three lithium mining claims in the eastern subbasin, and an active lithium mine in the western subbasin has a fresh groundwater well field in the southern portion of the eastern subbasin. There are currently no other significant economic activities or water users in the basin. The eastern subbasin contains a large brine lagoon (Laguna Verde) as well as a large wetlands complex called the Los Patos River Delta, which contains habitat for vulnerable flamingo species. The groundwater flow model for this subbasin evaluates the relative impact of the fresh groundwater withdrawals and halite brine withdrawals from lithium mining activities. We then measure the simulated change in groundwater discharge and the brine-freshwater interface relative to a baseline scenario and compare the relative impacts of each withdrawal scenario.

### **Model Design**

The Salar del Hombre Muerto Eastern Subbasin model is located along the southern margin of the subbasin where the Los Patos Aquifer enters the salar Transition Zone, extending past Laguna Verde (**Figure 10**). This domain follows the primary groundwater flow pathway into the eastern subbasin nucleus, including the Los Patos River Delta, containing fresh and brackish wetlands. We developed a geologic conceptual model of the subsurface in the area of the model domain. Then, using insights from the geologic conceptual model, we developed a hydrogeologic conceptual model as the framework for the groundwater flow model.

### **Geologic Conceptual Model**

A geologic conceptual model for the Eastern Subbasin was developed using several sources of information including surface geological maps, drill hole core logs, geophysical surveys, and available literature, as well as the commonly accepted concepts of sedimentary geology to further inform the distribution of lithologies in this part of the basin (Figure 11). The most important primary data came from technical reports with core log information and geophysical data conducted in the Los Patos Aquifer, Delta, and salar Transition Zone regions (locations of drill holes are shown in Figure 10). Generally, the geology can be described as ignimbrite overlying sedimentary and metamorphic basement, overlain by thick fluvial and colluvial sediments interfingering with evaporite sediments towards the north with a thick and extensive buried halite unit at the northern end of the transect. Lithostratigraphic units identified here include gravel & sand, sand & gravel w/clay, fine sand w/silt, red clay w/silt, calcareous gray silty clay, black calcareous clay, gypsum w/carbonate lenses, gypsum, halite, ignimbrite, and basement units (Figure 11). While smaller units that may not necessarily fit into one of the categorizations were observed in cores, it is important to note that lithologic characterization occurred at the meter scale and therefore not all geology at smaller scales were captured within the geologic conceptualization. The geologic model was subsequently categorized into distinct hydrostratigraphic units.



Figure 10. Map of the Salar del Hombre Muerto basin showing the location of the 2-D geologic and hydrogeologic models along with significant features.



**Figure 11.** Salar del Hombre Muerto Eastern Subbasin geologic conceptual model showing geological units and the wells along the transect with core data. The location of a geophysical survey line used to build this model is labeled (GFS – K-K'). The general locations of important surface water features are also labeled.

### Hydrogeologic Conceptual Model

We compiled hydraulic properties for each of the lithostratigraphic units identified in the geologic conceptual model based on either hydraulic testing conducted within the basin or adapted from previous hydrostratigraphic frameworks from our studies of hydrogeology in Salar de Atacama. In addition, the halite, gravel and sand, and fine sand and silt units exhibit depth-dependent hydraulic conductivity due to compaction and the resulting reduction in permeability with depth. We developed depth-dependent hydraulic conductivity values for these three units based on the available hydraulic testing data. **Table S2** lists the hydrogeologic parameters used in the model.

### **Groundwater Flow Model**

We created a two-dimensional, numerical, density-dependent groundwater flow model using the USGS program SEAWAT (5). The Salar del Hombre Muerto Eastern Subbasin groundwater flow model domain is 20 km long and 100 m wide. The bottom of the domain is fixed at elevation 3,800 m, and the top of the domain is set to a smoothed topographic DEM. The model contains 50 layers, with each layer discretized into 100 m wide by 100 m long cells (**Figure 12**). The thickness of the top layer is variable, depending on the elevation of the DEM which forms the top of the layer. The remaining layers have thicknesses of 2 m from elevation 3,964 m to 3,910 m and 5 m thick below elevation 3,910 m.

Groundwater recharge enters the model domain along the upgradient (left) boundary in the upper 25 model layers at a rate of  $350 \text{ m}^3/\text{d}$ . We estimated this recharge value based on climate data from Work Package 1 of this project. The right model boundary contains a general head boundary condition with a brine



elevation of 3,967 meters, which we estimated based on available well data in the subbasin. An evapotranspiration boundary condition on the top of the model domain simulates groundwater discharge and evapoconcentration. All remaining model boundaries are a no flow condition.

We ran the model until the salt concentrations in the model domain reached a pseudo-steady state. We then used the final conditions of the pseudo-steady state model as the initial conditions for the model simulations. These initial conditions represent long-term equilibrium groundwater conditions without anthropogenic influence. We then used this model to run 9 simulations representing baseline conditions, fresh groundwater withdrawals, and halite brine withdrawals from the buried halite aquifer (**Figure 12**). **Table 2** summarizes the simulation parameters.

	Fresh Groundwater	Fresh Groundwater	Halite Brine	Total		
Simulation	Recharge	Withdrawals	Withdrawals	Withdrawals		
Number	$(m^{3}/d)$	(m <sup>3</sup> /d)	(m <sup>3</sup> /d)	(% of Recharge)		
Baseline						
0	350	0	0	0%		
Fresh Groun	dwater Withdrav	vals				
1	350	35	0	10%		
2	350	70	0	20%		
3	350	105	0	30%		
4	350	140	0	40%		
Halite Brine	Halite Brine Withdrawals					
5	350	0	35	10%		
6	350	0	70	20%		
7	350	0	105	30%		
8	350	0	140	40%		

Table 2. Summary of Groundwater Model Simulations – Salar del Hombre Muerto Western Subbasin



**Figure 12.** Salar del Hombre Muerto Eastern Subbasin hydrogeologic conceptual model showing hydrogeologic units, starting conditions for all simulations, withdrawal locations for the various simulations, and boundary conditions for the groundwater flow model.

### Results

We investigate the effects of fresh groundwater and brine withdrawals on the groundwater system at the Salar del Hombre Muerto Eastern Subbasin by comparing the simulated conditions for the fresh groundwater and brine withdrawal scenarios to the baseline scenario. We measure the simulated conditions primarily as change relative to the baseline simulation, which isolates the impacts of the withdrawals from other variables. The aspects of the system that we evaluate are the overall flow dynamics, groundwater discharge, and the brine-freshwater interface.

### **Flow Dynamics**

When groundwater or brine withdrawals occur, flow dynamics in the system change as a result of perturbation in the amount of fluid flowing through the system as well as the relative proportion of fresh groundwater to brine. **Figure 13** shows the flow dynamics after 100 years of continuous withdrawals for the baseline scenario as well as the maximum withdrawal rate simulations for the fresh groundwater withdrawal and halite brine withdrawal scenarios.

In the baseline scenario, the majority of groundwater discharge occurs as fresh discharge, with almost no brackish or brine discharge. Most of the groundwater flow occurs in the fresh groundwater, with some flow in the brine-freshwater interface and very little flow in the brine body. In the fresh groundwater withdrawal scenario (Simulation 4), groundwater discharge at the most upgradient point decreases. Very little change occurs in the brine-freshwater interface, both at the water table and at depth.

In the halite brine withdrawal scenario (Simulation 8), groundwater discharge at the most upgradient point decreases, similar to Simulation 4. There is also a very slight increase in the relative proportion of brine and brackish discharge. Also similar to Simulation 4, very little change occurs in the brine-freshwater interface.



...very little change occurs in the brinefreshwater interface.





**Figure 13.** Flow dynamics after 130 years of simulated flow for the Salar del Hombre Muerto Eastern Subbasin model simulations. Fluid flux vectors show both the direction and relative magnitude of groundwater/brine flow. Groundwater discharge vectors at the water table show the location and relative magnitude of groundwater discharge. Image frames show a subset of the groundwater flow model focusing on the brine-freshwater interface. A) Baseline flow dynamics with no fluid withdrawals. B) Flow dynamics with fresh groundwater withdrawals on the left model boundary at a rate of 40% of the total fresh groundwater recharge. C) Flow dynamics with halite brine withdrawals from a well located to the right of the image frame at a rate of 40% of the total fresh groundwater recharge.

### Groundwater Discharge

We measure the simulated groundwater discharge as groundwater flux out of the top of the model domain along the evapotranspiration boundary. We then calculate  $\Delta Q$  to evaluate the relative impacts of withdrawals on groundwater discharge. In addition to evaluating the total groundwater discharge, we separate groundwater discharge into fresh discharge, brackish discharge, and brine discharge using the same methodology as the Salar de Atacama model. **Figure 14** shows the  $\Delta Q$  as a percentage of total groundwater recharge for each simulation across the whole model domain as well as for each of the three discharge categories.

Total groundwater discharge decreases relative to baseline in all withdrawal simulations. Similar to the Salar de Atacama model, fresh groundwater withdrawals cause the greatest decrease in groundwater discharge, with a 1:1 relationship between fresh groundwater withdrawals and  $\Delta Q$ . Halite brine withdrawals have a smaller impact on  $\Delta Q$ , with fresh groundwater withdrawals resulting in 135% more loss of groundwater discharge than halite brine withdrawals at the highest withdrawal rate.

For the fresh groundwater withdrawal scenario, the majority of the change in groundwater discharge occurs in the fresh discharge area. Fresh groundwater withdrawals have no significant effect on brackish discharge and cause a very small to negligible decrease in brine discharge.

In the halite brine withdrawal scenario, the majority of the change in groundwater discharge occurs in the brine discharge area. Halite brine withdrawals cause a small decrease in fresh discharge and a slight increase in brackish discharge.

### **Response Time**

We evaluate the groundwater discharge response time for each of the model simulations by calculating the rate of change (k) from the exponential decay function. **Figure 15** shows total  $\Delta Q$  over time along with the exponential decay trendline for the highest withdrawal rate simulations (40% of fresh groundwater recharge) for the fresh groundwater and halite brine withdrawal scenarios. The k value is highest for the fresh groundwater withdrawal scenario (k=1.34x10<sup>-1</sup>) and lowest for the halite brine withdrawal scenario (k=6.56x10<sup>-2</sup>), indicating that fresh groundwater withdrawals cause groundwater discharge to decrease at a rate more than twice as fast as halite brine withdrawals in the Salar del Hombre Muerto Eastern Subbasin model.



Halite brine withdrawals have a smaller impact on  $\Delta \mathbf{Q}...$ 



**Figure 14.** Maximum change in groundwater discharge ( $\Delta Q$ ) within 100 years of constant rate pumping in Salar del Hombre Muerto Eastern Subbasin simulations, reported as a percentage of freshwater recharge to the model. Simulations included fresh groundwater withdrawals and halite brine withdrawals at 10-40% of the model's groundwater recharge rate.  $\Delta Q$  is presented as total  $\Delta Q$  and is also subdivided into fresh, brackish, and brine discharge areas.





### **Brine-Freshwater Interface**

We evaluate changes in the water table surface expression of the brinefreshwater interface using the same methods described for the Salar de Atacama model. We measure the change in the position of the fresh interface limits and the brine interface limits relative to the baseline for each of the model simulations. **Figure 16** shows the changes in the water table surface expression of the brine-freshwater interface.

The position of the fresh interface limit and the brine interface limit remained relatively constant for all withdrawal simulations. The maximum migration distance was 10.3 m for the fresh interface limit and 12.9 m for the brine interface limit. The Salar del Hombre Muerto Eastern Subbasin model has a low topographic relief with a water table near the ground surface along most of the model domain. As a result, evapoconcentration affects salt concentrations in groundwater across a large portion of the water table, making evapoconcentration the primary driver of interface position at the water table rather than groundwater flow dynamics.



Figure 16. Salar del Hombre Muerto Eastern Subbasin model brine-freshwater interface boundaries' maximum migration distance during the first 100 years of continuous withdrawals.



### Salar del Hombre Muerto Western Subbasin

Model simulations of groundwater and brine withdrawals and relative impact evaluation in the Salar del Hombre Muerto Western Subbasin



The groundwater flow model for this subbasin evaluates the relative impact of the fresh groundwater withdrawals and halite brine withdrawals from lithium mining activities.

### **Introduction and Model Objectives**

The western subbasin of Salar del Hombre Muerto has one active lithium mine operating in the salar nucleus. There are currently no other significant economic activities or water users in this subbasin. The western subbasin contains a large brine lagoon (Laguna Catal) as well as a former vega that has been largely replaced by an artificial brine lagoon created by reinfiltration of brine from lithium mining operations. The groundwater flow model for this subbasin evaluates the relative impact of the fresh groundwater withdrawals and halite brine withdrawals from lithium mining activities. We then measure the simulated change in groundwater discharge and the brine-freshwater interface relative to a baseline scenario and compare the relative impacts of each withdrawal scenario.

### **Model Design**

The Salar del Hombre Muerto Western Subbasin model is located along the southern margin of the western part of the Hombre Muerto basin where the Trapiche Aquifer enters the salar transition zone, extending to the center of the salar nucleus to the west of Laguna Catal (**Figure 10**). This domain follows the primary groundwater flow pathway into the western subbasin nucleus, including the Artificial Lagoon. This study does not evaluate the effects of brine re-infiltration and therefore simulates a hypothetical scenario in which brine re-infiltration does not occur to isolate the effects of withdrawals on the system. We developed a geologic conceptual model of the subsurface in the area of the model domain. Then, using insights from the geologic conceptual model, we developed a hydrogeologic conceptual model as the framework for the groundwater flow model.

### **Geologic Conceptual Model**

A geologic conceptual model for the Western Subbasin was developed using several sources of information including surface geological maps, drill hole core logs, and available literature, as well as the commonly accepted concepts of sedimentary geology to further inform the distribution of lithologies in this part of the basin (Figure 17). The most important primary data came from technical reports provided by Livent Corp. from the Trapiche aquifer and the western salar nucleus (locations of drill holes are shown in Figure 10). Generally, the geology can be described as ignimbrite overlying sedimentary basement, with alluvial fan and halite deposits to the surface. Lithostratigraphic units of the western subbasin include coarse sand and gravel, silt and fine sand w/clay, halite with clastics, halite, ignimbrite, and Paleozoic basement (Figure 17). While smaller units that may not necessarily fit into one of the categorizations were observed in cores, it is important to note that lithologic characterization occurred at the meter scale and therefore not all geology at smaller scales were captured within the geologic conceptualization. The geologic model was subsequently categorized into distinct hydrostratigraphic units.



**Figure 17.** Salar del Hombre Muerto Western Subbasin geologic conceptual model showing geological units and the wells along the transect with core data. The general location of important hydrologic zones discussed in this report and important surface water features are labeled.

### Hydrogeologic Conceptual Model

The development of the hydrogeologic conceptual model of the Western Subbasin is similar to the hydrogeologic framework of the Eastern Subbasin. Both subbasins rely on the same hydraulic dataset that was collected and synthesized. Similar to the eastern subbasin, we developed depth-dependent hydraulic conductivity values for the halite, gravel, and sand, and fine sand and silt units. **Table S3** lists the hydrogeologic parameters used in the model.

### **Groundwater Flow Model**

We created a two-dimensional, numerical, density-dependent groundwater flow model using the USGS program SEAWAT (5). The Salar del Hombre Muerto Western Subbasin groundwater flow model domain is 20.9 km long and 100 m wide. The bottom of the domain is fixed at elevation 3,740 m, and the top of the domain is set to a smoothed topographic DEM. The model contains 51 layers, with each layer discretized into 100 m wide by 100 m long cells (**Figure 18**). The thickness of the top layer is variable depending on the elevation of the DEM. The remaining layers have thicknesses of 2 m from elevation 3,962 m to 3,910 m, 5 m from elevation 3,910 m to 3,840 m, and 10 m thick below elevation 3,840 m.

Groundwater recharge enters the model domain along the upgradient (left) boundary in the top model layer at a rate of 200 m<sup>3</sup>/d. We estimated this recharge value based on climate data from Work Package 1 as well as previous studies in the Trapiche Aquifer. The right model boundary contains a general head boundary condition with a brine elevation of 3,967.2 m, which we estimated based on a long-term average of available well data in the salar nucleus. An evapotranspiration boundary on the top of the model domain in the transition zone simulates groundwater discharge

and evapoconcentration. All remaining model boundaries are a no flow condition.

We ran the model until the salt concentrations in the model domain reached a pseudo-steady state. We then used the final conditions of the pseudosteady state model as the initial conditions for the model simulations. These initial conditions represent long-term equilibrium groundwater conditions without anthropogenic influence. We then used this model to run 9 simulations representing baseline conditions, fresh groundwater withdrawals, and halite brine withdrawals (**Figure 18**). **Table 3** summarizes the simulation parameters.



**Figure 18.** Salar del Hombre Muerto Western Subbasin hydrogeologic conceptual model showing hydrogeologic units, starting conditions for all simulations, withdrawal locations for the various simulations, and boundary conditions for the groundwater flow model.



### Results

We investigate the effects of fresh groundwater and brine withdrawals on the groundwater system at the Salar del Hombre Muerto Western Subbasin by comparing the simulated conditions for the fresh groundwater and brine withdrawal scenarios to the baseline scenario. We measure the simulated conditions primarily as change relative to the baseline simulation, which isolates the impacts of the withdrawals from other variables. The aspects of the system that we evaluate are the overall flow dynamics, groundwater discharge, and the brine-freshwater interface.

### **Flow Dynamics**

When groundwater or brine withdrawals occur, flow dynamics in the system change as a result of perturbation in the amount of fluid flowing through the system as well as the relative proportion of fresh groundwater to brine. **Figure 19** shows the flow dynamics after 100 years of continuous withdrawals for the baseline scenario as well as the maximum withdrawal rate simulations for the fresh groundwater withdrawal and halite brine withdrawal scenarios.



**Figure 19.** Flow dynamics after 130 years of simulated flow for the Salar del Hombre Muerto Western Subbasin model simulations. Fluid flux vectors show both the direction and relative magnitude of groundwater/brine flow. Groundwater discharge vectors at the water table show the location and relative magnitude of groundwater discharge. Image frames show a subset of the groundwater flow model focusing on the brine-freshwater interface. A) Baseline flow dynamics with no fluid withdrawals. **B)** Flow dynamics with fresh groundwater withdrawals on the left model boundary at a rate of 40% of the total fresh groundwater recharge. **C)** Flow dynamics with halite brine withdrawals from a well located to the right of the image frame at a rate of 40% of the total fresh groundwater recharge.

In the baseline scenario, the majority of groundwater discharge occurs as fresh discharge, with some significant brackish and brine discharge. Similar to the other two models, most of the groundwater flow occurs in the fresh groundwater, with some flow in the brine-freshwater interface and very little flow in the brine body. In the fresh groundwater withdrawal scenario (Simulation 4), groundwater discharge decreases in the more upgradient locations, with very little change in brackish and brine discharge. As the relative proportion of fresh groundwater in the system decreases, the brine-freshwater interface moves slightly upward; however, the location of the brine-freshwater interface at the water table remains relatively unchanged.

In the halite brine withdrawal scenario (Simulation 8), fresh discharge also shifts downgradient. The relative proportion of brackish and brine discharge also decreases. At the water table, the fresh groundwater limit of the brine-freshwater interface remains relatively unchanged, but the brine limit migrates somewhat towards the nucleus. At depth, the fresh groundwater limit of the brine-freshwater interface moves upward and the brine limit moves downward, resulting in an overall widening of the brine-freshwater interface.

### **Groundwater Discharge**

We measure the simulated groundwater discharge as groundwater flux out of the top of the model domain along the evapotranspiration boundary. We then calculate  $\Delta Q$  to evaluate the relative impacts of withdrawals on groundwater discharge. In addition to evaluating the total groundwater discharge, we separate groundwater discharge into fresh discharge, brackish discharge, and brine discharge using the same methodology as the Salar de Atacama model. **Figure 20** shows  $\Delta Q$  as a percentage of total groundwater recharge for each simulation across the whole model domain as well as for each of the three discharge categories.

Total groundwater discharge decreases relative to baseline in all withdrawal simulations. Similar to the other two models, fresh groundwater withdrawals cause the greatest decrease in groundwater discharge, with a 1:1 relationship between fresh groundwater withdrawals and  $\Delta Q$ . Halite brine withdrawals have a smaller impact on  $\Delta Q$ , with fresh groundwater withdrawals resulting in 130% more loss of groundwater discharge than halite brine withdrawals at the highest withdrawal rate.

For the fresh groundwater withdrawal scenario, the majority of the change in groundwater discharge occurs in the fresh discharge area. Fresh groundwater withdrawals cause small decreases in brackish and brine discharge as well, with the least impact on brackish discharge.

In the halite brine withdrawal scenario, the majority of the change in groundwater discharge occurs in the fresh and brine discharge areas, with a very small to negligible decrease in brackish discharge.

Total groundwater discharge decreases relative to baseline in all withdrawal simulations.



**Figure 20.** Maximum change in groundwater discharge ( $\Delta Q$ ) within 100 years of constant rate pumping in Salar del Hombre Muerto Western Subbasin simulations, reported as a percentage of freshwater recharge to the model. Simulations included fresh groundwater withdrawals and halite brine withdrawals at 10-40% of the model's groundwater recharge rate.  $\Delta Q$  is presented as total  $\Delta Q$  and is also subdivided into fresh, brackish, and brine discharge areas.

### **Response Time**

We evaluate the groundwater discharge response time for each of the model simulations by calculating the rate of change (k) from the exponential decay function. **Figure 21** shows total  $\Delta Q$  over time along with the exponential decay trendline for the highest withdrawal rate simulations (40% of fresh groundwater recharge) for the fresh groundwater and halite brine withdrawal scenarios. The k value is highest for the fresh groundwater withdrawal scenario (k=3.49x10<sup>-2</sup>) and lowest for the halite brine withdrawal scenario (k=2.68x10<sup>-2</sup>), indicating that fresh groundwater withdrawals cause groundwater discharge to decrease at a rate of approximately 30% faster than halite brine withdrawals in the Salar del Hombre Muerto Eastern Subbasin model.

### **Brine-Freshwater Interface**

We evaluate changes in the water table surface expression of the brine-freshwater interface using the same methods described for the Salar de Atacama model. We measure the change in the position of the fresh interface limits and the brine interface limits relative to the baseline for each of the model simulations. **Figure 22** shows the changes in the water table surface expression of the brine-freshwater interface.

For the fresh groundwater withdrawal scenario, no significant change occurs in the fresh interface limit. The brine interface limit migrates somewhat, up to 71 m in the highest withdrawal simulation. Significantly more interface migration occurs in the brine withdrawal scenario, with the fresh interface limit migrating up to 130 m towards the nucleus and the brine interface limit migrating up to 655 m towards the nucleus.



**Figure 21.** Salar del Hombre Muerto Western Subbasin model timeseries of  $\Delta Q$  over the first 200 years of extraction for the maximum withdrawal rate simulations (40% of groundwater recharge) for the fresh groundwater withdrawal and halite brine withdrawal scenarios. Dashed lines represent the exponential decay trendline for each simulation, along with the rate of change constant (k) for each trend.

Significantly more interface migration occurs in the brine withdrawal scenario...



Figure 22. Salar del Hombre Muerto Western Subbasin model brine-freshwater interface boundaries' maximum migration distance during the first 100 years of continuous withdrawals.



### Conclusions

We summarize our data-driven interpretations & their implications.

### **Comparison of Salar Systems**

Each salar system has unique hydrogeologic characteristics that cause the systems to respond differently to perturbations from fresh groundwater and brine withdrawals. We compare the Salar de Atacama and the Salar del Hombre Muerto Eastern and Western Subbasin models to identify features that may impact how these systems respond to withdrawals.

### Geology & Hydroclimate

These salar systems are quite similar in terms of their geological and hydroclimatological characteristics but exhibit a few key differences that have a strong influence on the hydrodynamics observed in each. The salars and their watersheds have hyper-arid to arid climates and are composed of some combination of alluvial deposits overlying thick volcanic units in their inflow/upgradient zone which then interfinger to different degrees with evaporite sediments near the basin floors, the general framework of which is outlined in **Figure 1**. Their arid climate creates high evaporation rates, low recharge rates, and deep water tables. However, the basin floor at Salar del Hombre Muerto is >1,700 meters higher in elevation than Salar de Atacama. This leads to a somewhat wetter climate and lower evaporation rates in the transition zones of the basin. These slightly different hydroclimatic conditions and the particular orientation of the Eastern Subbasin also allow for the persistence of a large river, Rio Los Patos, that feeds the Salar del Hombre Muerto transition zone. The interaction between these conditions as well as geographical and geological differences is important to interpreting the results presented here.

The topographic gradients of these three systems are also distinct, leading to corresponding differences in hydraulic gradients from their inflows to discharge. These differences result from variations in thickness, lateral width, and composition of the sedimentary deposits and surface expression of their Transition Zones. The Eastern Subbasin of Salar del Hombre Muerto has the shallowest topographic gradient of the three systems as it lies within the well-developed alluvial aquifer of the Los Patos River and its Delta. This creates a wide and thick Transition Zone from freshwater inflow to brackish to brine and allows for a relatively shallow water table throughout the system. This system also has the overall lowest hydraulic conductivity. The Western Subbasin of Salar del Hombre Muerto has the steepest gradient and as a result, a very narrow Transition Zone, with groundwater discharge occurring over a short lateral distance. The Salar de Atacama system lies in between these two in terms of gradient and the resulting width of the Transition Zone and the zone of groundwater discharge. This system also has an overall higher hydraulic conductivity, which is a coefficient used to describe the permeability of subsurface porous media, within the model domain than the other two. These natural distinctions and the resulting hydrology drive much of the results we describe here and can serve as effective frameworks to understanding a large range of salar systems across the Altiplano region.



Their arid climate creates high evaporation rates, low recharge rates, and deep water tables.

### **Flow Dynamics**

In all systems, withdrawals can affect the distribution of groundwater discharge, the direction of fluid flow, and the size and position of the brine-freshwater interface. We compare these aspects of each of the three groundwater flow models as well as how these flow dynamics respond to withdrawals in each of the systems.

In all three models, the majority of groundwater discharge occurs as fresh discharge relatively close to the brine-freshwater interface. This relationship exists because the denser brine tends to flow downward, whereas the less dense fresh groundwater flows upward towards the ground surface where it comes into contact with the brine body. Topography and the depth of the water table relative to the ground surface also play a role in the distribution of groundwater discharge. Systems with lower topographic relief (i.e., Salar de Atacama) have a wide distribution of groundwater discharge locations (Figure 6), whereas systems with high topographic relief (Salar del Hombre Muerto Western Subbasin) have a focused zone of groundwater discharge (Figure 19). The position of the water table relative to the ground surface also affects the relative proportion of brackish and brine discharge. For example, in Salar de Atacama, where the water table is further below ground surface in the nucleus, there is very little brackish and brine discharge in the system. A larger proportion of brackish and brine discharge occurs in the Salar del Hombre Muerto models, where the water table is comparatively shallow in the nucleus. In all models, groundwater discharge locations migrate downgradient (towards the brine-freshwater interface) as the water table declines due to withdrawals.

The brine-freshwater interface width and orientation are also influenced by topographic relief and subsurface architecture. In the Salar de Atacama model, where topographic relief is relatively low and hydraulic conductivities are relatively high, the brine-freshwater interface is relatively narrow and shallow dipping. In the Salar del Hombre Muerto Western Subbasin model where topographic relief is relatively high and hydraulic conductivities are also high, the brine-freshwater interface is relatively narrow and steeply dipping. In contrast, in the Salar del Hombre Muerto Eastern Subbasin model topographic relief is relatively low, and hydraulic conductivities near the water table are low. In this model (Figure 13), the brinefreshwater interface is wide near the water table where evapoconcentration dominates, and the brine-freshwater interface is shallow dipping beneath this zone. These show that topographic relief is the dominant control on brine-freshwater interface orientation, with the brine-freshwater interface steepness mirroring topographic relief, and hydraulic conductivity is the dominant control on brinefreshwater interface width, with the interface becoming wider with lower hydraulic conductivities.

...the denser brine tends to flow downward, whereas the less dense fresh groundwater flows upward towards the ground surface...

### Groundwater Discharge

Fresh groundwater and brine withdrawals reduce the total amount of groundwater discharge in all systems. However, the relative magnitude of those changes depends on where the withdrawals occur and the type of fluid withdrawn.

In all three systems, upgradient fresh groundwater withdrawals cause the greatest decrease in groundwater discharge. There is a linear, 1:1 relationship between fresh groundwater withdrawals and change in groundwater discharge (Figure 23), meaning that for any amount of fresh groundwater withdrawn, total groundwater discharge will decrease by the same amount. The same cannot be said for halite brine withdrawals, which have an impact on total groundwater discharge that is buffered by flow dynamics in the system. In these salar systems, the majority of groundwater discharge occurs as fresh discharge, which is hydraulically upgradient of the halite brine. In addition, the density-driven flow dynamics of the brine-freshwater interface effectively forms a flow barrier which makes it difficult for changes in flow on one side of the barrier to affect flow on the other side. As a result, halite brine withdrawals demonstrate an exponential decay relationship with total groundwater discharge. Where transitional brine withdrawals are simulated we see a hybrid relationship, with the transitional brine withdrawals causing fresh groundwater level declines due to their position beneath the fresh groundwater aquifer and leading to more significant reductions in total discharge relative to halite brine withdrawals.

Fresh groundwater withdrawals and transitional brine withdrawals have the greatest impact on fresh discharge across all three systems. Halite and transitional brine withdrawals have the greatest impact on brine discharge across all three systems. These relationships result from the flow dynamics described above in which the brine-freshwater interface buffers withdrawal effects on the opposite side of the interface. Brackish discharge is less affected by either type of withdrawal, either increasing somewhat or decreasing somewhat. Migration of the brine-freshwater interface and the position of the water table relative to the ground surface primarily affect whether brackish discharge slightly increases or decreases. When the brinefreshwater interface migrates towards the nucleus, brackish discharge tends to increase; however, the salt concentration of this discharge also decreases (freshens). In addition, when brackish discharge increases, it is always accompanied by a much larger decrease in the upgradient fresh discharge, which also supplies water by overland or near-surface flow to any wetlands that may be located in the brackish discharge area (e.g. Lagunas Punta and Brava), resulting in a net decrease in recharge to those wetlands.

In all three systems, upgradient fresh groundwater withdrawals cause the greatest decrease in groundwater discharge.



**Figure 23.** Summary of the impacts of withdrawal rate on  $\Delta Q$  for the Salar de Atacama, Salar del Hombre Muerto Eastern Subbasin, and Salar del Hombre Muerto Western Subbasin models. Dashed lines indicate the best fit line for the  $\Delta Q$  values across the three models. Solid filled areas represent the range of all values across the three models.

### **Response Time**

The rate of exponential change (k) describes how quickly groundwater discharge responds to fresh groundwater or brine withdrawals. **Figure 24** shows the rate of change constant for the highest withdrawal rate (40% of fresh groundwater recharge) scenarios for each of the three models. The magnitude of k is specific to each of the groundwater model domains due to their unique hydrogeologic conditions. However, across all three systems, groundwater discharge responds faster to fresh groundwater withdrawals than halite brine withdrawals. Transitional brine withdrawals also cause a faster groundwater discharge response than halite brine withdrawals.



**Figure 24.** Summary of  $\Delta Q$  response times for the Salar de Atacama, Salar del Hombre Muerto Eastern Subbasin, and Salar del Hombre Muerto Western Subbasin models for the maximum withdrawal (40% of fresh groundwater recharge) simulations.

### Brine-Freshwater Interface

Fresh groundwater and brine withdrawals change the relative proportions of fresh groundwater and brine within the system, and the brine-freshwater interface responds according to these changes. In general, when fresh groundwater withdrawals occur, the brine-freshwater interface becomes shallower, increasing in elevation at the upgradient boundary of the model domain. When brine withdrawals occur, the brine-freshwater interface widens with the majority of interface migration occurring on the brine limit of the interface, reflecting a decrease in the total salt mass in the brine body (**Figure 22**).

In addition, the water table surface expression of the brine-freshwater interface plays an important role in evapoconcentration and the subsequent formation of new brine. More evapoconcentration occurs where the water table surface expression of the brine-freshwater interface is wider, such as the Salar del Hombre Muerto Eastern Subbasin. This evapoconcentration makes the water table surface expression of the brine-freshwater interface resistant to change because evapoconcentration is buffering changes in salt concentrations in this region. The effects of withdrawals must exceed this evapoconcentration threshold for the water table surface expression of the brine-freshwater interface to migrate. At Salar de Atacama and the Salar del Hombre Muerto Western Subbasin, where the water table surface expression of the brine-freshwater interface is relatively narrow, halite brine withdrawals exceed the evapoconcentration threshold at rates of 20% of fresh groundwater recharge (**Figures 9 and 22**). However, at the Salar del Hombre Muerto Eastern Subbasin, When brine withdrawals occur, the brinefreshwater interface widens where the water table surface expression of the brine-freshwater interface is relatively wide, withdrawals never exceed the evapoconcentration threshold, resulting in very little interface migration (Figure 16).

### Implications and Recommendations

We present the following implications and recommendations based on the results and conclusions from this modeling study.

- Fresh groundwater and transitional brine withdrawals have a larger impact on total GW discharge than halite brine extractions.
  - **Implication:** Emphasis should be placed on minimizing fresh groundwater and transitional brine withdrawals. Avoid development of lithium mining operations in transitional brine environments. Minimize at all costs fresh groundwater withdrawals.
  - **Implication:** Focus environmental monitoring on innovative ways to measure spring discharge through remote sensing and *in situ* monitoring.
- Extent of shallow depth to water is a key factor in controlling sensitivity of impacts on the brine-freshwater interface.
  - **Implications:** Monitoring of salinity (and chemical composition) is more important where depths to water are greater than average.
- In general, fresh groundwater or brine withdrawals have little impact on brackish discharge and reduce fresh discharge.
  - **Implication:** Total amount of inflow to wetlands from groundwater will decrease and impact the salinity distribution of wetlands. Monitoring of salinity is key to understanding the impacts.
- Timescales of impacts in general are longer for halite brine withdrawals and shorter for fresh groundwater and transitional brine withdrawals.
  - **Implication:** Monitoring strategies need to be long-term and consistent with high QA/QC standards.

Environmental Impact Metric	Result	Li Brine Development Implications
Water Quantity	<ol> <li>Impacts from freshwater and transitional brine withdrawals are greater than brine withdrawals</li> </ol>	Focus on minimizing fresh groundwater withdrawals and avoid development in transitional brine environments. Focus on environmental monitoring of spring discharge
	<ol> <li>Timescales of impacts are longer from halite brine withdrawals than fresh groundwater or transitional brines</li> </ol>	Monitoring strategies require a long-term and consistent commitment with high QA/QC standards
Water Quality	3. Freshwater extraction has little impact on brackish water discharge but will lead to decreases of flow to wetlands and increasing salinity	Monitoring of spatial and temporal distributions of salinity is key to quantifying impacts
	<ol> <li>Extent of shallow water table is key control on sensitivity of impacts on brine-freshwater interface</li> </ol>	Monitoring of salinity is more important where depths to water are greater than average

### References

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# Appendix

Supplemental data tables.

Parameter	Value	Unit
Domain Length	24	km
Domain thickness (b)	~360	m
Longitudinal dispersivity (aL)	10	m
Horizontal transverse dispersivity ( $\alpha_H$ )	1	m
Vertical transverse dispersivity ( $\alpha_v$ )	0.01	m
Diffusion coefficient	1x10 <sup>-6</sup>	$m^2 \cdot s^{-1}$
Effective porosity (φ)	0.3	-
Storativity (S <sub>s</sub> )	1x10 <sup>-4</sup>	$m^{-1}$
Vertical anisotropy (Kh/Kv)	100	-
General head elevation	2,302.33	m
Freshwater density (ρ <sub>0</sub> )	1,000	kg·m <sup>-3</sup>
Brine density (ρ <sub>max</sub> )	1,200	kg·m <sup>-3</sup>

### Table S1a. Salar de Atacama Hydrogeologic Model Parameters

 Table S1b.
 Salar de Atacama Hydrogeologic Unit Parameters

Hydrogeologic Unit	Specific Yield (Sy)	Horizontal Hydraulic Conductivity (K <sub>h</sub> ) (m/d)
Alluvium & Clay	0.02	5 - 100
Carbonate	0.21	3 - 100
Gypsum	$1.8 \times 10^{-3} - 0.46$	1-44.5
Halite	$1.8 \times 10^{-3} - 0.46$	1 - 1,000
Ignimbrite	0.02	5 - 50
Silt	0.02	5
Ash	0.02	1
Undifferentiated Bedrock	1.8x10 <sup>-3</sup>	1

Parameter	Value	Unit
Domain Length	20	km
Domain thickness (b)	~170	m
Longitudinal dispersivity (aL)	10	m
Horizontal transverse dispersivity ( $\alpha_H$ )	1	m
Vertical transverse dispersivity ( $\alpha_v$ )	0.01	m
Diffusion coefficient	1x10 <sup>-6</sup>	$m^2 \cdot s^{-1}$
Effective porosity (φ)	0.3	-
Storativity (S <sub>s</sub> )	1x10 <sup>-4</sup>	$m^{-1}$
Vertical anisotropy (K <sub>h</sub> /K <sub>v</sub> )	10	-
General head elevation	3,967	m
Freshwater density (ρ <sub>0</sub> )	1,000	kg·m <sup>-3</sup>
Brine density (ρ <sub>max</sub> )	1,200	kg·m <sup>-3</sup>

#### Table S2a. Salar del Hombre Muerto Eastern Subbasin Hydrogeologic Model Parameters

#### Table S2b. Salar del Hombre Muerto Eastern Subbasin Hydrogeologic Unit Parameters

Hydrogeologic Unit	Specific Yield (Sy)	Horizontal Hydraulic Conductivity (K <sub>h</sub> ) (m/d)
Gravel and Sand	0.02	60
Fine Sand With Silt	0.25	1
Red Clay and Silt	0.008 - 0.01	0.1 - 0.6
Calcareous Gray Silty Clay	0.01	0.6
Black Calcareous Clay	0.01	0.6
Halite	0.03	2.1-15.4
Gypsum	0.1	0.1 - 0.95
Gypsum with Carbonate Lenses	0.13	6
Ignimbrite	0.065	7
Sand and Gravel with Clay	0.01	1
Catal Formation	0.008	0.1

Parameter	Value	Unit
Domain Length	20.9	km
Domain thickness (b)	~224	m
Longitudinal dispersivity (aL)	100	m
Horizontal transverse dispersivity ( $\alpha_H$ )	10	m
Vertical transverse dispersivity ( $\alpha_v$ )	0.1	m
Diffusion coefficient	$1 x 10^{-6}$	$m^2 \cdot s^{-1}$
Effective porosity (φ)	0.3	-
Storativity (S <sub>s</sub> )	1x10 <sup>-3</sup>	$m^{-1}$
Vertical anisotropy (K <sub>h</sub> /K <sub>v</sub> )	10	-
General head elevation	3,967.2	m
Freshwater density (ρ <sub>0</sub> )	1,000	kg·m <sup>-3</sup>
Brine density (ρ <sub>max</sub> )	1,200	kg·m <sup>-3</sup>

#### Table S3a. Salar del Hombre Muerto Western Subbasin Hydrogeologic Model Parameters

### Table S3b. Salar del Hombre Muerto Western Subbasin Hydrogeologic Unit Parameters

Hydrogeologic Unit	Specific Yield (Sy)	Horizontal Hydraulic Conductivity (K <sub>h</sub> ) (m/d)
Gravel and Sand	0.01	2 - 10.6
Fine Sand With Silt	0.25	1.6 – 9.9
Nucleus Halite	0.25	1.6 - 1,000
Halite with Clastics	0.01	1 – 17.5
Ignimbrite	0.05	2
Basement	0.001	1