

1 **Imbalance in the modern hydrologic budget of topographic**  
2 **catchments along the western slope of the Andes (21–25°S)**

3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29

David F. Boutt<sup>1</sup>, Lilly G. Coenthall<sup>1</sup>, LeeAnn Munk<sup>2</sup>, Scott A. Hynek<sup>3</sup>,

<sup>1</sup> Department of Geosciences, University of Massachusetts-Amherst, Amherst, MA, USA

<sup>2</sup> Department of Geological Sciences, University of Alaska-Anchorage, Anchorage, AK, USA

<sup>3</sup> Earth and Environmental Systems Institute and Department of Geosciences, Pennsylvania State University, University Park, PA, USA

Mailing Address:

Department of Geosciences  
611 North Pleasant Street  
233 Morrill Science Center  
University of Massachusetts  
Amherst, MA 01003-9297

30 **Key Points**

- 31 • Modern hydrologic budgets in topographic watersheds along the western margin of the  
32 Andes (21–25°S) do not close.
- 33 • Steady state regional flow from outside these basins yield large contributing areas of  
34 conflicting nature.
- 35 • Transient groundwater storage is essential to balance water budget and is consistent with  
36 paleohydrologic observations.

37

38 **Keywords:** Salar de Atacama, Chile, recharge, regional groundwater flow, plateau, paleo-  
39 recharge, pulsed recharge, recharge events

40

41 **Abstract**

42 Rates of water discharge often exceed groundwater recharge in arid catchments. This  
43 apparent mass imbalance within a catchment may be reconciled through either regional-scale  
44 groundwater flow between topographic drainages and/or the draining of stored groundwater  
45 recharged during pluvial periods. We investigate discrepancies in the modern hydrologic budget  
46 of catchments along the west flank of the Andes in northern Chile (21–25° S), focused on the  
47 endorheic Salar de Atacama basin, and adjacent basins. Our new, uncertainty bounded, estimates  
48 of modern recharge rates do not come close to balancing observed modern groundwater  
49 discharge within topographic catchments. Two geologically realistic conceptualizations of  
50 hydrogeologic catchments discharging to Salar de Atacama were explored with a 2D  
51 groundwater model. Results from models support the interpretation that both regional flow and  
52 transient drainage of groundwater from storage are required to balance water budgets along the  
53 plateau margin. The models further examine whether this system is still responding to climatic  
54 forcing from pluvial periods and highlight general characteristics for similar plateau margin  
55 systems including: (1) water level changes at the plateau margin are highly sensitive to changes  
56 in recharge on the plateau, (2) extent and magnitude of the changes in water table are controlled  
57 by the distribution of hydraulic conductivity at the margin, (3) contributing area to the lower  
58 elevation catchment is itself dynamic, and not coincident with the topographic boundary, and (4)



59 difficulty in reconciling the modern position of “the water table” on the Andean plateau with the  
60 regional groundwater flow conceptualization and modern discharge to low lying catchments.

61

## 62 **1. Introduction**

63 Rates of anthropogenic water extraction and natural water discharge often exceed  
64 groundwater recharge in arid catchments [e.g. *van Beek et al.*, 2011; *Gleeson et al.*, 2012]. This  
65 mass imbalance within a catchment may be reconciled by regional-scale groundwater flow  
66 between topographic drainages and/or the draining of stored groundwater recharged during past  
67 pluvial periods. While these processes are well documented globally [e.g. *Alley et al.*, 2002;  
68 *Gleeson et al.*, 2011; *Condon and Maxwell*, 2015], debates exist over methods to physically and  
69 quantitatively distinguish between these mechanisms since both depend on processes operating  
70 on large spatial and temporal scales difficult to directly observe [e.g. *Nelson et al.*, 2004;  
71 *Masbruch et al.*, 2016; *Nelson and Mayo*, 2014]. Both mechanisms invalidate the steady state  
72 assumption often targeted for water resource management [e.g. Salar de Atacama (SdA) in  
73 *Dirección General de Aguas*, 2013; *Gorelick and Zheng*, 2015; *Currell et al.*, 2016]; this  
74 assumption also underpins lake-based paleo-precipitation reconstructions [e.g. *Urbano et al.*,  
75 2004]. Therefore, we aim to better constrain the spatial and temporal dimensions of regional-  
76 scale groundwater flow and transient draining of groundwater storage at a site where extreme  
77 modern hydrologic imbalance is documented in the topographic watershed.

78 Plateau margins, especially in arid regions, are characterized by steep gradients in  
79 topography and climate that are conducive to the development of regional-scale groundwater  
80 systems [*Haitjema and Mitchell-Bruker*, 2005; *Gleeson et al.*, 2011]. Closed basins within or  
81 adjacent to plateau may preserve geologic records of water fluxes over  $10^2$ – $10^6$  year time frames  
82 in the accumulation of evaporite minerals [e.g. *Godfrey et al.*, 2003; *Jordan et al.*, 2007; *Munk et*  
83 *al.*, 2018]. The Salar de Atacama (SdA) hosts  $>1800$  km<sup>3</sup> of halite in a closed basin adjacent to  
84 the Altiplano-Puna plateau, and provides an extreme case to evaluate the potential role of  
85 regional-scale groundwater flow and transient draining of groundwater storage in sustaining  
86 water discharge rates over both modern and geologic timescales (Figure 1).

87           Sustaining the accumulation of massive (>1500 m thick [*Jordan et al.*, 2007]) evaporites  
88 in the basin necessitates maintaining the water table within several meters of land surface over a  
89 5–10 million year time period [*Tyler et al.*, 2006]. Individual components of the water budget of  
90 the relatively small and hyperarid topographic watershed of SdA, and of the adjacent Altiplano-  
91 Puna plateau [*Kampf and Tyler*, 2006; *Salas et al.*, 2010] have been studied extensively. While  
92 evidence for modern recharge in the central Andes exists [*Houston*, 2007, 2009]; rates, spatial  
93 extent, and mechanisms are poorly constrained [e.g. *Montgomery et al.*, 2003; *Jordan et al.*,  
94 2015; *Rissmann et al.*, 2015]. Halite accumulation, a proxy for long term average water inflow,  
95 confirms the observed hydrologic imbalance over geologically meaningful timescales [up to 10  
96 Myr; *Corenthal et al.*, 2016]. However, recently Munk et al. (2018) presented solute fluxes for  
97 the surface and shallow sub-surface that account for halite and brine hosted solutes in the  
98 uppermost (30 m) on a Myr timescale. Regional scale groundwater flow, interbasin transfer,  
99 and pulsed recharge events (10–100 year timescale) are documented [*Houston*, 2006b; *Rissmann*  
100 *et al.*, 2015] in the region, and modern discharge in the Atacama Desert is suggested to reflect  
101 the draining of groundwater recharged during episodic pluvial periods ( $10^3$ – $10^4$  year timescale)  
102 in the Late Pleistocene and Holocene [*Houston and Hart*, 2004; *Gayo et al.*, 2012].

103           Previous efforts to balance the water budget of SdA [*Dirección General de Aguas*, 2013;  
104 *Rissmann et al.*, 2015; *Corenthal et al.*, 2016], nearby closed basins [*Houston and Hart*, 2004],  
105 and plateau margins in general [e.g. *Andermann et al.*, 2012] identified large uncertainties in  
106 water flux estimates. Here, recharge from precipitation is quantified by scaling point  
107 measurements to regional satellite datasets including uncertainty, further defining the magnitude  
108 and time-scale of the hydrologic imbalance inferred from observations of halite accumulation in  
109 SdA [*Corenthal et al.*, 2016]. Unique datasets are integrated to: (1) quantify the area of a  
110 regional-scale groundwater catchment necessary to balance modern discharge, and (2)  
111 approximate the role of late-Pleistocene recharge in modern discharge, and (3) elucidate the  
112 mechanisms and process by which water is delivered to the basin. These data are then used to  
113 constrain a 2D groundwater model that further explores the dynamic temporal and spatial scales  
114 of both regional groundwater flow and transient drainage in two  
115 hydrogeological conceptualizations. The focused nature of groundwater discharge allows re-

116 assessment of the contributing areas of plateau margins, and steady state water management  
117 strategies that may apply to similar arid, high-relief regions globally.

### 118 **1.1 Regional Geologic and Hydrologic Framework**

119 Salar de Atacama (SdA), a significant topographic depression with an area over 17,000  
120 km<sup>2</sup> adjacent to the Altiplano-Puna plateau of the Central Andes, serves as the focal point for our  
121 analysis of the hydrologic imbalance in the region (Figure 1). SdA began accumulating a  
122 massive halite deposit ~7 Ma, coincident with uplift of the Central Andean Plateau [*Jordan et*  
123 *al.*, 2002a, 2007; *Reutter et al.*, 2006]. The halite nucleus of the basin hosts a lithium-rich brine  
124 that provides approximately one-third of the global lithium supply [*Maxwell*, 2014]. Alluvial  
125 fans conduct water to SdA, and numerous springs and seeps discharge in the transition zone  
126 around the halite nucleus, supporting environmentally sensitive wetlands and lagoons. The  
127 spatial trend of alluvium, carbonate, gypsum and halite downgradient along flowpaths through  
128 the transition zone documents the evaporation of inflow water until it reaches halite saturation  
129 [*Risacher et al.*, 2003]. Seven perennial and ephemeral streams emerge at stratigraphic and  
130 structural contacts but lose all surface flow through alluvium before reaching gypsum and halite  
131 facies, however, shallow groundwater again emerges in complex lagoon systems above a  
132 freshwater/brine interface that rims the salar margin. A series of hydrogeologically important  
133 Plio-Pleistocene ignimbrites originate from the Altiplano-Puna Volcanic Complex on the plateau  
134 [e.g. *Jordan et al.*, 2007; *Salisbury et al.*, 2011] and extend into the SdA subsurface; in the  
135 northern half of the basin these units are interpreted to be highly continuous. The north-south  
136 trending blind, high-angle, down-to-the-east Salar Fault System accommodates over 1 km of  
137 offset through the halite nucleus [*Lowenstein et al.*, 2003; *Jordan et al.*, 2007; *Rubilar et al.*,  
138 2017; *Martinez et al.*, 2018]. *Jordan et al.* [2002] suggest that this fault acts as a barrier causing  
139 orogenic scale groundwater flow paths to discharge in SdA.

140 There are many studies that either explicitly document [*Magaritz et al.*, 1990;  
141 *Montgomery et al.*, 2003; *Rissmann et al.*, 2015; *Jayne et al.*, 2016] or implicate [*Pérez-Fodich*  
142 *et al.*, 2014; *Jordan et al.*, 2002a, 2002b] that water from the Andean Cordillera via regional  
143 groundwater flow feeds downgradient basins to the west. In their analysis on the MNT aquifer,  
144 *Rissman et al.* [2015] provide one of the few examples in the region where hydrologic and

145 geochemical information have been used to investigate and imply a connection from a high  
146 elevation recharge area southeast of SdA to discharge at the salar margin. The  $^{87}\text{Sr}/^{86}\text{Sr}$  values  
147 for discharging water reported by Munk et al. [2018] support the possible link between high  
148 elevation salt lakes and brines in SdA proposed by Rissman et al. [2015], and consistent with the  
149 observation of Grosjean et al. [1995] that many of the high elevation lakes never reach Na-Cl  
150 saturation and that the water enriched in Na and Cl then drains to lower elevation basins.  
151 Investigation and refinement of the hydrogeologic framework for many such aquifers is required  
152 to understand the regional scale imbalances in water budgets, and we embark on broad scale  
153 characterization of such systems in this work.

154 The SdA basin, in the core of Atacama Desert, is characterized by a hyperarid to arid climate  
155 [Hartley and Chong, 2002]. Significant inter-annual precipitation variability [Garreaud et al.,  
156 2003] includes infrequent high-intensity rainfall events that produce pulsed groundwater  
157 recharge [Houston, 2006b; Boutt et al., 2016]. Because the basin has been closed since at least  
158 the late Miocene [Jordan et al., 2002a], surface water discharge occurs only through  
159 evapotranspiration. Sedimentary records suggest that variable arid to hyperarid climates have  
160 dominated since 53 ka [Bobst et al., 2001; Godfrey et al., 2003], with at least four periods wetter  
161 than modern occurring since 106 ka [Gayo et al., 2012]. Hydrologic models for lakes in the  
162 Bolivian Altiplano similarly suggest that precipitation may have been 2–3 times more than the  
163 modern during at least four intervals in the previous 130 ka, with the most recent wet period  
164 occurring during the Tauca (Heinrich 1) wet phase in the late Pleistocene [Placzek et al., 2013].  
165 SdA sedimentary records document variations in the hydrologic balance of SdA from >100 ka to  
166 present [Bobst et al., 2001; Godfrey et al., 2003; Lowenstein et al., 2003]. Paleo-wetland deposits  
167 south of SdA suggest two wetter periods from 15.9–13.8 ka and 12.7–9.7 ka, (Central Andean  
168 Pluvial Event) [Quade et al., 2008] consistent with vegetation records and wetland deposits  
169 throughout the region [Betancourt et al., 2000; Rech et al., 2002, 2003; Quade et al., 2008].  
170 Climate around SdA has been drier since the mid-Holocene based on the water table being below  
171 ground surface at paleo-wetland sites and observations from sediment cores at SdA [Rech et al.,  
172 2002; Quade et al., 2008; Placzek et al., 2013]. The most recent period from 3.0 ka to present  
173 has been the driest since the late Pleistocene [e.g. Betancourt et al., 2000].

174 **2. Study Approach, Data Sources, and Methods**

175 *2.1 Conceptualization of the Modern Water Budget*

176 Constraining the modern hydrologic budget is critical to evaluating whether the system is  
 177 balanced within the topographic watershed. If the system is at steady state within the topographic  
 178 watershed, groundwater recharge from precipitation ( $GW_{RCH}$ ) plus surface water runoff (R)  
 179 would balance all evapotranspiration (discharge) from SdA ( $D_{SdA}$ ) with no change in storage (S).  
 180 When considering the water budget beyond the topographic watershed one must also consider an  
 181 additional loss term of evapotranspiration from salars and lakes in high elevation closed basins  
 182 ( $D_{HighElevSalars}$ ). The most conservative (more balanced) conceptualization of the modern  
 183 hydrologic balance can be described by:

184 
$$\Delta S = GW_{RCH} + R - D_{SdA} - D_{HighElevSalars} \quad (1)$$

185 In the context of this equation, we provide uncertainty-bounded estimates of spatially distributed  
 186  $GW_{RCH}$  and  $D_{HighElevSalars}$  throughout the region as the critically under-constrained term for  
 187 assessing the hydrologic balance. A negative change in storage would suggest that water from  
 188 outside the topographic basin or drawn from storage is needed to close the modern budget,  
 189 whereas a positive change in storage would reflect recharge and surface water inputs currently  
 190 outpacing evapotranspiration. We evaluate equation (1) for both the topographic watershed and  
 191 the hydrogeologic watershed. We define the hydrogeologic watershed as the smallest potential  
 192 contributing area within which the steady state hydrologic budget closes within reasonable  
 193 uncertainty bounds (i.e. scenario M in *Corenthal et al.*, [2016]). This conservative scenario has  
 194 the potential to double count some discharge in both the  $GW_{RCH}$  and R terms.

195 A less conservative (less balanced) water budget conceptualization assumes that baseflow  
 196 in streams within the SdA watershed is sourced entirely from groundwater. In this  
 197 conceptualization precipitation events recharge aquifers ( $GW_{RCH}$ ), but do not generate runoff  
 198 (R). This conceptualization of the modern hydrologic balance can be described by:

199 
$$\Delta S = GW_{RCH} - D_{SdA} - D_{HighElevSalars} \quad (2)$$

200 Equation 2 does not include a surface water runoff term (R) and therefore yields a more negative  
 201 estimate of change in groundwater storage. A more negative change in storage would suggest  
 202 that even more water from outside the topographic watershed or drawn from storage is needed to

203 close the modern water budget. These equations and the following budgets do not consider  
204 anthropogenic water extraction.

## 205 2.2 Precipitation

206 Precipitation estimates were obtained from the publicly-available Tropical Rainfall  
207 Measurement Mission (TRMM) 2B31 dataset of Mean Annual Precipitation (MAP) derived  
208 from 1–3 daily measurements at a resolution of 25 km<sup>2</sup>. A processed TRMM 2B31 dataset was  
209 calibrated, validated, and provided by *Bookhagen and Strecker* [2008] over the period January 1,  
210 1998 to December 31, 2009. This dataset was compared to gage measurements from 28  
211 meteorological stations in the Region of Antofagasta maintained by the Chilean Dirección  
212 General de Aguas (DGA) and one station on the salar maintained by the Sociedad Chilena de  
213 Litio/Rockwood Lithium Inc./Albemarle (Figure S1 and S2). Power-law functions are fit to the  
214 lower and upper bound of the DGA station-TRMM data (Figure S2) to provide constraints on  
215 bias and uncertainty in the precipitation estimates. These bounds are used to estimate the median  
216 (most plausible), lower and upper ranges of MAP in the region and to provide a range of possible  
217 precipitation scenarios (Text S1). These ranges are incorporated in other dependent calculations  
218 below.

## 219 2.3 Groundwater Recharge

220 To determine  $GW_{RCH}$  from precipitation (P), we apply the chloride mass balance (CMB)  
221 method, which has been successfully applied to basins to the north and northwest of SdA  
222 [*Houston, 2007, 2009*], whereby

$$223 \quad GW_{RCH} = \frac{P * Cl_p}{Cl_{gw} - Cl_{rw}} \quad (3)$$

224 Where:

225  $Cl_p$  = chloride concentration in precipitation

226  $Cl_{gw}$  = chloride concentration in groundwater

227  $Cl_{rw}$  = chloride contribution to groundwater from rock weathering

228 Common assumptions used in the application of the CMB method include (1) precipitation (P) is  
229 the only source of chloride (Cl) to groundwater, and (2) Cl is conservative in the groundwater  
230 system [*Bazuhair and Wood, 1996*]. Table 1 presents analyses of precipitation samples and  
231 locations.

232 We apply equation (3) to selected sample sites (Text S2) using the median, lower, and  
233 upper TRMM 2B31 derived P estimates [Bookhagen and Strecker, 2008] to determine a range of  
234 potential  $GW_{RCH}$  rates. A power function (Figure 3a) fit to the P and calculated  $GW_{RCH}$  is  
235 applied to the TRMM 2B31 datasets to generate lower, median, and upper estimate of the  
236 fraction of P that becomes  $GW_{RCH}$ . Estimates of  $GW_{RCH}$  were confined to areas that do not  
237 contain permanent discharge features (salar and lakes). The fraction of P that does not recharge  
238 groundwater is assumed to evapo-transpire or contribute to R. Herein, we pursue a conservative  
239 approach for closing the water budget and consider that R is generated by precipitation runoff  
240 (equation 1).

#### 241 *2.4 Evapotranspiration*

242 Evapotranspiration ( $D_{SdA}$ ) estimates from the nucleus and transition zone of SdA are  
243 summarized from works that (1) coupled eddy covariance station measurements taken in 2001 to  
244 remotely sensed land energy budgets ( $D_{SdA}$  range from 1.6–27.1  $m^3/s$ ) [Kampf and Tyler, 2006]  
245 and (2) coupled lysimeter measurements collected from 1983–1985 to land-type classifications  
246 from 1983–1985 ( $D_{SdA}$  of 5.6  $m^3/s$ ) [Mardones, 1986]. Of this range, we consider a maximum  
247  $D_{SdA}$  of 22.7  $m^3/s$  because higher estimates significantly over-predict fluxes from the nucleus  
248 [Kampf and Tyler, 2006]. We consider a minimum  $D_{SdA}$  of 5.6  $m^3/s$  because it is the current  
249 estimate used to manage water resources of the basin [Dirección General de Aguas, 2013]. The  
250 infiltration rate determined through the CMB method is assumed to account for diffuse ET from  
251 anywhere not covered by a salar or lake.

252 Many closed basins above 3500 m in elevation host zones of focused evapotranspiration  
253 ( $D_{HighElevSalar}$ ). Because no reliable  $D_{HighElevSalar}$  measurements were available, a linear regression  
254 for potential ET (PET) (mm/year) as a function of ground elevation (m) for the Atacama region  
255 was used (Text S2).

#### 256 *2.5 Incorporation of Uncertainty in Hydrologic Balance Estimates*

257 Each component of the water balance contains uncertainty that propagates through the  
258 calculations described above to consider a range of possible hydrologic balance estimates. At  
259 each stage of the calculations we consider these uncertainties and include them in final lower,  
260 median, and upper-end recharge scenarios which then are used to assess closure of the water

261 budget. The precipitation amounts from TRMM impact both the precipitation-recharge CMB  
262 functional relationship ( $P$  in equation 3) as well as the assessed distributed recharge calculations.  
263 Additionally, uncertainty in the chloride composition ( $Cl_p$  in Equation 3) of the precipitation also  
264 impacts the effective recharge through the CMB calculation and functional relationship (equation  
265 3). Our lower-end recharge estimates are calculated using the lowest possible precipitation  
266 estimates (Figure S2 – Upper Curve), the lowest precipitation chloride concentration, and  
267 omitting any  $Cl^-$  in groundwater sourced from rock weathering ( $Cl_{rw}$ ). The median recharge  
268 estimate is produced using the TRMM 2B31 directly with the average chloride concentration in  
269 precipitation. Finally, the upper-end recharge estimates are calculated using highest possible  
270 precipitation estimates (Figure S2 – Lower Curve), the highest precipitation chloride  
271 concentration, and the possibility of  $Cl^-$  sourced from rock weathering.

### 272 3. Assessment and Uncertainty in the Hydrologic Balance

#### 273 3.1 Precipitation

274 On the SdA salt flat annual precipitation averages 16 mm/year [*Sociedad Chilena de Litio*  
275 *Ltda.*, 2009], whereas  $>300$  mm/year [*Bookhagen and Strecker*, 2008; *Quade et al.*, 2008] may  
276 occur above 5,000 m within the topographic watershed (Figure 1, Figure 3b). Approximately 50–  
277 80 mm/year of snow water equivalent occurs at 4500 m asl [*Vuille and Ammann*, 1997] but the  
278 majority likely sublimates before infiltrating [*Johnson et al.*, 2010; *Dirección General de Aguas*,  
279 2013]. Based on the TRMM 2B31 dataset mean annual precipitation from 1998 to 2009,  
280 including the wetter than average 2001/2002, is  $30.7 \text{ m}^3/\text{s}$  ( $23.4$  for lower bound and  $51.7 \text{ m}^3/\text{s}$   
281 for upper bound) in recharge zones in the topographic watershed (Table 2), equivalent to a mean  
282 of 48 mm/year with a range of 0–340 mm/year (standard deviation of 45 mm/year). For the  
283 median precipitation scenario only 7% of the watershed area receives more than the 120  
284 mm/year of precipitation threshold required for significant  $GW_{RCH}$  [*Scanlon et al.*, 2006;  
285 *Houston*, 2009] (Figure 3, Figure 4a), and most precipitation occurs above 3,500 m. Using this  
286 (median) scenario, infiltration rates of nearly 100% throughout the topographic watershed would  
287 be required to balance the highest estimates of  $D_{SdA}$ .



288 *3.2 Groundwater Recharge*

289 Chloride concentration in precipitation samples ranged from 5–16 mg/L (Table 1). We  
 290 combine our CMB results with those from the Turi and Linzor basins [Houston, 2007, 2009] to  
 291 establish a new relationship for  $GW_{RCH}$  as a function of P (Figure 3) in this region. The range of  
 292 chloride concentrations in precipitation bounds does not impact the fit of this function to the  
 293 data, therefore we use a single relationship between precipitation and recharge (Text S3 and  
 294 Figure S5). This relationship is fit using a power law with an  $R^2$  of 0.82 as described:

$$295 \quad GW_{RCH} = (1.3 * 10^{-4}) * P^{2.3} \quad (4)$$

296 Applying equation (4) to the median TRMM 2B31 dataset predicts 1.1 m<sup>3</sup>/s (1.1 for lower bound  
 297 and 2.1 m<sup>3</sup>/s for upper bound) of recharge within the topographic watershed (Figure 4b, Table 2),  
 298 with infiltration rates ranging from 0.5–3.5% based on a precipitation Cl<sup>-</sup> concentration of 8  
 299 mg/l, or 0.3–9.0% considering a range of precipitation Cl<sup>-</sup> concentrations from 5–16 mg/l (Table  
 300 3, Figure 3). Similar to arid regions globally [Scanlon *et al.*, 2006] and the Central Andes  
 301 [Houston, 2007], more than 1 mm/year of  $GW_{RCH}$  only occurs when precipitation exceeds 120  
 302 mm/year.

303 *3.3 Evapotranspiration*

304 Estimates of  $D_{SdA}$  range from 5.6–13.4 m<sup>3</sup>/s [Mardones, 1986; Kampf and Tyler, 2006]  
 305 (Table 2). We use a range of  $D_{SdA}$  estimates in our calculations, considering a minimum of 9.5  
 306 m<sup>3</sup>/s based on the spatially variable latent heat flux method [Kampf and Tyler, 2006] and  
 307 lysimeter study [Mardones, 1986]. We predict that  $D_{HighElevSalars}$  in the hydrogeologic watershed  
 308 totals 5.0 m<sup>3</sup>/s (uncertainty range of 1.8–17.8 m<sup>3</sup>/s) (Figure 4c, Table 2).

309 *3.4 Surface Water and Shallow Groundwater Inflows*

310 Approximately 3.19 m<sup>3</sup>/s of shallow subsurface groundwater enters SdA (Corenthal *et al.*  
 311 2016; Munk *et al.*, 2018). The *Direccion General de Aguas* [2013] estimates total streamflow to  
 312 SdA is 1.58 m<sup>3</sup>/s based on gage measurements, which alone matches our range of estimates of  
 313  $GW_{RCH}$  within the topographic watershed (1.1–2.1 m<sup>3</sup>/s). The sum of shallow groundwater and  
 314 streamflow (4.77 m<sup>3</sup>/s) is consistent with but smaller than low estimates of  $D_{SdA}$  (5.6 m<sup>3</sup>/s);  
 315 however,  $GW_{RCH}$  within the topographic watershed accounts for only 24% of these inflows and  
 316 only 5–20% of  $D_{SdA}$ . In order to balance the full range of discharge from evapotranspiration (5.6

317 to 13.4 m<sup>3</sup>/s) with GW<sub>RCH</sub> in the SdA topographic watershed, an average infiltration rate of 21 to  
318 86 % is required. Such rates greatly exceed average infiltration rates for arid regions globally of  
319 0.1–5% [Scanlon *et al.*, 2006] and infiltration rates observed in the Linzor and Turi basins  
320 [Houston, 2007, 2009].

### 321 3.5 Steady State Hydrologic Balance

322 Within the topographic watershed, D<sub>SdA</sub> is 2–8 times (5.6–13.4 m<sup>3</sup>/s) higher than the  
323 combined inputs of modern recharge from precipitation and streamflow (Figure 5; Table 2).  
324 Some streamflow is likely sourced from groundwater [e.g. Hoke *et al.*, 2004] and therefore  
325 counted twice, yielding more conservative estimates of hydrologic imbalance. The  
326 hydrogeologic watershed required for GW<sub>RCH+R</sub> to balance evapotranspiration for our estimated  
327 range of recharge values has a surface area over 75,000 km<sup>2</sup>, 4 times larger than the topographic  
328 watershed (Figure 5 scenario M). Even the high estimates of GW<sub>RCH+R</sub> fail to explain the low  
329 estimates of D<sub>SdA</sub> for watersheds A through I.

## 330 4. Closing the hydrologic budget

331 The modern hydrologic balance of the SdA topographic watershed does not close within  
332 reasonable uncertainty bounds (Table 2; Figure 5). We consider a range of uncertainties in the  
333 data including evapotranspiration estimates and bias in the TRMM 2B31 precipitation dataset  
334 correlation with gage data. To close this apparent hydrologic imbalance, the missing water could  
335 be explained by several sources [Corenthal *et al.*, 2016]. Here we explore two mechanisms to  
336 close the budget: (1) a larger watershed area that encompasses regional-scale inter-basin  
337 groundwater flow paths recharged from precipitation at higher elevations and (2) the modern  
338 hydrologic balance includes drainage of transient groundwater storage recharged during wetter  
339 conditions.

340 These two mechanisms are inferred to account for the majority of the missing water flux;  
341 however, additional flowpaths (e.g. orogenic groundwater) and systematic errors in ET and/or  
342 GW<sub>RCH</sub> estimates could also explain portions of the imbalance. Infrequent, high-intensity  
343 precipitation events are known to rapidly recharge the groundwater system in areas where the  
344 water table is near the surface [Boutt *et al.*, 2016]. For the brine budget of SdA, such events are  
345 important to balance discharge from pumping and the low ET rates (<0.1 mm/year) in the halite

346 aquifer. By applying equation 4 uniformly to the precipitation dataset, the potential for higher  
347 recharge rates in salars during intense rainfall is not included in the budget calculations.  
348 Nonetheless, the CMB method integrates over long-term timescales of recharge and accounts for  
349 these events in alluvium elsewhere in the Atacama [*Bazuhair and Wood, 1996; Houston, 2006b*].  
350 Therefore, the CMB equation accounts for different types of freshwater recharge mechanisms

351 *4.1 Steady state hydrologic system with residual water from regional groundwater flow*  
352 *(Mechanism 1)*

353 The arid climate, high topographic relief and presence of laterally continuous permeable  
354 volcanic units dipping towards SdA support the potential for regional groundwater flow paths  
355 [*Tóth, 1963; Haitjema and Mitchell-Bruker, 2005*]. Within the proposed hydrogeologic  
356 watershed of 75,900 km<sup>2</sup> (Table 2; Figure 5 scenario M),  $GW_{RCH} + R$  balance evapotranspiration  
357 while maintaining an overall topographic gradient driving groundwater flow towards SdA;  
358 however, this watershed delineation is non-unique. Proposed recharge areas for many adjacent  
359 watersheds along the western Altiplano-Puna plateau margin overlap (Figure 5). We propose  
360 that regional groundwater flow plays an important role in the modern hydrologic balance of SdA;  
361 however, it likely cannot fully explain the observed discrepancy.

362 Inter-basin groundwater flow is assumed to occur in the central Andes [*Anderson et al.,*  
363 *2002; Jordan et al., 2015*], including the Monturaqui-Negrillar (MNT) aquifer that discharges in  
364 the southern SdA [*Rissmann et al., 2015*]. To explain the existence of giant nitrate deposits in the  
365 Central Depression southwest and northwest of SdA, *Pérez-Fodich et al. [2014]* also suggest  
366 regional groundwater flowpaths. To the north, interbasin groundwater flow is necessary to close  
367 the hydrologic budget of the Río Loa catchment (drainage area = 33,570 km<sup>2</sup>), where the Chilean  
368 DGA estimates a total of 6.4 m<sup>3</sup>/s of water discharge [*Jordan et al., 2015*], but we calculate only  
369 1.6 – 4.0 m<sup>3</sup>/s of  $GW_{RCH}$  within that topographic watershed. Using the plausible groundwater  
370 system of the Río Loa proposed by *Jordan et al. [2015]* (Figure 5); we estimate that  
371 approximately 8.5 – 14.3 m<sup>3</sup>/s of  $GW_{RCH}$  occurs within this zone; however, this boundary  
372 overlaps with the major discharge zone of the Salar de Uyuni as well as a significant portion of  
373 our proposed hydrogeologic watershed for SdA. In order for these adjacent watersheds to have  
374 distinct recharge zones and be hydrologically balanced, it is apparent that some water must be  
375 drawn from transient storage.

376 *4.2 Transient, draining groundwater storage (Mechanism 2)*

377 Here we consider transient draining of groundwater storage to reconcile these budgets.  
378 We calculate the mean residence time of water [Gelhar and Wilson, 1974; Lasaga and Berner,  
379 1998] within the SdA watershed is calculated to be 4.9 kyr using the conservative  $D_{SdA}$  rate of  
380  $5.6 \text{ m}^3/\text{s}$ , an active aquifer thickness of 500 m, an area of  $17,257 \text{ km}^2$  (i.e. area of the topographic  
381 watershed), and an effective porosity of 0.25. The dynamic response time [Houston and Hart,  
382 2004] for the topographic watershed is 9.2 kyr and 42 kyr for the hydrogeologic watershed (Text  
383 S4). In systems with long residence and response times, the assumption that modern recharge  
384 rates must balance discharge rates is invalidated by having equilibration times greater than the  
385 timescale of documented climatic changes [Currell et al., 2016]. High and low elevation  
386 groundwater age estimates lack a significant component of modern recharge further suggesting  
387 that these systems respond over long time scales [Houston, 2006b]. Evaporation at a smaller  
388 salar 50 km southwest of SdA exceeds modern recharge, and this imbalance has been explained  
389 by residual hydraulic head decay (i.e. groundwater storage) due to episodic recharge [Houston  
390 and Hart, 2004]. We now investigate these processes in a physically-based numerical model of  
391 a plateau-margin hydrogeologic system.

392

393 *5 Numerical Simulations of a Plateau Margin Groundwater System*

394 A transient 2-dimensional groundwater model simulating the Altiplano-Puna plateau and  
395 adjacent SdA system was constructed using the MODFLOW finite difference code for saturated  
396 flow [McDonald and Harbaugh, 1988]. The purpose of the model is to examine: (1) the dynamic  
397 response times of a regional groundwater system to changes in groundwater recharge that are of  
398 a magnitude similar to that predicted by paleoclimatic reconstructions [Betancourt et al., 2000;  
399 Placzek et al., 2013]; (2) the sensitivity of water level responses to these changes in groundwater  
400 recharge; and (3) whether the groundwater divide between water draining to SdA and water  
401 discharging to shallow basins on the plateau is dynamic or static with respect to changing  
402 recharge. The framework model is based on previous work in the Atacama by Houston and Hart  
403 [2004] and in the Murray Basin in Australia by Urbano et al., [2004]. We evaluate this model for  
404 two scenarios of hydraulic conductivity that are designed to be conducive and restrictive to  
405 regional groundwater flowpaths; the conducive scenario is intended to approximate the northern

406 half of SdA, and the restrictive scenario is intended to approximate the southern half of SdA.  
407 Together these scenarios represent the range of conditions expected along the plateau margin.  
408 Initial hydraulic head geometries were assigned based on the results of steady-state simulations,  
409 and the transient models simulate a period of 100,000 years (using 100 year timesteps)  
410 immediately following a step decrease in precipitation (recharge).

#### 411 *5.1 Base Model Geometry and Properties*

412 The model domain is 250,000 m long (active domain of 219,200 m), unit width, and  
413 3,000 m thick with grid dimensions of 200 m x 1 m x 200 m in the upper 7 layers and 200 m x 1  
414 m x 400 m in the lower 4 layers. Elevations of the top grid cells were interpolated from a  
415 smoothed ASTER GDEM. The right, bottom and left faces of the model are no-flow boundaries.  
416 On the top boundary, there are 207 constant head cells in the upper layer at an elevation of 2300  
417 m asl, representing the SdA surface. Specified flux boundaries were assigned to all other top  
418 cells. There are 119 drains along the plateau with conductance of 1,000 m<sup>2</sup>/day. Along the  
419 plateau at an elevation of 3,893 m, 28 drains were assigned an elevation of 3,993 m and  
420 conductance of 10 m<sup>2</sup>/day for the steady state run to produce a high elevation lake similar to  
421 those described by *Grosjean et al.* [1995], *Condom et al.*, [2004] and others. For the transient  
422 runs, the drains were assigned an elevation of 3,893m (top cell elevation) and conductance of  
423 1,000 m<sup>2</sup>/day to simulate a salar. Recharge was assigned to any top cell that was not already a  
424 drain or constant head boundary. For the initial steady-state simulations, recharge was  
425 determined by multiplying the TRMM 2B31 precipitation raster by a factor of three and applying  
426 equation 4 to this precipitation raster. This multiplication factor represents the upper (and  
427 conservative) estimates of precipitation during past pluvial periods in the most recent 130 ka  
428 [*Placzek et al.*, 2013]. This resultant raster was then interpolated to the model grid. For the  
429 transient run, modern  $GW_{RCH}$  rates were determined by applying equation 4 for  $GW_{RCH}$  to the  
430 modern TRMM 2B31 dataset and interpolating the resultant raster to the model grid.  
431 Interpolating the 3x modern precipitation and modern precipitation estimates for recharge inputs  
432 to the model captures the spatial distribution of  $GW_{RCH}$  across the plateau as well as the  
433 predicted relative magnitude of paleo- to modern-  $GW_{RCH}$  from the late Pleistocene to present  
434 based on *Betancourt et al.* [2000] and *Placzek et al.* [2013]. There are no other hydraulic sources

435 or sinks in the model and the 2D model does not account for flow transverse to the domain,  
436 surface water features or direct precipitation runoff.

437 Two heterogeneous, isotropic hydraulic conductivity distributions were examined based  
438 on a geologic cross section through the SdA basin and the western Altiplano-Puna plateau by  
439 *Reutter et al.* [2006] (Figure 6). These scenarios are designed to represent: (1) southeastern SdA  
440 characterized by an uplifted, low permeability block of Precambrian to Carboniferous basement  
441 that interrupts the plateau margin (restrictive to regional flow) and (2) northeastern SdA  
442 characterized by monoclinal folding of laterally extensive ignimbrites (conductive to regional  
443 flow). Hydraulic conductivities for the geologic units described in *Reutter et al.* [2006] were  
444 assigned based on standard values from *Ingebritsen and Manning* [1999] and range from 0.01 to  
445 10 m/day. For the transient simulations, a confined specific storage of  $10^{-4}$  was assigned  
446 uniformly to the entire domain.

447

## 448 *5.2 Model Results*

### 449 5.2.1 Water Table Response to Changing Recharge Conditions

450 Water level responses to changing recharge conditions showed strong spatial variability  
451 with the most sensitivity observed in the area of the western and eastern plateau margins (Figure  
452 7). Water levels showed a greater magnitude of response to recharge in the restrictive than in the  
453 conducive to regional flow simulations; however, the pattern of head decline was consistent  
454 between the models. In both simulations, less than 10 m of change in head was observed in cells  
455 within 7 km of a constant head cell at SdA throughout the 100,000 year simulation. Maximum  
456 head decline occurred near observation points 15 and 16, reaching 845 m of decline in the  
457 restrictive simulation and 370 m of decline in the conducive simulation. The magnitude of head  
458 decline increased with increasing elevation along the plateau margin. From west to east across  
459 the plateau, the magnitude of head decline decreased, reaching a minimum of 100 m decline by  
460 the high elevation drain cells for both simulations.

461 Figure 8 presents a comparison between modern and paleo hydraulic head observations  
462 in the conducive and restrictive cases. Observation points (see actual locations on Figure 5 and  
463 projected locations in Figure 7) are placed along corresponding model locations with modern and  
464 paleo- water table elevation estimates (Table S6). The two models are conceptual, and not

465 specifically developed to match field observations, nonetheless, comparing results from the two  
466 models with field observations supports general interpretations and places first-order constraints  
467 on permeability structure of the plateau and plateau margin.

468 The pattern of modeled head declines is consistent with observed patterns inferred from  
469 paleoclimatic studies. During the Central Andean Pluvial Event in the late Pleistocene and early  
470 Holocene, paleowetland deposits and river incision records at elevations <3500 m show water  
471 table fluctuations on the scale of 1–25 m [e.g. *Betancourt et al.*, 2000; *Rech et al.*, 2002; *Quade*  
472 *et al.*, 2008], while lake levels and core records on the Altiplano-Puna plateau at elevations  
473 >3500 m show water table fluctuations of up to 130 m [*Grosjean et al.*, 1995; *Placzek et al.*,  
474 2006, 2013]. In both modeled scenarios, after 10,000 years, the water table on the plateau margin  
475 at elevations <2,600 m declined by <25 m. The water table on the plateau near the discharge  
476 zone at elevations between 3880 m and 4120 m declined approximately 100 m by 10,000 years.  
477 No field observations of changes in water table are available on the plateau in areas distal to  
478 discharge zones.

479

#### 480 5.2.2 Flow Budget Response Time to Changing Recharge

481 The ratio of flow out of the model from constant head cells on the salar surface and  
482 drains on the plateau margin ( $D_{SdA}$ ) to total domain model recharge ( $GW_{TotalRCH}$ ) is used as a  
483 metric to evaluate changes in the flow budget of the model over time. For all time steps of the  
484 100,000 year transient simulation,  $GW_{RCH}$  was assigned modern values. This ratio is plotted as a  
485 function of simulation time in Figure 9. If the ratio of  $D_{SdA}$  to  $GW_{TotalRCH}$  is greater than 1, then  
486  $D_{SdA}$  must be supported in part by draining groundwater storage because the volume of water  
487 entering the salar exceeds total model recharge, where the fraction of discharging water supplied  
488 by draining storage is described by  $(D_{SdA} - GW_{TotalRCH}) / D_{SdA}$ . If  $D_{SdA} / GW_{TotalRCH}$  equals 1, then  
489  $D_{SdA}$  is entirely balanced by  $GW_{TotalRCH}$  in the model domain, and no  $GW_{TotalRCH}$  is discharging  
490 on the plateau. If  $D_{SdA} / GW_{TotalRCH}$  is less than 1, then  $D_{SdA}$  is less than  $GW_{TotalRCH}$  in the model  
491 domain and some fraction of  $GW_{TotalRCH}$  is discharging from drains on the plateau ( $D_{HIGHELEV}$ ).

492 For the restrictive to regional groundwater flow simulation, 60% of  $D_{SdA}$  is sourced from  
493 draining storage at  $t=100$  years, and  $D_{SdA}$  is supplied entirely by  $GW_{TotalRCH}$  after 19,200 years.  
494 For the conducive to regional groundwater flow simulation, 70% of  $D_{SdA}$  is sourced from

495 draining storage at  $t=100$  years, and  $D_{SdA}$  is supplied entirely by  $GW_{TotalRCH}$  after 37,600 years.  
496 In the “conductive” simulation the water table on the plateau lies below the ground surface  
497 elevation and thus the elevation of the drains. For the “restrictive” simulation,  $D_{SdA}/GW_{TotalRCH}$  is  
498 less than 1 for all times greater than 19,200 years. These results suggest that the low permeability  
499 zones must be present between the plateau and the basin floor to allow the high-elevation lakes  
500 and salars to exist.

501 If the change in  $D_{SdA}/GW_{TotalRCH}$  between each time step is small, then the model has  
502 approached a steady state and the modeled system is adjusted to a reduction in groundwater  
503 recharge from 3x modern to modern. For the restrictive simulation, the dynamic response time is  
504 approximately 85,000 years, and approximately 38,000 years for the conducive simulation.  
505 These results are consistent with the dynamic response time calculations using general bulk  
506 aquifer properties of 42 kyr for the hydrogeologic watershed. The modeling results do not  
507 support the modern steady-state budget for systems with long response times.

### 508 5.2.3 Dynamic groundwater divides

509 Model results confirm that the groundwater divide separating water flowing to SdA and  
510 water discharging within the plateau do not coincide with topographic watershed boundaries  
511 (Figure 7 – vertical lines). In the initial steady state simulations for both scenarios, the  
512 groundwater divide occurs approximately 100 km from the easternmost constant head cell at  
513 SdA (or 50–70 km from the topographic divide). This length scale defines the upper distribution  
514 of flowpaths discharging at SdA. Once the transient simulation begins, the groundwater divide  
515 moves westward closer towards SdA as water is released from storage to augment discharge at  
516 SdA. In both simulations, after approximately 1,000 years, the groundwater divide reverses  
517 direction and begins to migrate eastward away from SdA as the volume of water released from  
518 storage decreases and regional groundwater recharge is captured. In the restrictive simulation,  
519 the position of the groundwater divide stabilizes by approximately 50,000–100,000 years around  
520 130 km east of the easternmost constant head cell of SdA. In the conducive simulation, the  
521 groundwater divide reaches the easternmost boundary of the model after 9,000 years and does  
522 not move westward for the remainder of the simulation.

523 These results show that the groundwater divide of the plateau-margin system is dynamic  
524 at time scales similar to changes in climate and moves in response to changing recharge



525 conditions. The position of the divide is also sensitive to presence of a lower conductivity block  
526 separating the discharge zone from the plateau, especially over longer time periods. While the  
527 “restrictive” case shows the greatest head change, it should be expected that groundwater divides  
528 along such portions of the plateau margin will be more stable over time than other segments of  
529 the plateau margin.

## 530 **6. Discussion and Conclusions**

531 The persistence and scope of questions relating to water imbalance in the Atacama Desert  
532 [e.g. *Magaritz et al.*, 1990; *Houston and Hart*, 2004; *Jordan et al.*, 2015], which is subject to  
533 high water resource demand for mining purposes, highlights the importance of better  
534 constraining groundwater divides and groundwater storage in these modern and paleo hydrologic  
535 systems. Observations of discharge along the Altiplano-Puna plateau margin greatly exceed our  
536 estimates of modern recharge rates to groundwater aquifers. In the absence of substantial  
537 overland flow this leaves an extreme hydrologic imbalance for catchments along the plateau  
538 margin.

539 The steady state assumption that recharge equals discharge is clearly not appropriate in  
540 this setting, despite its prevalence in the watershed management approaches throughout the  
541 region and globally. In order for recharge to equal discharge with hydrologic closure at steady  
542 state conditions, groundwater infiltration rates must be unrealistically high (15–24%; cf. *Scanlon*  
543 *et al.*, 2006). Recharge rates in basins to the east (Tuyajto; *Herrera, et al.* [2016]) and north (e.g.  
544 Pampa del Tamarugal; *Jayne et al.*, [2016]; and Salar de Huasco; *Uribe et al.*, [2016]) using  
545 steady-state conditions are fundamentally flawed in the conceptualization of the sources of  
546 recharge water. Recharge during infrequent and sporadic precipitation events [*Boutt, et al.*, 2016;  
547 *Masbruch et al.*, 2016] could be a potential source of water but it must be explained in the  
548 context of recharge rates constrained using CMB estimates, which should reflect long-term  
549 average recharge rates.

550 Assuming that recharge water is moving from up-gradient closed basins requires a  
551 reorganization of how topographic boundaries are treated in the catchment hydrologic budgets as  
552 widely applied elsewhere [*Haitjema and Mitchell-Bruker*, 2005; *Gleeson et al.*, 2011]. Effort  
553 should be spent on identifying the hydrogeologic controls on the flowpaths of water and being

554 able to distinguish this regional groundwater from local groundwater inputs using elemental,  
555 isotopic, and molecular tracers. In the case of regional steady state flow towards these basins,  
556 shallow hydraulic heads should show strong downward gradients and should have equipotentials  
557 that lower in magnitude towards the basins. Both of these conditions have significant  
558 implications for the water budget of the high elevation (> 4000 m) closed basins on the plateau.  
559 The water budget of these basins should be negative with some fraction of water flowing out of  
560 the basin to neighboring basins. One nearby basin, Laguna Tuayito shows that this is case  
561 [Herrera *et al.*, 2016]. Secondly, water levels in these basin floors are likely perched above a  
562 regional water table. Both considerations have strong implications for lake-based precipitation  
563 paleo-climate reconstruction. Reconstructions assume that lake levels in the closed basin  
564 respond solely to precipitation minus evaporation. If one were to assume losses of water  
565 between basins or out the basin bottom to the regional groundwater table, it would lead to an  
566 underestimate of precipitation in the region. Basins that receive substantial groundwater input  
567 from another would lead to an overestimate of precipitation. Understanding the magnitude of  
568 hydraulic losses through infiltration from perched basins above a regional water table is critical.

569 By coupling solute and water budgets, additional constraints may be gained. *Corenthal*  
570 *et al.* [2016] demonstrated that the modern sodium (Na) flux to the SdA basin could account for  
571 the halite and brine deposits over ~10 Ma, consistent with geological constraints. If the Na  
572 concentration of inflow water is assumed to be constant over this interval, the long-term average  
573 discharge rates are also required to remain relatively constant. Both the water molecule and any  
574 conservative solutes must achieve mass balance in a realistic conceptualization of the hydrologic  
575 system. Therefore, agreement in water and solute budgets is strong support for a realistic and  
576 reasonable hydrologic model. Similarly, recharge area and transient storage can be constrained  
577 by solute budgets. Although this is outside the scope of this manuscript, we suggest that the  
578 yield of weathering derived solutes per unit area must fall within the range observed for arid  
579 montane catchments globally to justify the size of the hydrogeologic watershed. Additionally,  
580 solute release rates from groundwater systems must match the modeled draining of transient  
581 storage and predicted mean residence times for groundwater. In the case where such constraints  
582 cannot be reconciled, a reconceptualization of the hydrogeologic model will require adjustments

583 that effect any combination of the following: (1) the scale of regional groundwater flow, (2) the  
584 mean residence time of water, and (3) the potential for deep, under-sampled flowpaths.

585 Our present conceptualization of the extreme modern hydrologic imbalance along the  
586 western margin of the Altiplano-Puna plateau can be explained by a combination of regional  
587 groundwater flow and transient draining of groundwater storage. Transient draining of  
588 groundwater storage is required because presumed recharge areas for many watersheds at the  
589 plateau margin overlap. Dynamic groundwater flow modeling suggests: (1) the water level  
590 changes at the salar margins (and discharging water to fans) are highly sensitive to changes in  
591 recharge on plateau, (2) the extent and magnitude of the changes in hydraulic head are controlled  
592 by the distribution of hydraulic conductivity at the plateau margin, (3) the contributing area to  
593 SdA changes, is not coincident with the topographic boundary, and is a dynamic feature, and (4)  
594 it is difficult to reconcile the modern position of the “water table” on the plateau with the  
595 regional groundwater flow conceptualization and the modern discharge to SdA (i.e., modern  
596 salars are perched and lose water to surrounding basins and ultimately to SdA and adjacent  
597 plateau margin catchments).

598 The hydrologic imbalance at SdA has important implications for paleoclimatic  
599 reconstructions, implying that paleo-lakes on the Altiplano lost water to SdA altering their  
600 hydrologic budgets and further complicating lake-level based paleoclimatic reconstructions.  
601 Because water resources of SdA are managed under the steady state assumption, these findings  
602 have implications for efforts to sustainably allocate water resources for mining, agricultural and  
603 environmental interests. Such considerations apply to many continental settings with strong  
604 gradients in landscape and climate, though the margins of large orogenic plateau are likely to  
605 exhibit the greatest hydrologic imbalance by virtue of their scale.

## 606 **7. Acknowledgments**

607  
608 The authors would like to acknowledge Rockwood Lithium, Inc./Albemarle Corporation for their  
609 continued support of this and related research to improve the understanding of the hydrogeology  
610 and geochemistry of the SdA environment. The ASTER DEM and Landsat 8 OLI were retrieved  
611 from EarthExplorer, courtesy of the NASA Land Processes Distributed Active Archive Center,

612 USGS/Earth Resources Observation and Science Center. Processed and calibrated TRMM 2B31  
613 dataset, a joint mission between NASA and the Japan Aerospace Exploration Agency, was  
614 kindly made publically accessible by B. Bookhagen.  
615

616 **8. References**

- 617 Alley, W. M., R. W. Healy, J. W. LaBaugh, and T. E. Reilly (2002), Flow and storage in  
 618 groundwater systems., *Science*, 296(5575), 1985–1990, doi:10.1126/science.1067123.
- 619 Andermann, C., L. Longuevergne, S. Bonnet, A. Crave, P. Davy, and R. Gloaguen (2012),  
 620 Impact of transient groundwater storage on the discharge of Himalayan rivers, *Nat.*  
 621 *Geosci.*, 5(2), 127–132, doi:10.1038/ngeo1356.
- 622 Anderson, M., R. Low, and S. Foot (2002), Sustainable groundwater development in arid, high  
 623 Andean basins, *Geol. Soc. London, Spec. Publ.*, 193(1), 133–144,  
 624 doi:10.1144/GSL.SP.2002.193.01.11.
- 625 Aron, F., G. González, E. Veloso, and J. Cembrano (2008), Architecture and style of  
 626 compressive Neogene deformation in the eastern-southeastern border of the Salar de  
 627 Atacama Basin (22°30′-24°15′S): A structural setting for the active volcanic arc of the  
 628 Central Andes, *Int. Symp. Andean Geodyn.*, (1998), 52–55.
- 629 Bazuhair, A. S., and W. W. Wood (1996), Chloride mass-balance method for estimating ground  
 630 water recharge in arid areas: Examples from western Saudi Arabia, *J. Hydrol.*, 186(1-4),  
 631 153–159, doi:10.1016/S0022-1694(96)03028-4.
- 632 Betancourt, J. L., C. Latorre, J. A. Rech, J. Quade, and K. A. Rylander (2000), A 22,000-Year  
 633 Record of Monsoonal Precipitation from Northern Chile’s Atacama Desert, *Science* (80-  
 634 .), 289(5484), 1542–1546, doi:10.1126/science.289.5484.1542.
- 635 Blodgett, T. a., J. D. Lenters, and B. L. Isacks (1997), Constraints on the origin of paleolake  
 636 expansions in the Central Andes, *Earth Interact.*, 1(1), 1–1, doi:10.1175/1087-  
 637 3562(1997)001<0001:CotOoP>2.0.CO;2.
- 638 Bobst, A. L., T. K. Lowenstein, T. E. Jordan, L. V. Godfrey, T. L. Ku, and S. Luo (2001), A 106  
 639 ka paleoclimate record from drill core of the Salar de Atacama, northern Chile,  
 640 *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 173(1-2), 21–42, doi:10.1016/S0031-  
 641 0182(01)00308-X.
- 642 Bookhagen, B., and M. R. Strecker (2008), Orographic barriers, high-resolution TRMM rainfall,  
 643 and relief variations along the eastern Andes, *Geophys. Res. Lett.*, 35(6), 1–6,  
 644 doi:10.1029/2007GL032011.
- 645 Boutt D. F., Hynek S. A., Munk L. A., and Corenthal L. G. (2016) Rapid recharge of fresh water  
 646 to the halite-hosted brine aquifer of Salar de Atacama, Chile, *Hydrol. Process.*, 30: 4720–  
 647 4740. doi: 10.1002/hyp.10994.
- 648 Cervetto Sepulveda, M. M. (2012), Caracterización hidrogeológica e hidrogeoquímica de las  
 649 cuencas: Salar de Aguas Calientes 2, Puntas Negras, Laguna Tuyajto, Pampa Colorada,  
 650 Pampa las Tecas y Salar el Laco, II Region de Chile [M.S. Thesis], Universidad de Chile.
- 651 Condom, T., A. Coudrain, A. Dezetter, D. Brunstein, F. Delclaux, and S. Jean-Emmanuel (2004),  
 652 Transient modelling of lacustrine regressions: two case studies from the Andean  
 653 Altiplano, *Hydrol. Process.*, 18(13), 2395–2408, doi:10.1002/hyp.1470.
- 654 Condon, L. E., and R. M. Maxwell (2015), Evaluating the relationship between topography and  
 655 groundwater using outputs from a continental-scale integrated hydrology model, *Water*  
 656 *Resour. Res.*, 51, 6602–6621, doi:10.1002/2014WR016774.
- 657 Corenthal, L. G., D. F. Boutt, S. A. Hynek, and L. A. Munk (2016), Regional groundwater flow  
 658 and accumulation of a massive evaporite deposit at the margin of the Chilean Altiplano,  
 659 *Geophys. Res. Lett.*, 43, doi:10.1002/2016GL070076

- 660 Craig, H. (1961), Isotopic Variations in Meteoric Waters., *Science*, 133(3465), 1702–3,  
661 doi:10.1126/science.133.3465.1702.
- 662 Crossey, L. J., T. P. Fischer, P. J. Patchett, K. E. Karlstrom, D. R. Hilton, D. L. Newell, P.  
663 Huntoon, A. C. Reynolds, and G. a. M. de Leeuw (2006), Dissected hydrologic system at  
664 the Grand Canyon: Interaction between deeply derived fluids and plateau aquifer waters  
665 in modern springs and travertine, *Geology*, 34(1), 25, doi:10.1130/G22057.1.
- 666 Currell, M., Gleeson, T., & Dahlhaus, P. (2016). A new assessment framework for transience in  
667 hydrogeological systems. *Groundwater*, 54(1), 4–14.
- 668 Dirección General de Aguas (2013), Análisis de la Oferta Hídrica del Salar de Atacama,  
669 Santiago, Chile.
- 670 Dunai, T. J., G. a. González López, and J. Juez-Larré (2005), Oligocene-Miocene age of aridity  
671 in the Atacama Desert revealed by exposure dating of erosion-sensitive landforms,  
672 *Geology*, 33(4), 321–324, doi:10.1130/G21184.1.
- 673 Fritz, S. C., P. a. Baker, T. K. Lowenstein, G. O. Seltzer, C. a. Rigsby, G. S. Dwyer, P. M. Tapia,  
674 K. K. Arnold, T. L. Ku, and S. Luo (2004), Hydrologic variation during the last 170,000  
675 years in the southern hemisphere tropics of South America, *Quat. Res.*, 61(1), 95–104,  
676 doi:10.1016/j.yqres.2003.08.007.
- 677 Garreaud, R., M. Vuille, and A. C. Clement (2003), The climate of the Altiplano: Observed  
678 current conditions and mechanisms of past changes, *Palaeogeogr. Palaeoclimatol.*  
679 *Palaeoecol.*, 194(1-3), 5–22, doi:10.1016/S0031-0182(03)00269-4.
- 680 Gayo, E. M., C. Latorre, T. E. Jordan, P. L. Nester, S. A. Estay, K. F. Ojeda, and C. M. Santoro  
681 (2012), Late Quaternary hydrological and ecological changes in the hyperarid core of the  
682 northern Atacama Desert (~21°S), *Earth-Science Rev.*, 113(3-4), 120–140,  
683 doi:10.1016/j.earscirev.2012.04.003.
- 684 Gelhar, L. W. & Wilson, J. L. Ground-Water Quality Modeling. *Ground Water* **12**, 399–408  
685 (1974).
- 686 Gorelick, S. M., and C. Zheng (2015), Global change and the groundwater management  
687 challenge, *Water Resour. Res.*, 51, 3031–3051, doi:[10.1002/2014WR016825](https://doi.org/10.1002/2014WR016825).
- 688 Gleeson, T., L. Marklund, L. Smith, and A. H. Manning (2011), Classifying the water table at  
689 regional to continental scales, *Geophys. Res. Lett.*, 38(5), 1–6,  
690 doi:10.1029/2010GL046427.
- 691 Gleeson, T., Y. Wada, M. F. P. Bierkens, and L. P. H. van Beek (2012), Water balance of global  
692 aquifers revealed by groundwater footprint, *Nature*, 488(7410), 197–200,  
693 doi:10.1038/nature11295.
- 694 Godfrey, L. V., T. E. Jordan, T. K. Lowenstein, and R. L. Alonso (2003), Stable isotope  
695 constraints on the transport of water to the Andes between 22o and 26oS during the last  
696 glacial cycle, *Palaeogeogr. Palaeoclimatol. Palaeoecol.*, 194(1-3), 299–317,  
697 doi:10.1016/S0031-0182(03)00283-9.
- 698 Grosjean, M., M. A. Geyh, B. Messerli, and U. Schotterer (1995), Late-glacial and early  
699 Holocene lake sediments, ground-water formation and climate in the Atacama Altiplano  
700 22°24'S, *J. Paleolimnol.*, 14(3), 241–252, doi:10.1007/BF00682426.
- 701 Haitjema, H. M., and S. Mitchell-Bruker (2005), Are Water Tables a Subdued Replica of the  
702 Topography?, *Ground Water*, 43(6), 781–786, doi:10.1111/j.1745-6584.2005.00090.x.

- 703 Hartley, A. J., and G. Chong (2002), Late Pliocene age for the Atacama Desert: Implications for  
704 the desertification of western South America, *Geology*, 30(1), 43–46, doi:10.1130/0091-  
705 7613(2002)030<0043:LPAFTA>2.0.CO;2.
- 706 Herrera Lameli, C. (2011), Informe Final Estudio Hidrogeológico Proyecto “ Planta de Sulfato  
707 de Cobre Pentahidratado ,” , 0–33.
- 708 Herrera, C. et al., 2016. Groundwater flow in a closed basin with a saline shallow lake in a  
709 volcanic area: Laguna Tuyajto, northern Chilean Altiplano of the Andes. *Science of the*  
710 *Total Environment*, 541, p.303-318.
- 711 Hoke, G. D., B. L. Isacks, T. E. Jordan, and J. S. Yu (2004), Groundwater-sapping origin for the  
712 giant quebradas of northern Chile, *Geology*, 32(7), 605–608, doi:10.1130/G20601.1.
- 713 Houston, J. (2006a), Evaporation in the Atacama Desert: An empirical study of spatio-temporal  
714 variations and their causes, *J. Hydrol.*, 330(3-4), 402–412,  
715 doi:10.1016/j.jhydrol.2006.03.036.
- 716 Houston, J. (2006b), The great Atacama flood of 2001 and its implications for Andean  
717 hydrology, *Hydrol. Process.*, 20(3), 591–610, doi:10.1002/hyp.5926.
- 718 Houston, J. (2007), Recharge to groundwater in the Turi Basin, northern Chile: An evaluation  
719 based on tritium and chloride mass balance techniques, *J. Hydrol.*, 334(3-4), 534–544,  
720 doi:10.1016/j.jhydrol.2006.10.030.
- 721 Houston, J. (2009), A recharge model for high altitude, arid, Andean aquifers, *Hydrol. Process.*,  
722 23(16), 2383–2393, doi:10.1002/hyp.7350.
- 723 Houston, J., and D. Hart (2004), Theoretical head decay in closed basin aquifers: an insight into  
724 fossil groundwater and recharge events in the Andes of northern Chile, *Quat. J. Eng.*  
725 *Geol. Hydrogeol.*, 37(December 2007), 131–139, doi:10.1144/1470-9236/04-007.
- 726 Ingebritsen SE, Manning CE (1999) Geological implications of a permeability-depth curve for  
727 the continental crust. *Geology*, 27, 1,107–10.
- 728 Jayne, R. S., R. M. Pollyea, J. P. Dodd, E. J. Olson, and S. K. Swanson (2016), Spatial and  
729 temporal constraints on regional-scale groundwater flow in the Pampa del Tamarugal  
730 Basin, Atacama Desert, Chile, *Hydrogeol. J.*, (August), 1921–1937, doi:10.1007/s10040-  
731 016-1454-3.
- 732 Johnson, E., J. Yáñez, C. Ortiz, and J. Muñoz (2010), Evaporation from shallow groundwater in  
733 closed basins in the Chilean Altiplano, *Hydrol. Sci. J.*, 55(4), 624–635,  
734 doi:10.1080/02626661003780458.
- 735 Jordan, T., C. Herrera Lameli, N. Kirk-Lawlor, and L. Godfrey (2015), Architecture of the  
736 aquifers of the Calama Basin, Loa catchment basin, northern Chile, *Geosphere*, 11(5),  
737 GES01176.1, doi:10.1130/GES01176.1.
- 738 Jordan, T. E., N. Munoz, M. Hein, T. Lowenstein, L. Godfrey, and J. Yu (2002a), Active faulting  
739 and folding without topographic expression in an evaporite basin, Chile, *Bull. Geol. Soc.*  
740 *Am.*, 114(11), 1406–1421, doi:10.1130/0016-7606(2002)114<1406:AFAFWT>2.0.CO;2.
- 741 Jordan, T. E., L. V. Godfrey, N. Munoz, R. N. Alonso, T. K. Lowenstein, G. D. Hoke, N.  
742 Peranginangin, B. L. Isacks, and L. Cathles (2002b), Orogenic-scale ground water  
743 circulation in teh Central Andes: evidence and consequences., 5th ISAG (International  
744 Symp. Andean Geodyn., 331–334.
- 745 Jordan, T. E., C. Mpodozis, N. Muñoz, N. Blanco, P. Pananont, and M. Gardeweg (2007),  
746 Cenozoic subsurface stratigraphy and structure of the Salar de Atacama Basin, northern  
747 Chile, *J. South Am. Earth Sci.*, 23(2-3), 122–146, doi:10.1016/j.jsames.2006.09.024.

- 748 Kampf, S. K., and S. W. Tyler (2006), Spatial characterization of land surface energy fluxes and  
749 uncertainty estimation at the Salar de Atacama, Northern Chile, *Adv. Water Resour.*,  
750 29(2), 336–354, doi:10.1016/j.advwatres.2005.02.017.
- 751 Lamb, S., and P. Davis (2003), Cenozoic climate change as a possible cause for the rise of the  
752 Andes., *Nature*, 425(6960), 792–797, doi:10.1038/nature02049.
- 753 Lasaga, A. C. & Berner, R. A. Fundamental aspects of quantitative models for geochemical  
754 cycles. *Chem. Geol.* **145**, 161–175 (1998).
- 755 Lowenstein, T. K., and F. Risacher (2009), Closed basin brine evolution and the influence of Ca–  
756 Cl inflow waters: Death Valley and Bristol Dry Lake California, Qaidam Basin, China,  
757 and Salar de Atacama, Chile, *Aquat. Geochem.*, 15, 71–94, doi:10.1007/s10498-008-  
758 9046-z.
- 759 Lowenstein, T. K., M. C. Hein, A. L. Bobst, T. E. Jordan, T.-L. Ku, and S. Luo (2003), An  
760 Assessment of stratigraphic completeness in climate-sensitive closed-basin lake  
761 sediments: Salar de Atacama, Chile, *J. Sediment. Res.*, 73(1), 91–104,  
762 doi:10.1306/061002730091.
- 763 Magaritz, M., R. Aravena, H. Pena, O. Suzuki, and A. Grilli (1990), Source of ground water in  
764 the deserts of Northern Chile: Evidence of deep circulation of ground water from the  
765 Andes, *Ground Water*, 28(4), 513–517, doi:10.1111/j.1745-6584.1990.tb01706.x.
- 766 Mardones, L. (1986), Características geológicas e hidrogeológicas del Salar de Atacama, in *El*  
767 *litio: Un nuevo recurso para Chile*, edited by G. Lagos, pp. 181–216, Universidad de  
768 Chile, Santiago, Chile.
- 769 Martínez, F., López, C., Bascuñan, S., & Arriagada, C. (2018). Tectonic interaction between  
770 Mesozoic to Cenozoic extensional and contractional structures in the Preandean  
771 Depression (23°–25°S): Geologic implications for the Central Andes. *Tectonophysics*,  
772 744, 333–349. <https://doi.org/10.1016/j.tecto.2018.07.016>
- 773 Masbruch, M. D., C. A. Rumsey, S. Gangopadhyay, D. D. Susong, and T. Pruitt (2016),  
774 Analyses of infrequent (quasi-decadal) large groundwater recharge events in the northern  
775 Great Basin: Their importance for groundwater availability, use, and management, *Water*  
776 *Resour. Res.*, 52, doi:10.1002/2016WR019060.
- 777 Maxwell, P. (2014), Analysing the lithium industry: Demand, supply, and emerging  
778 developments, *Miner. Econ.*, 26(3), 97–106, doi:10.1007/s13563-013-0041-5.
- 779 McDonald, M. G., and A. W. Harbaugh (1988), A modular three-dimensional finite-difference  
780 ground-water flow model, United States Geological Survey.
- 781 McGuire, K. J., and J. J. McDonnell (2010), Hydrological connectivity of hillslopes and streams:  
782 Characteristic time scales and nonlinearities, *Water Resour. Res.*, 46, W10543,  
783 doi:10.1029/2010WR009341.
- 784 McNamara, J. P., D. Tetzlaff, K. Bishop, C. Soulsby, M. Seyfried, N. E. Peters, B. T. Aulenbach,  
785 and R. Hooper (2011), Storage as a metric of catchment comparison, *Hydrol. Processes*,  
786 25, 3364–3371.
- 787 Montgomery, E. L., M. J. Rosko, S. O. Castro, B. R. Keller, and P. S. Bevacqua (2003),  
788 Interbasin underflow between closed Altiplano basins in Chile., *Ground Water*, 41(4),  
789 523–31.
- 790 Munk, L.A., Hynek, S.A., Bradley, D.C., Boutt, D.F., Labay, K., Jochens, H., 2016. Lithium  
791 Brines: A Global Perspective, in Verplanck, P.L. and Hitzman, M.W., eds., *Rare Earth and*  
792 *Critical Elements in Ore Deposits. Reviews in Economic Geology* (18), 339–365.



- 793 Munk, L. A., Boutt, D. F., Hynek, S. A., & Moran, B. J. (2018). Hydrogeochemical fluxes and  
794 processes contributing to the formation of lithium-enriched brines in a hyper-arid  
795 continental basin. *Chemical Geology*, 493, 37–57.  
796 <https://doi.org/10.1016/j.chemgeo.2018.05.013>
- 797 Muñoz, Enrique, Álvarez, César, Billib, Max, Arumí, José Luis, & Rivera, Diego. (2011).  
798 Comparison of Gridded and Measured Rainfall Data for Basin-scale Hydrological  
799 Studies. *Chilean journal of agricultural research*, 71(3), 459-468. doi: 10.4067/S0718-  
800 58392011000300018
- 801 Nelson, S.T., Anderson, K.W., and Mayo, A.L., 2004, Testing the interbasin flow hypothesis at  
802 Death Valley, CA, USA: *Eos*, v. 85, p. 349, 355-356.
- 803 Nelson, S.T., and Mayo, A.L., 2014, The role of interbasin groundwater transfers in geologically  
804 complex terranes, demonstrated by the Great Basin in the western United States:  
805 *Hydrogeology Journal*, v. 22, p. 807-822, DOI 10.1007/s10040-014-1104-6.
- 806 Pérez-Fodich, A., M. Reich, F. Álvarez, G. T. Snyder, R. Schoenberg, G. Vargas, Y. Muramatsu,  
807 and U. Fehn (2014), Climate change and tectonic uplift triggered the formation of the  
808 atacama desert's giant nitrate deposits, *Geology*, 42(3), 251–254, doi:10.1130/G34969.1.
- 809 Placzek, C., J. Quade, and P. J. Patchett (2006), Geochronology and stratigraphy of late  
810 Pleistocene lake cycles on the southern Bolivian Altiplano: Implications for causes of  
811 tropical climate change, *Bull. Geol. Soc. Am.*, 118(5-6), 515–532, doi:10.1130/B25770.1.
- 812 Placzek, C. J., J. Quade, and P. J. Patchett (2013), A 130ka reconstruction of rainfall on the  
813 Bolivian Altiplano, *Earth Planet. Sci. Lett.*, 363, 97–108, doi:10.1016/j.epsl.2012.12.017.
- 814 Quade, J., J. a. Rech, J. L. Betancourt, C. Latorre, B. Quade, K. A. Rylander, and T. Fisher  
815 (2008), Paleowetlands and regional climate change in the central Atacama Desert,  
816 northern Chile, *Quat. Res.*, 69(3), 343–360, doi:10.1016/j.yqres.2008.01.003.
- 817 Ramirez, C., and M. Gardeweg (1982), *Carta Geologica de Chile, escala 1:250000, Hoja*  
818 *Toconao, Region de Antofagasta, Chile No. 54, Santiago, Chile.*
- 819 Rech, J. a, J. L. Betancourt, and J. Quade (2002), Late Quaternary paleohydrology of the central  
820 Atacama Desert (lat 22 -24 S), Chile, *Geol. Soc. Am. Bull.*, 114(3), 334–348,  
821 doi:10.1130/0016-7606(2002)114<0334:LQPOTC>2.0.CO;2.
- 822 Rech, J. a., J. S. Pigati, J. Quade, and J. L. Betancourt (2003), Re-evaluation of mid-Holocene  
823 deposits at Quebrada Puripica, northern Chile, *Palaeogeogr. Palaeoclimatol. Palaeoecol.*,  
824 194, 207–222, doi:10.1016/S0031-0182(03)00278-5.
- 825 Reutter, K.-J. et al. (2006), The Salar de Atacama Basin : A Subsiding block within the western  
826 edge of the Altiplano-Puna Plateau, *Andes. Act. Subduction Orogeny*, 303–325.
- 827 Risacher, F., H. Alonso, and C. Salazar (2003), The origin of brines and salts in Chilean salars: A  
828 hydrochemical review, *Earth-Science Rev.*, 63(3-4), 249–293, doi:10.1016/S0012-  
829 8252(03)00037-0.
- 830 Rissmann, C., M. Leybourne, C. Benn, and B. Christenson (2015), The origin of solutes within  
831 the groundwaters of a high Andean aquifer, *Chem. Geol.*, 396, 164–181,  
832 doi:10.1016/j.chemgeo.2014.11.029.
- 833 Rubilar, J., Martínez, F., Arriagada, C., Becerra, J., 2017. Structure of the Cordillera de la Sal: A  
834 key tectonic element for the Oligocene-Neogene evolution of the Salar de Atacama basin,  
835 Central Andes, northern Chile. *J. S. Am. Earth Sci.*  
836 <https://doi.org/10.1016/j.jsames.2017.11.013>.

- 837 Salas, J., J. Guimerà, O. Cornellà, R. Aravena, E. Guzmán, C. Tore, W. Von Igel, and R. Moreno  
838 (2010), Hidrogeología del sistema lagunar del margen este del Salar de Atacama (Chile),  
839 Bol. Geol. y Min., 121(4), 357–372.
- 840 Salisbury, M. J., B. R. Jicha, S. L. de Silva, B. S. Singer, N. C. Jiménez, and M. H. Ort (2011),  
841  $^{40}\text{Ar}/^{39}\text{Ar}$  chronostratigraphy of Altiplano-Puna volcanic complex ignimbrites reveals  
842 the development of a major magmatic province, Bull. Geol. Soc. Am., 123(5), 821–840,  
843 doi:10.1130/B30280.1.
- 844 Scanlon, B. R., K. E. Keese, A. L. Flint, L. E. Flint, C. B. Gaye, W. M. Edmunds, and I.  
845 Simmers (2006), Global synthesis of groundwater recharge in semiarid and arid regions,  
846 Hydrol. Process., 20(15), 3335–3370, doi:10.1002/hyp.6335.
- 847 Schaller, M. F., and Y. Fan (2009), River basins as groundwater exporters and importers:  
848 Implications for water cycle and climate modeling, J. Geophys. Res. Atmos., 114(4),  
849 doi:10.1029/2008JD010636.
- 850 Sociedad Chilena de Litio Ltda. (2009), Informe Hidrología e Hidrogeología Adenda 1 - EIA  
851 Modificaciones y Mejoramiento del Sistema de Pozas de Evaporación Solar en el Salar  
852 de Atacama, Antofagasta, Chile.
- 853 Tóth, J. (1963), A theoretical analysis of groundwater flow in small drainage basins, J. Geophys.  
854 Res., 68(16), 4795–4812, doi:10.1029/JZ068i016p04795.
- 855 Tyler, S. W., J. F. Muñoz, and W. W. Wood (2006), The response of playa and sabkha  
856 hydraulics and mineralogy to climate forcing, Ground Water, 44(3), 329–338,  
857 doi:10.1111/j.1745-6584.2005.00096.x.
- 858 Urbano, I. D., M. Person, K. Kelts, and J. S. Hanor (2004), Transient groundwater impacts on the  
859 development of paleoclimatic lake records in semi-arid environments, Geofluids, 4(3),  
860 187–196, doi:10.1111/j.1468-8123.2004.00081.x.
- 861 Uribe, J., Muñoz, J.F., Gironás, J. Oyarzún, R., Aguirre, E. and Aravena, R. (2016)  
862 Assessing groundwater recharge in an Andean closed basin using isotopic characterization and a  
863 rainfall-runoff model: Salar del Huasco basin, Chile, Hydrogeology Journal 23 (7), 1535-  
864 1551
- 865 van Beek, L. P. H., Y. Wada, and M. F. P. Bierkens (2011), Global monthly water stress: 1.  
866 Water balance and water availability, Water Resour. Res., 47, W07517,  
867 doi:10.1029/2010WR009791
- 868 Vuille, M., and C. Ammann (1997), Regional snowfall patterns in the high, arid Andes, Clim.  
869 Change, 36, 413–423, doi:10.1007/978-94-015-8905-5\_10.
- 870 Ward, K. M., G. Zandt, S. L. Beck, D. H. Christensen, and H. McFarlin (2014), Seismic imaging  
871 of the magmatic underpinnings beneath the Altiplano-Puna volcanic complex from the  
872 joint inversion of surface wave dispersion and receiver functions, Earth Planet. Sci. Lett.,  
873 404, 43–53, doi:10.1016/j.epsl.2014.07.022.
- 874

875 FIGURE CAPTIONS:

876 **Figure 1:** Locator map and cross-section of elevation and precipitation for the SdA region of 21  
877 to 25° S. (a) Elevations (0 to > 6,000 m) from an Advanced Spaceborne Thermal Emission and  
878 Reflection Radiometer (ASTER) Global Digital Elevation Model (GDEM). Gages maintained by  
879 the Chilean DGA. (b) ASTER GDEM derived elevation and TRMM 2B31 estimates of MAP  
880 from 1998 to 2009.

881 **Figure 2:** (a) Stable isotopic composition of samples considered for CMB calculations. Samples  
882 collected as part of this study and from *Cervetto* [2012]. Only samples of inflow groundwater  
883 from wells in alluvial fans that plot closely to the global meteoric water line, shown as solid  
884 black diamonds, were selected for the CMB calculations. (b) Stable isotopic composition of  
885 groundwater samples within the topographic watershed used for CMB analysis. CMB cutoff is  
886 deuterium excess <5 ‰.

887 **Figure 3** Results of chloride mass balance (CMB). (a) GWR determined by the CMB method  
888 based on samples in the SdA, Linzor basin and Chilean Puna Plateau. Uncertainty includes a  
889 range of Cl in precipitation from 5–16 mg/l. MAP from the TRMM 2B31 dataset. Solid line is  
890 the best fit to calculated CMB results, while the dashed line shows the infiltration rate required to  
891 close the steady state hydrologic budget in the SdA topographic watershed. (b) Frequency  
892 distribution of gridded (25 km<sup>2</sup>) MAP from the TRMM 2B31 dataset within the topographic  
893 watershed.

894 **Figure 4** Spatial distribution of P, GWR for the median precipitation scenario and PET used to  
895 evaluate the topographic and regional scale hydrologic budget of the SdA. (a) MAP from 1998–  
896 2009 based on TRMM 2B31 dataset. Black circles are groundwater Cl sample locations. (b)  
897 Median annual GWR from 1998-2009 determined by applying equation (2) to median  
898 precipitation dataset. (c) PET determined as a function of elevation applied to discharge zones  
899 (gray polygons), excluding SdA.

900 **Figure 5** A) Regional watersheds from Corenthal et al. (2016) for the evaluation the steady state  
901 contributing regions for the Salar de Atacama and Rio Loa Basins using full uncertainty  
902 considerations for the water balance. Each lettered zone for SdA includes the cumulative area of

903 all smaller zones. A is the topographic watershed, and M is the inferred hydrogeologic watershed  
904 where  $GWR+R$  balances ET in the full uncertainty bounds. The Rio Loa contributing areas are  
905 from Jordan et al. (2015). Background is an ASTER DEM. Dashed line indicates the position of  
906 the 2-D model presented in Figure 6. Numbered locations refer to modern and paleo hydraulic  
907 head estimates in Figures 7 and 8. B) Chloride mass balance recharge + runoff estimates (blue  
908 diamonds) for lower (light blue) and upper bound recharge (dark blue) scenarios.  
909 Evapotranspiration bounds (red diamonds) for lower (light red) and upper estimates (dark red).  
910 Letters indicate the areas represented by the black polygons in A).

911 **Figure 6** Model geometry, boundary conditions, hydraulic conductivity distribution, and initial  
912 water table positions for the restrictive and conducive simulations described in text.

913 **Figure 7:** Simulated hydraulic head distributions for (A) restrictive and (B) conducive  
914 simulations at time of 0, 1000, 10000, 100000 years after change in recharge. Position of the  
915 hydrogeologic divides are shown for labeled times in vertical black lines. Numbered locations  
916 correspond to head observations locations plotted in Figure 8.

917 **Figure 8** Simulated hydraulic heads (y-axis) for restrictive and conducive simulations compared  
918 to modern field and paleo-hydraulic head estimates (x-axis). Sizes of colored polygons (pink –  
919 conducive, blue – restrictive) are based on simulated heads with the highest position being the  
920 initial condition for the models. Highest observed heads are estimated based on paleo-  
921 hydrologic information cited in text and presented in Supporting Information 5.

922 **Figure 9** Plot of the ratio of discharge from constant head cells at SdA and drains along the  
923 plateau margin ( $D_{SdA}$ ) to specified total model recharge ( $GW_{TOTALRCH}$ ). The un-shaded region  
924 represents discharge to SdA that is greater than total model recharge (i.e. some contribution of  
925 groundwater storage). The shaded represents times in the simulation where discharge to SdA is  
926 less than the total model recharge.

927

928 TABLE CAPTIONS:

929 **Table 1:** Locations and characteristics of precipitation samples used to determine chloride  
930 concentrations in precipitation

931 **Table 2:** Predicted precipitation and recharge for topographic and hydrogeologic catchments

932 **Table 1:** Tabulated attributes and results of groundwater chloride analyses including estimates of

933 groundwater recharge from the CMB method.

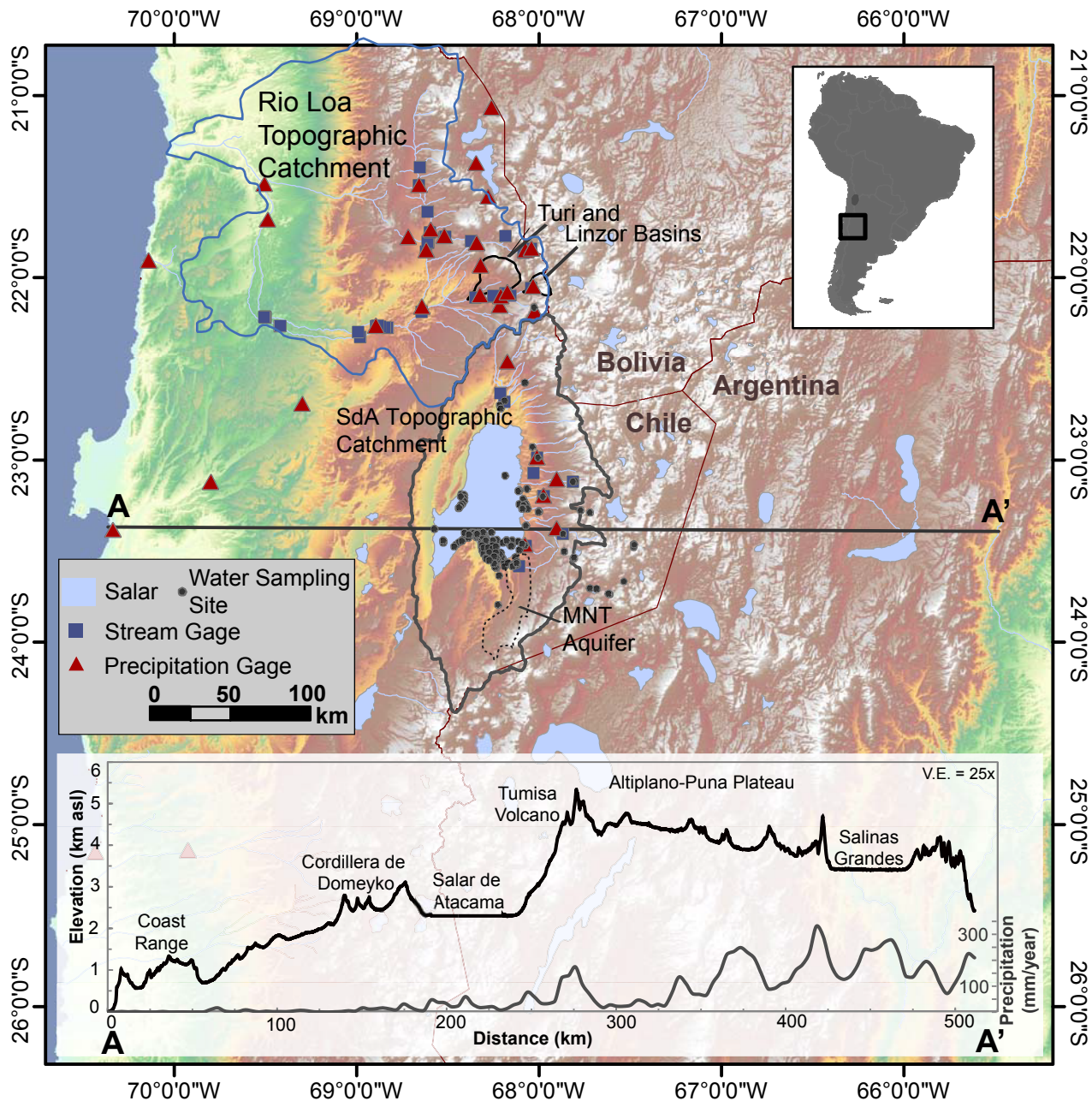


Figure 1: Locator map and cross-section of elevation and precipitation for the SdA region of 21 to 25° S. (a) Elevations (0 to > 6,000 m) from an Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER) Global Digital Elevation Model (GDEM). Gages maintained by the Chilean DGA. (b) ASTER GDEM derived elevation and TRMM 2B31 estimates of MAP from 1998 to 2009.



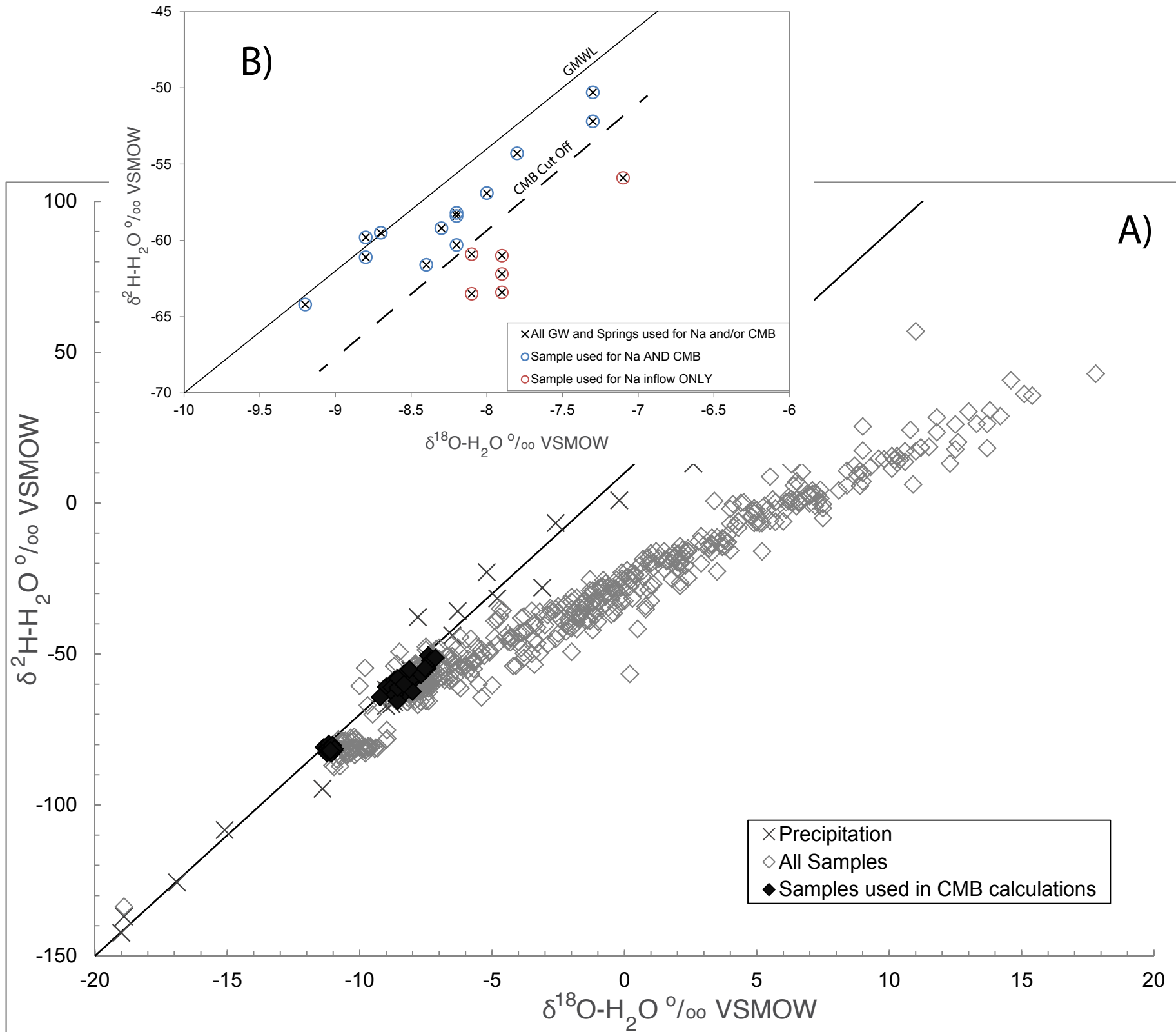


Figure 2: (a) Stable isotopic composition of samples considered for CMB calculations. Samples collected as part of this study and from Cervetto [2012]. Only samples of inflow groundwater from wells in alluvial fans that plot closely to the global meteoric water line, shown as solid black diamonds, were selected for the CMB calculations. (b) Stable isotopic composition of groundwater samples within the topographic watershed used for CMB analysis. CMB cutoff is deuterium excess  $<5$  ‰.

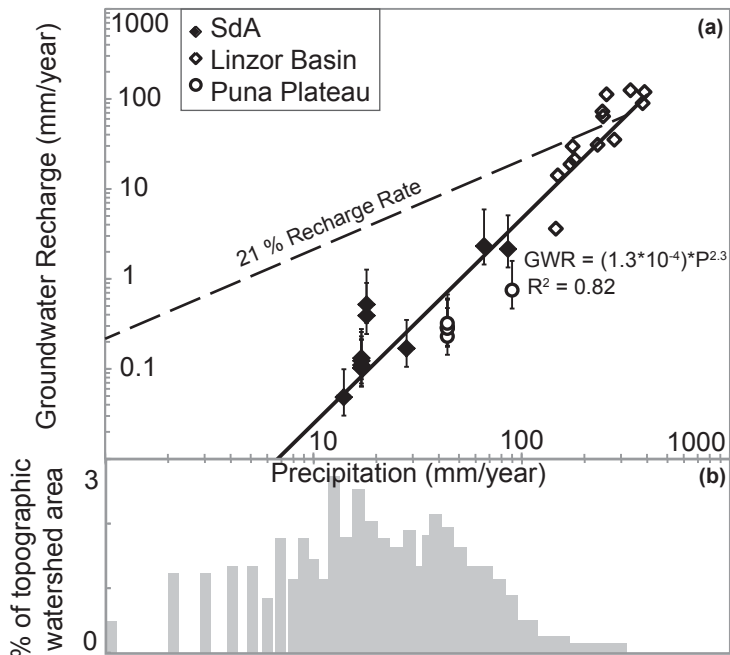


Figure 3 Results of chloride mass balance (CMB). (a) GWR determined by the CMB method based on samples in the SdA, Linzor basin and Chilean Puna Plateau. Uncertainty includes a range of Cl in precipitation from 5–16 mg/l. MAP from the TRMM 2B31 dataset. Solid line is the best fit to calculated CMB results, while the dashed line shows the infiltration rate required to close the steady state hydrologic



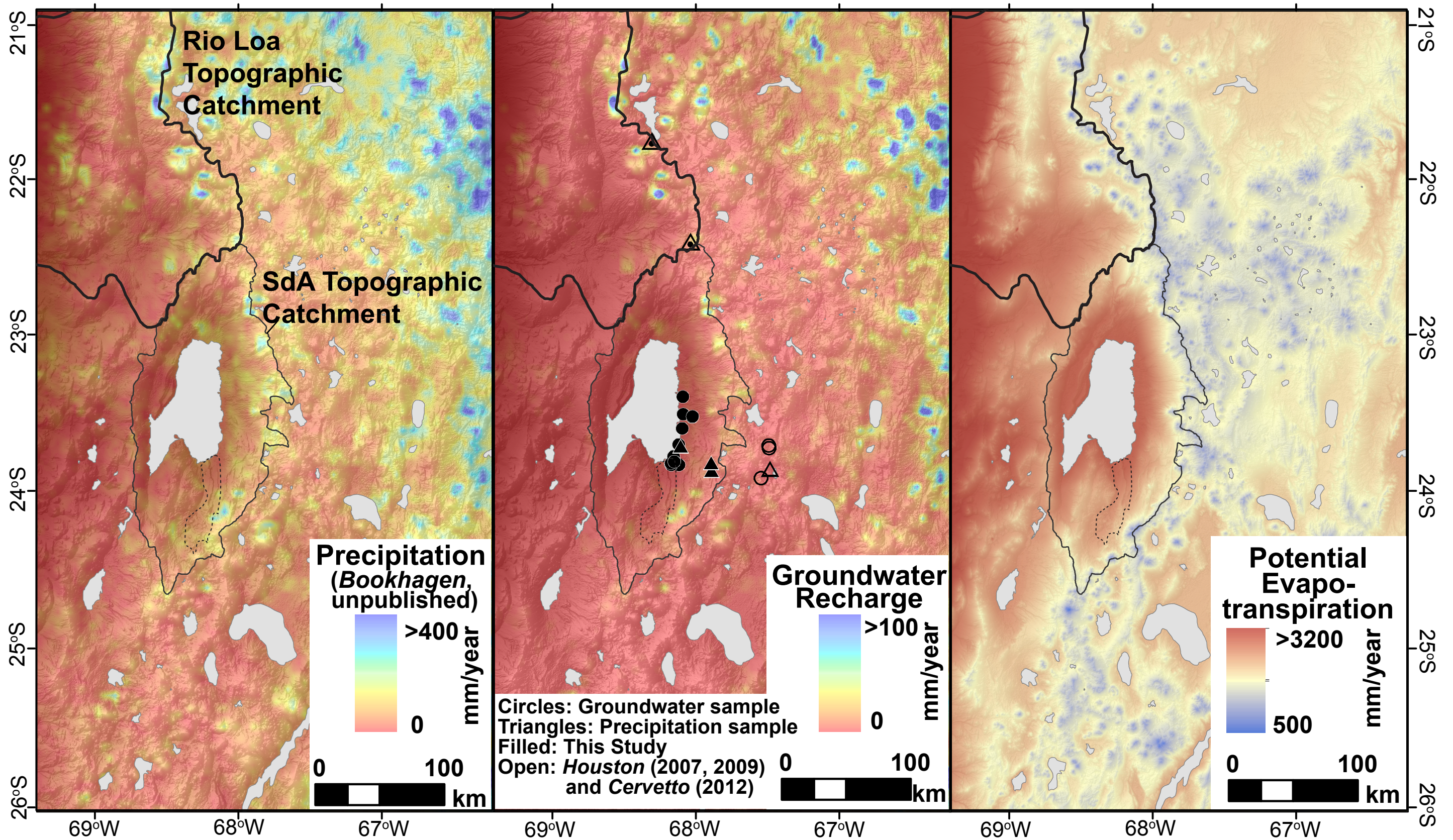


Figure 4 Spatial distribution of P, GWR for the median precipitation scenario and PET used to evaluate the topographic and regional scale hydrologic budget of the SdA. (a) MAP from 1998–2009 based on TRMM 2B31 dataset. Black circles are groundwater CI sample locations. (b) Median annual GWR from 1998–2009 determined by applying equation (2) to median precipitation dataset. (c) PET determined as a function of elevation applied to discharge zones (gray polygons), excluding SdA.



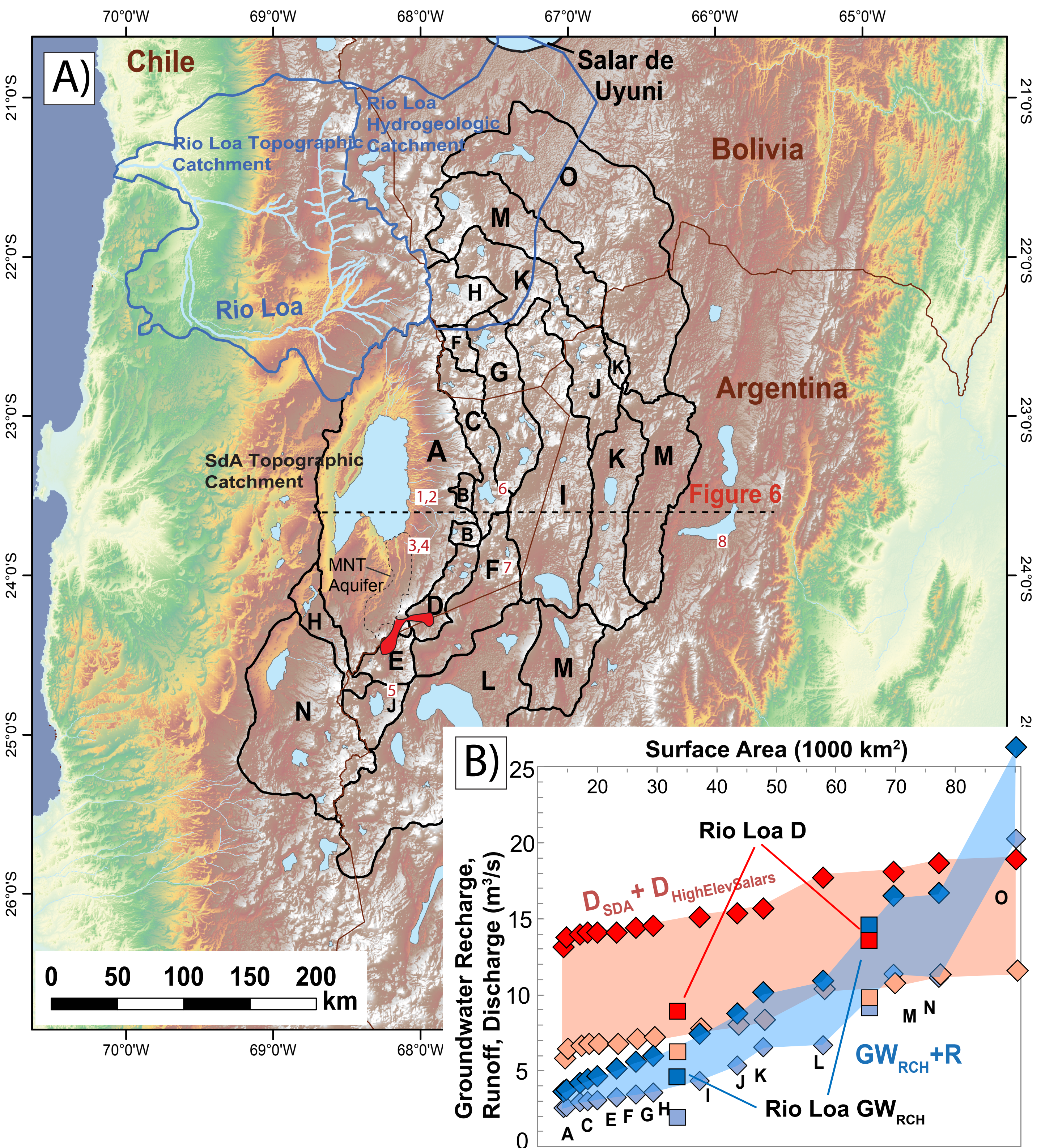


Figure 5 A) Regional watersheds from Corenthal et al. (2016) for the evaluation of the steady state contributing regions for the Salar de Atacama and Rio Loa Basins using full uncertainty considerations for the water balance. Each lettered zone for SdA includes the cumulative area of all smaller zones. A is the topographic watershed, and M is the inferred hydrogeologic watershed where GWR+R balances ET in the full uncertainty bounds. The Rio Loa contributing areas are from Jordan et al. (2015). Background is an ASTER DEM. Dashed line indicates the position of the 2-D model presented in Figure 6. Numbered locations refer to modern and paleo hydraulic head estimates in Figures 7 and 8. B) Chloride mass balance recharge + runoff estimates (blue diamonds) for lower (light blue) and upper bound recharge (dark blue) scenarios. Evapotranspiration bounds (red diamonds) for lower (light red) and upper estimates (dark red). Letters indicate the areas represented by the black polygons in A).



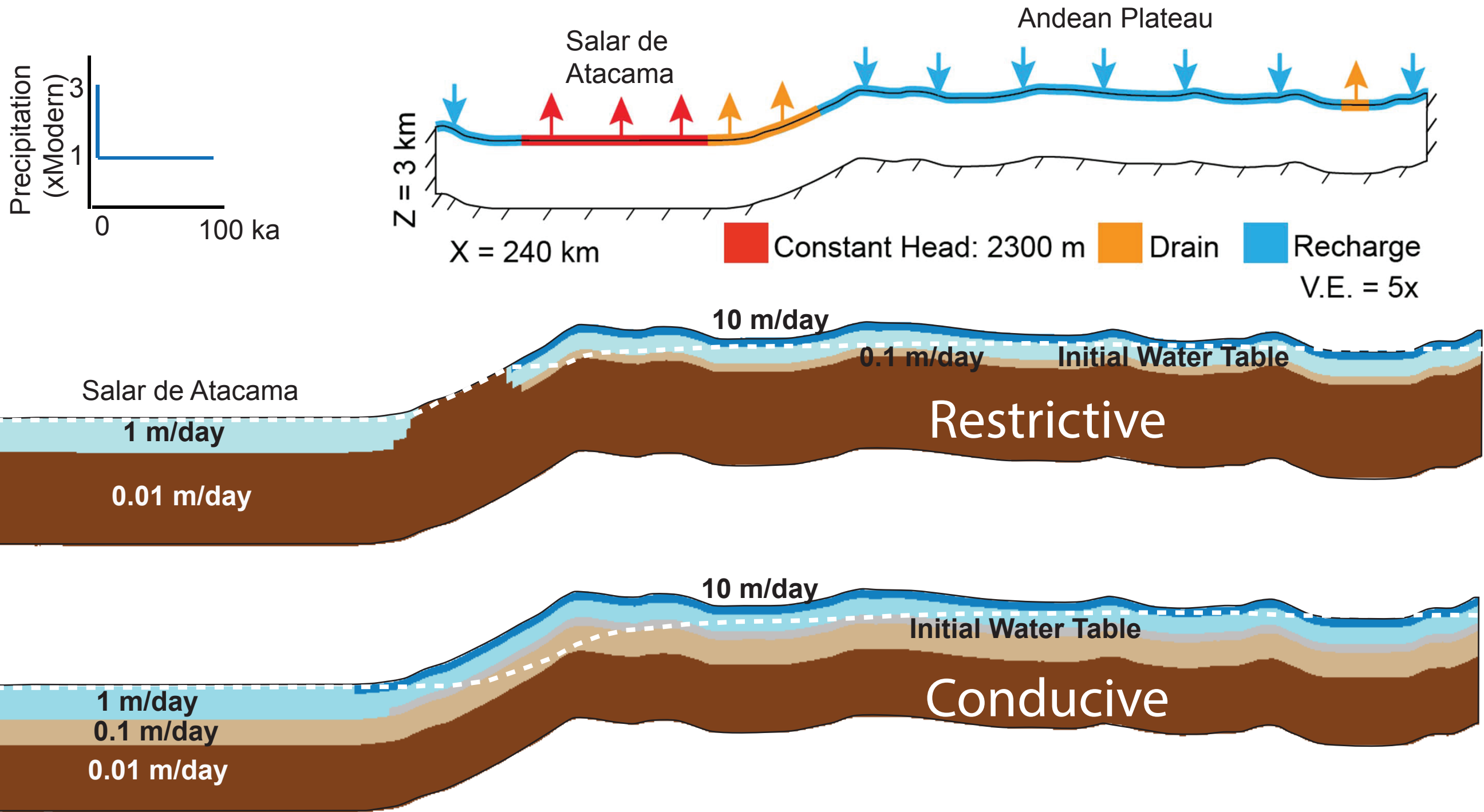


Figure 6 Model geometry, boundary conditions, hydraulic conductivity distribution, and initial water table positions for the restrictive and conductive simulations described in text.

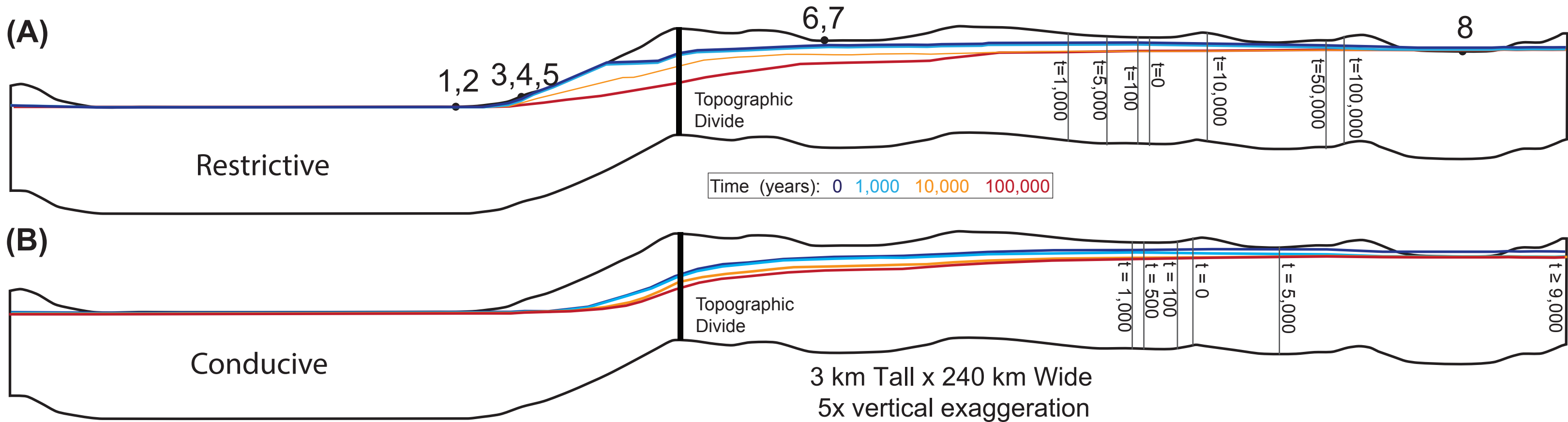


Figure 7: Simulated hydraulic head distributions for (A) restrictive and (B) conducive simulations at time of 0, 1000, 10000, 100000 years after change in recharge. Position of the hydrogeologic divides are shown for labeled times in vertical black lines. Numbered locations correspond to head observations locations plotted in Figure 8.

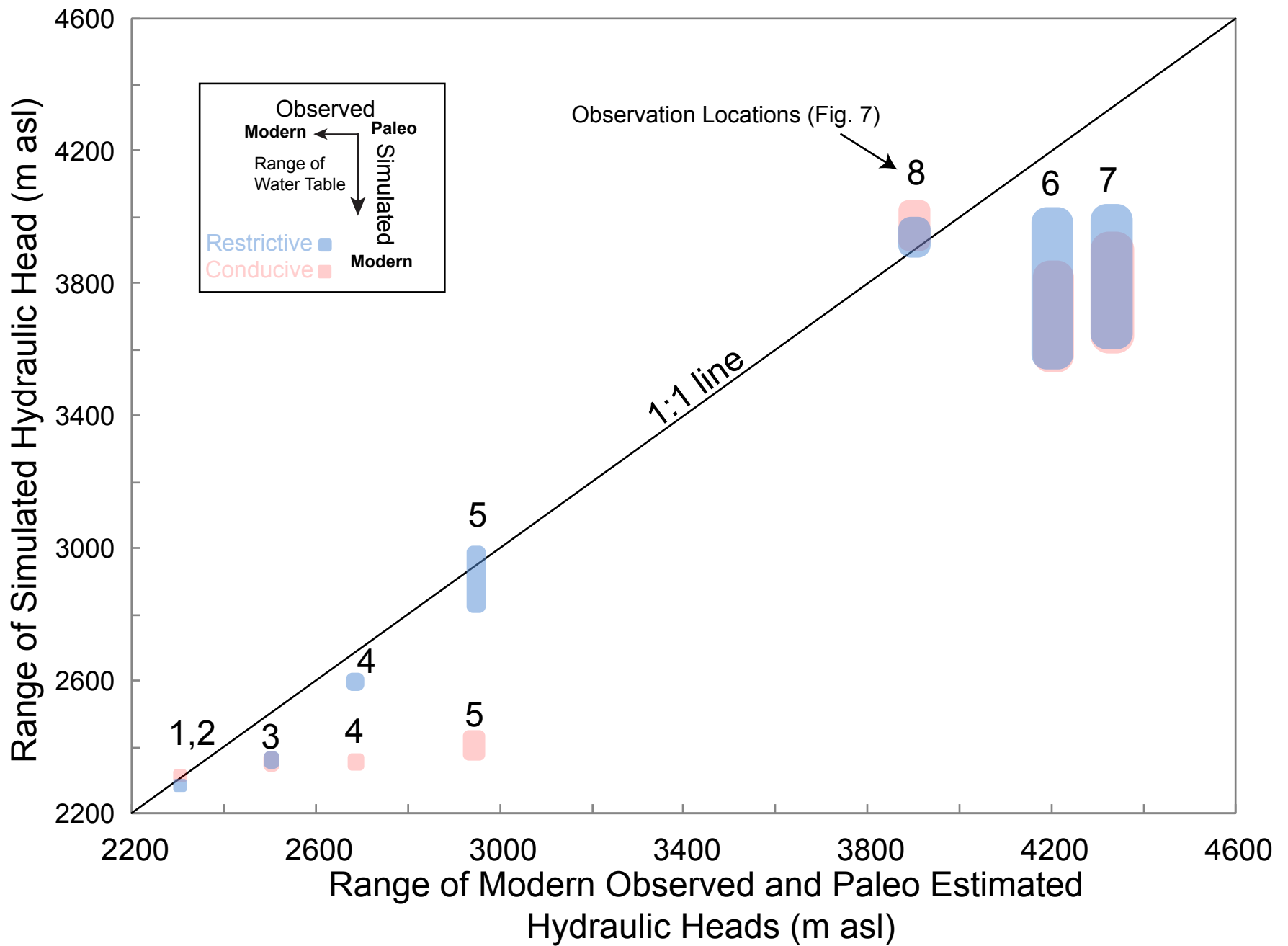


Figure 8 Simulated hydraulic heads (y-axis) for restrictive and conducive simulations compared to modern field and paleo-hydraulic head estimates (x-axis). Sizes of colored polygons (pink – conducive, blue – restrictive) are based on simulated heads with the highest position being the initial condition for the models. Highest observed heads are estimated based on paleo-hydrologic information cited in text and presented in Supporting Information 5.

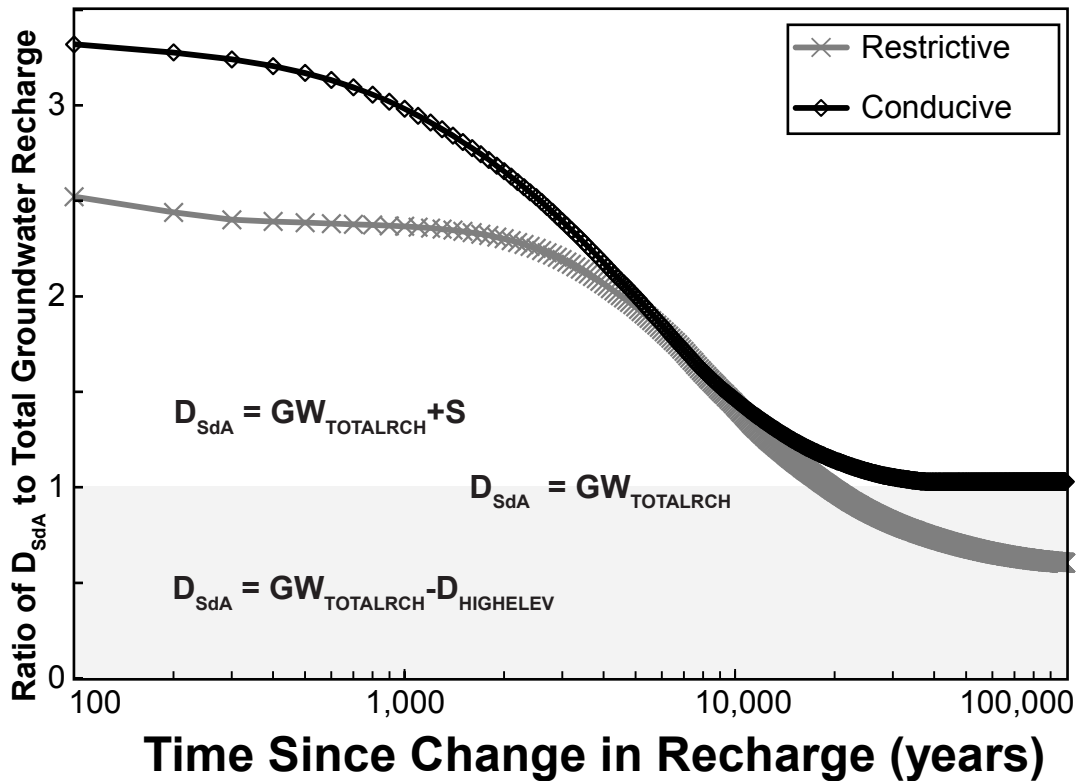


Figure 9 Plot of the ratio of discharge from constant head cells at SdA and drains along the plateau margin ( $D_{SdA}$ ) to specified total model recharge ( $GW_{TOTALRCH}$ ). The un-shaded region represents discharge to SdA that is greater than total model recharge (i.e. some contribution of groundwater storage). The shaded represents times in the simulation where discharge to SdA is less than the total model recharge.

Table 1: Locations and characteristics of precipitation samples used to determine chloride concentrations in precipitation

Sample ID	Date	Longitude	Latitude	Elevation	Cl	$\delta^{18}\text{O}$	$\delta^2\text{H}$	Reference
		WGS84		m asl	mg/l	‰ VSMOW		
SDA185W	41293	-67.8534	-23.8374	3940	9.6	-16.9	-125.7	N/A
SDA190W	41411	-68.0673	-23.6818	2381	8.1	-18.9	-137.0	N/A
SDA220W	41655	-67.8549	-23.7876	3825	15.8	-2.6	-6.6	N/A
LAC.P001	38718	-67.4450	-23.8281	4307	<10	-18.0	-135.4	Cervetto, 2012
Ascotan	1999-2000	-68.27	-21.72	3956	28	ND	ND	Houston, 2007
Colchane	1999-2000	-68.65	-19.28	3965	10	ND	ND	Houston, 2007
Collacagua	1999-2000	-68.83	-20.05	3990	4	ND	ND	Houston, 2007
El Tatio	1999-2000	-68	-22.37	4345	5	ND	ND	Houston, 2007

Table 2: Predicted Precipitation and Recharge for Topographic and Hydrogeologic Catchments

		<b>Topographic Watershed</b>	<b>Hydrogeologic Watershed</b>
Total Surface Area (km <sup>2</sup> )		17257	75924
Area of Recharge Zones (km <sup>2</sup> )		14319	69676
Precipitation	(P)	30.7 (23.4-51.7)	199.4 (171.4-284.4)
Recharge from precipitation	(GWR)	1.1 (1.1-2.1)	10.0 (9.7-14.6)
Surface water inflow	(R)	1.6 (0.5-2)	1.6 (0.5-2)
Evapotranspiration from SdA	(ET <sub>SdA</sub> )	9.5 (5.6-13.4)	9.5 (5.6-13.4)
Evapotranspiration from higher elevation salars	(ET <sub>HighElevSalars</sub> )	0	5.0 (1.8-17.8)
$\Delta$ Storage	( $\Delta$ S)	-6.8 (-11.8--1.5)	-2.9 (-21.0+9.2)



Table 3: Tabulated attributes and results of groundwater chloride analyses including estimates of groundwater recharge from

Site ID	Long.	Lat.	Elev.	Approx. Depth to Water	Date	Cl <sub>gw</sub>	Na <sub>gw</sub>	δ <sup>18</sup> O	δ <sup>2</sup> H	D-ex	P	GWR	GWR/P
	WGS84		m asl	m		mg/l	mg/l	‰	VSMOW		mm/year	mm/year	%
SDA139W	-67.988	-23.495	2568	Spring	Average	320.9	222.7	-8.0	-56.9	7.4	86	2.1 (1.3-5.1)	2.5 (1.6-5.9)
					4/3/12	207.7	231.8	-8.3	-60.0				
					9/24/12	201.9	220.5	-7.7	-56.8				
					1/11/13	555.3	225.1	-7.5	-54.8				
					5/19/13	311.6	224.0	-8.1	-55.1				
					1/17/14	327.9	212.0	-8.6	-58.0				
SDA140W	-68.050	-23.476	2340	18.5	Average	277.3	217.0	-8.2	-60.3	5.3	18	0.5 (0.3-1.3)	2.9 (1.8-7.0)
					4/3/12	224.8	210.9	-8.0	-62.4				
					9/24/12	366.5	215.6	-8.4	-63.3				
					1/11/13	319.9	238.2	-8.1	-58.7				
					5/19/13	197.9	203.2	-8.3	-56.6				
SDA161W	-68.112	-23.771	2338	23.4	Average	1579.7	913.0	-7.8	-54.3	8.2	15	0.1 (0-0.2)	0.5 (0.3-1.0)
					9/29/12	1650.2	1075.0	-7.6	-54.6				
					1/12/13	1745.9	904.9	-7.8	-54.2				
					1/13/13	1732.7	779.4	-7.9	-53.8				
					5/14/13	1949.4	904.9	-7.8	-54.2				
					1/14/14	1525.9	1001.9	-8.0	-55.9				
					8/18/14	873.8	811.9	-7.9	-53.1				
SDA186W	-67.985	-23.491	2574	21.7	Average	228.5	215.4	-8.2	-58.2	7.4	66	2.3 (1.4-5.9)	3.5 (2.2-9.0)
					1/19/13	231.3	220.1	-7.9	-57.9				
					5/19/13	225.7	210.8	-8.5	-58.5				
SDA226W	-68.137	-23.794	2329	13.3	Average	1339.9	768.5	-7.3	-50.3	8.3	17	0.1 (0.1-0.2)	0.6 (0.4-1.2)
					1/19/14	1457.5	811.6	-7.5	-52.2				
					8/18/14	1222.3	725.5	-7.1	-48.3				
SDA227W	-68.134	-23.800	2338	25.2	Average	1032.3	621.5	-8.7	-59.5	9.9	17	0.1 (0.1-0.3)	0.8 (0.5-1.6)
					1/19/14	842.1	686.9	-8.6	-58.0				
					8/18/14	1222.6	556.1	-8.8	-60.9				
SDA228W	-68.136	-23.789	2335	11.6	Average	1107.7	705.5	-8.8	-59.8	10.9	17	0.1 (0.1-0.3)	0.7 (0.5-1.5)
					1/19/14	948.0	711.2	-9.0	-60.8				
					8/18/14	1267.4	699.8	-8.7	-58.8				
SDA229W	-68.118	-23.746	2313	4.7	1/19/14	1232.9	882.3	-8.8	-61.1	9.3	17	0.1 (0.1-0.2)	0.6 (0.4-1.4)
SDA2W	-68.081	-23.671	2333	15	Average	1328.4	782.6	-7.3	-52.2	6.4	28	0.2 (0.1-0.4)	0.6 (0.4-1.3)
					9/30/11	1983.3	696.2	-7.7	-56.2				
					1/12/12	1097.1	718.6	-8.7	-63.4				
					4/8/12	1414.2	798.8	-8.6	-65.6				
					9/26/12	819.0	916.8	-8.6	-61.2				
SDA76W	-68.053	-23.365	2387	43.6	Average	369.0	340.3	-7.3	-52.2	6.5	18	0.4 (0.2-0.9)	2.2 (1.4-5.0)
					1/13/12	354.2	301.1	-7.3	-52.2				
					4/3/12	370.6	365.1	-7.5	-54.7				
					1/11/13	382.9	341.9	-7.1	-51.3				
					5/19/13	368.4	353.1	-7.4	-50.5				
SDA84W	-68.057	-23.569	2329	9.9	Average	2309.2	1460.1	-8.2	-58.4	7.0	14	0.0 (0-0.1)	0.3 (0.2-0.7)
					1/14/12	2214.0	1446.5	-8.3	-59.3				
					4/7/12	2830.9	1620.1	-8.2	-60.5				
					9/24/12	2197.2	1334.5	-8.2	-59.6				
					5/19/13	2042.3	1284.0	-7.9	-56.2				
					1/11/13	2259.0	1722.3	-8.2	-56.2				
					1/9/14	2312.0	1353.2	-8.3	-58.7				
SDA85W	-68.114	-23.780	2351	23.1	Average	1632.6	958.5	-8.3	-59.2	7.1	15	0.1 (0-0.2)	0.5 (0.3-1.0)
					1/14/12	1527.8	921.6	-8.3	-60.5				
					4/8/12	1867.5	1048.1	-8.3	-62.5				
					9/25/12	1576.7	1000.4	-8.4	-62.4				
					1/12/13	1668.3	957.3	-8.2	-57.6				
					5/14/13	1483.7	852.0	-8.2	-55.8				
					1/9/14	1671.4	971.4	-8.3	-56.5				
SDA8AW	-68.109	-23.791	2373	Spring	1/9/14	1323.1	867.0	-9.2	-64.2	9.6	17	0.1 (0.1-0.2)	0.6 (0.4-1.3)
COL.T008.1	-67.507	-23.885	4261	No Data	Average	960.0	652.0	-11.3	-81.1	9.3	90	0.8 (0.5-1.6)	0.9 (0.5-1.8)
					10/30/08	994.3	692.0	-11.2	-81.2				
					10/30/08	925.6	612.0	-11.4	-80.9				
PN.T005.1	-67.451	-23.676	4376	31.1	Average	1220.0	0.0	-11.0	-81.5	6.6	44	0.3 (0.2-0.6)	0.7 (0.4-1.4)
					11/26/04	1250.0	0.0	-11.0	-80.2				
					11/26/04	1220.0	0.0	-10.9	-81.4				
					11/27/04	1190.0	0.0	-11.1	-82.9				
PN.T006.1	-67.451	-23.693	4364	44.4	11/27/04	1220.0	0.0	-11.1	-82.0	6.9	44	0.3 (0.2-0.6)	0.7 (0.4-1.4)
PN.T007.6	-67.452	-23.692	4361	No Data	Average	1533.3	593.3	-11.2	-81.7	7.8	44	0.2 (0.1-0.5)	0.5 (0.3-1.1)
					9/1/05	1510.0	593.0	-11.2	-79.8				
					1/16/05	1540.0	597.0	-11.2	-82.8				
					1/20/05	1550.0	590.0	-11.2	-82.5				
PN.T008.2	-67.453	-23.677	4374	52.89	Average	1250.0	434.7	-11.1	-81.8	7.2	44	0.3 (0.2-0.6)	0.6 (0.4-1.3)
					12/22/04	1260.0	425.0	-11.2	-81.4				
					1/16/05	1240.0	442.0	-11.2	-82.0				
					1/20/05	1250.0	437.0	-11.0	-81.9				
PN.T014	-67.449	-23.678	4383	No Data	10/29/08	1097.1	0.0	-11.1	-81.7	7.2	44	0.3 (0.2-0.7)	0.7 (0.5-1.5)

Supporting Information for

**Imbalance in the modern hydrologic budget of topographic catchments along the western slope of the Andes (21–25°S)**

David F. Boutt<sup>1</sup>, Lilly G. Coenthall<sup>1</sup>, Scott A. Hynek<sup>2</sup>, LeeAnn Munk<sup>3</sup>,

<sup>1</sup> Department of Geosciences, University of Massachusetts-Amherst, Amherst, MA, USA

<sup>2</sup> Earth and Environmental Systems Institute and Department of Geosciences,  
Pennsylvania State University, University Park, PA, USA

<sup>3</sup> Department of Geological Sciences, University of Alaska-Anchorage, Anchorage, AK,  
USA

**Contents of this file**

Text S1 to S3

Figures S1 to S4

Tables S1 to S6

### Text S1.

**Calculation of Median, Lower and Upper Precipitation Bounds.** To incorporate uncertainty into our analysis of precipitation in the regional precipitation dataset we consider 3 instances of precipitation estimates. The median precipitation value is the processed TRMM 2B31 dataset from [Bookhagen and Strecker, 2008], which has already been evaluated against gauge data in the region and is shown to be a good estimate of precipitation. To evaluate the bias of TRMM 2B31 in this region we can compare the satellite estimates against station data from Direccion General de Aguas (DGA) (Figure S1) in Figure S2. Specific station data are provided in Table 1. While the fit between the remote sensed and station data is very good, we consider two different scenarios for estimating precipitation bias. In each case, we modify the magnitude of the spatial distribution of precipitation using the TRMM data to calculate both a lower and upper bound precipitation values. A power law is fit to the data in the region of the greatest misfit ( $< 75$  mm/yr) in the TRMM dataset. Functions fit to the lower and upper bounds are used to calculate a modified precipitation map and for the corresponding water balance calculations. For both bounds for values greater than 75 mm/yr we simply revert back to the TRMM 2B31 data. These estimates capture more than a 100% of variation in precipitation estimates.

### Text S2.

#### Expanded Groundwater Recharge Methods

Given abundant chlorine (Cl) in volcanic glass (~0.1 weight %) and biotite (~0.2 weight %) from ignimbrites in the region we allow (in the upper-end recharge scenario) for the potential that 50 mg/l of Cl<sup>-</sup> in groundwater could be sourced from rock weathering.

The basis for the  $GW_{RCH}$  estimates is data derived from over 600 water samples collected between 2011 and 2014. All samples were collected in clean HDPE bottles after passing through a 0.45  $\mu$ m filter. Samples were shipped to the University of Alaska Anchorage where all chemical analyses were performed. Sample dilutions based on specific conductance were performed prior to analysis of Cl<sup>-</sup> by ion chromatography. The isotopic composition of water samples ( $\delta^2H$ ,  $\delta^{18}O$ ) was measured by a Picarro L-1102i WS-CRDS analyzer (Picarro, Sunnyvale, CA).

Searching our water sample database revealed 9 wells and 2 springs sampled 1 to 6 times within the SdA watershed and 6 wells sampled by *Cervetto Sepulveda* [2012] in the Chilean Puna Plateau that fit our criteria for CMB calculations (Figure 2). Table 1 lists the details of the repeated sampling of each sample site and precipitation characteristics. These sites are located in recharge zones, and yield samples with stable isotopic compositions near the global meteoric water line [Craig, 1961]. These criteria minimize the influences of evaporation and salt recycling known to occur in discharge zones. The Cl<sup>-</sup> concentration was averaged for each site. Measurements of Cl<sup>-</sup> in

precipitation include 4 rain samples collected in SdA as part of this study and published measurements from the Chilean Puna Plateau [*Cervetto Sepulveda*, 2012] and Turi and Linzor region in the upper reaches of the Río Loa catchment [*Houston*, 2007, 2009].

#### **Expanded Evapotranspiration Estimates Methods**

This equation, whereby  $PET[mm] = 4367[m] - (0.59 * \text{ground elevation}[m])$ , was developed by *Houston* [2006a] based on pan evaporation from 12 meteorological stations. This equation was applied to a 30 m<sup>2</sup> resolution Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER) Global Digital Elevation Model (GDEM) to calculate gridded PET for the region. Polygons outlining the borders of fresh lakes and salars were manually constructed based on Landsat imagery, and estimates of PET were applied to permanent zones of discharge (salars and fresh lakes). The mean PET (mm/year) for each polygon (m<sup>2</sup>) is used to derive an estimated PET from each high elevation salar or lake (m<sup>3</sup>/s) (Figure S3 and Tables S2 and S3). To determine the actual evapotranspiration (AET) we compare our PET estimates to published AET estimates for SdA [*Mardones*, 1986; *Kampf and Tyler*, 2006] and Salar de Pedernales [*Johnson et al.*, 2010]. Mean annual AET is approximately 2% of PET for the lower  $D_{SdA}$  estimate (5.6 m<sup>3</sup>/s) and the Salar de Pedernales, and 8% of the upper  $D_{SdA}$  estimate (22.7 m<sup>3</sup>/s). Conservatively, we therefore consider that AET could vary from 0.5 to 8% of PET for salars in the region depending on depth to water table and other factors. We assume that AET is 80% of PET for Miniques and Miscanti Lakes (specific conductance between 7,780 and 10,640  $\mu\text{S}/\text{cm}$ ).

#### **Text S3.**

**Evaluation of the impact of Chloride Concentration in Precipitation and Precipitation magnitude on Chloride Mass Balance Recharge Calculations.** Inspecting equation 3 in the manuscript both total precipitation and the chloride concentration have a positive impact on the predicated amount of groundwater recharge. Additionally, setting the term  $Cl_{rw}$  (chloride from rock weathering) to zero we also can increase the groundwater recharge. Here, we consider the sources of uncertainty in the derived precipitation-recharge relationship in the chloride mass balance method using the median precipitation (described in S1) and an average chloride concentration of 8 mg/L and the upper precipitation bound and a high chloride concentration (16 mg/L) with no contribution of Cl from rock weathering. The resulting precipitation-recharge estimates calculated for our groundwater samples described in the text is depicted in Figure S5 along with a power law fit to the data. The estimated precipitation-recharge amounts, for observed chloride concentrations in both groundwater and precipitation fall along a distribution that is well fit by a single power law. Therefore, in the manuscript we use a single power-law to calculate recharge amounts from precipitation scenarios.

#### **Text S4.**

**Groundwater Footprint and Water Table Ratio.** The groundwater footprint [*Gleeson et al.*, 2012] of SdA, considering only non-anthropogenic discharge, is 5–21 times larger than the topographic watershed (Text S4, Figure S6, and Table S5), which ranks among the largest footprints of aquifers studied in the world. This is especially significant since

the calculations of Gleeson et al. (2012) are based predominantly on anthropogenic discharge rates. The discharge to recharge ratio (Qr:R; [Schaller and Fan, 2009]), a metric of whether a basin is a groundwater importer or exporter, indicates the topographic watershed, with a Qr:R ratio of 4.9–19.9, is a strong importer of groundwater. Modern shallow groundwater and surface water inflow to SdA reasonably balance low estimates of evapotranspiration; however, modern  $GW_{RCH}$  within the topographic watershed alone cannot explain the magnitude of these fluxes. We modify the groundwater footprint calculation [Gleeson et al., 2012], to include groundwater abstraction only from natural sources (evapotranspiration) in order to approximate the area required to support discharge rates. The groundwater footprint (GF) can be described according to Gleeson et al. [2012] by  $GF = A[ET/(GWR-R)]$ , where A is the area of interest, ET is the groundwater abstraction rate, GWR is the recharge rate and R is baseflow. For the SdA topographic watershed, we calculate a groundwater footprint of 87,850-365,121 km<sup>2</sup>, or 5-21 times the area of the topographic watershed. The range is based using median GWR and the upper and lower ranges of reasonable ET estimates.

We calculate the water table ratio (WTR) for the topographic watershed of SdA following methods outlined in Haitjema and Mitchell Bruker [2005] and Gleeson et al. [2011]. The WTR is a dimensionless criterion that describes whether the water table is likely (1) topographically controlled where the water table follows topography, or (2) recharge controlled where the water table is disconnected from topography and there is strong potential for inter-basin flow. The WTR is defined by  $\log(WTR) = \log\left(\frac{RL^2}{mKHd}\right)$ , with abbreviations explained in Gleeson et al., [2011], where a more positive  $\log(WTR)$  suggests topography controlled water tables and a more negative  $\log(WTR)$  suggests recharge controlled water tables. We estimate a range of  $\log(WTR)$  for the watershed from -3.6 to -5.3, which suggests a strongly recharge-controlled water table similar to those of the arid southwestern United States [Gleeson et al., 2012].

## Text S5.

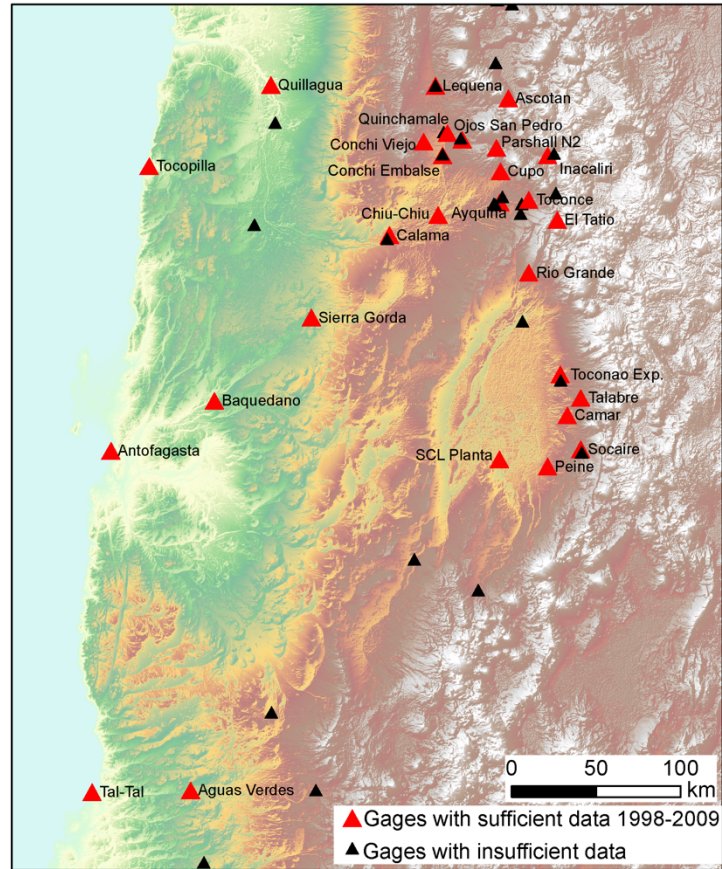
**Dynamic Response Time Calculations.** The residence time of a system is equated to its response time using a simple box model of an aquifer system to calculate the e-folding time, or the time to readjust to new boundary conditions. The above residence time estimate does not take into account the dynamics of the hydraulic response of the system (i.e. how changes in hydraulic head are propagated from the plateau to the basin). The

dynamic response time ( $\tau_{DRes}$ ) of a 1-dimensional homogenous aquifer can be

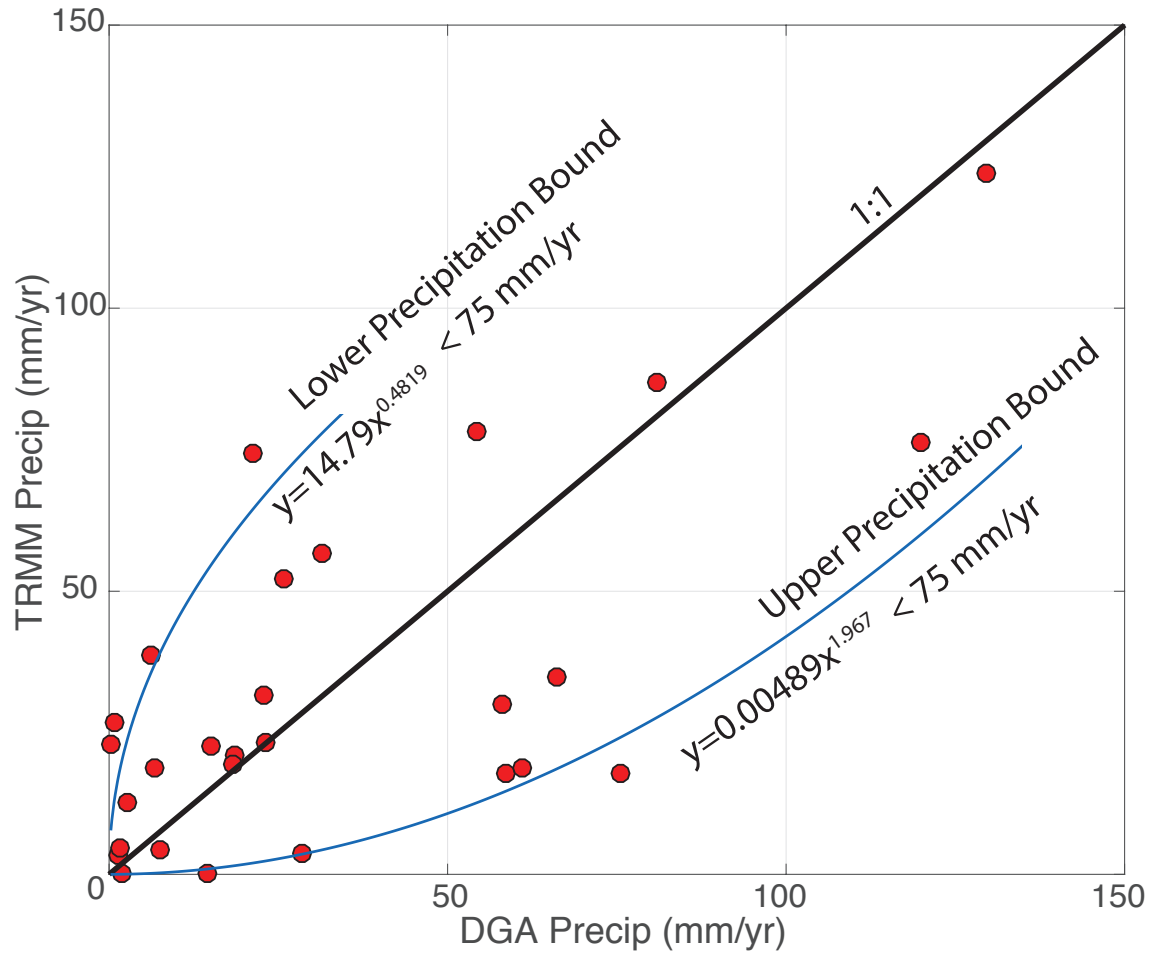
approximated as  $\tau_{DRes} = \frac{L^2}{D} = \frac{L^2 S}{bK}$ , where L is a characteristic length of the flow system

(here taken as the maximum length of a flow path), D is the hydraulic diffusivity  $D = \frac{Kb}{S}$ ,

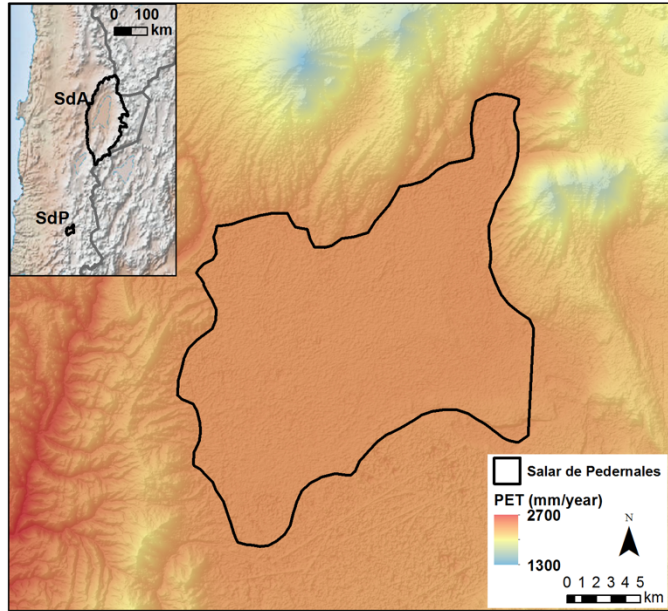
K is the hydraulic conductivity, b is the aquifer thickness (here assumed to be 500 m) and S is the aquifer storage coefficient.



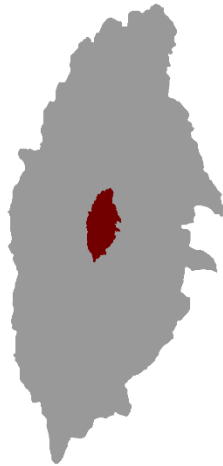
**Figure S1. Meteorological stations in the Region of Antofagasta, Chile** with sufficient data from the period of 1998-2009 (red triangles) and stations with discontinuous or discontinued measurements (black triangles). All stations are maintained by the Chilean Government’s Direccion General de Aguas (DGA), with the exception of SCL Planta that has been maintained by the Sociedad Chilena de Litio/Rockwood Lithium, Inc/Albermarle.



**Figure S2. Comparison of gage data with remotely sensed precipitation estimates.** Average annual precipitation from 1998-2009 calculated from monthly DGA raw measurements (<http://snia.dga.cl/BNAConsultas/reportes>) and one measurement from the Sociedad Chilena de Litio/Rockwood Lithium Inc. meteorological station. TRMM 2B31 estimate of precipitation from the TRMM 2B31 dataset [Bookhagen and Strecker, 2008] for sites in the Region of Antofagasta, Chile. Power functions are fit to calculate lower and upper bound precipitation maps for water balance calculations.



**Figure S3. Map of PET computed by equation (a) in the Salar de Pedernales (SdP) region.** Background is an ASTER DEM. SdP border modified from *Johnson et al.* [2010]. The SdP has a surface area of 315 km<sup>2</sup>, average elevation of 3356 m asl and average PET of 2384 mm/year (standard deviation of 6 mm/year).



**Figure S4. Conservative groundwater footprint of the SdA topographic watershed.** The red zone is the topographic watershed area, and the gray shaded region is its inferred groundwater footprint based on a groundwater discharge rate of 5.6 m<sup>3</sup>/s.



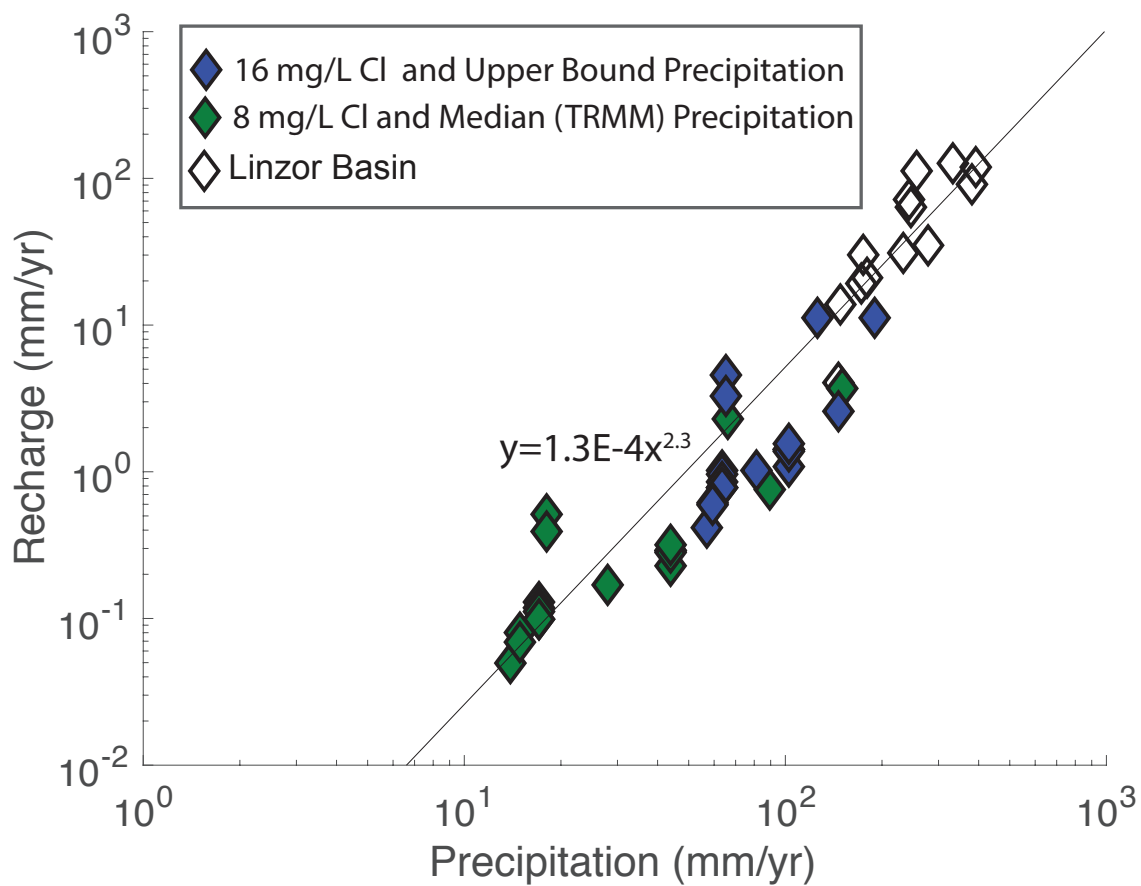


Figure S5. Precipitation-Recharge Relationship from Chloride Mass Balance Calculations.

Meteorological Station	Easting	Northing	Elevation	Distance from SdA	Precipitation	
					Gage	TRMM 2B31
	WGS84		m asl	km	mm/year	mm/year
Aguas Verdes	403389	7190650	1600	270	6.7	18.7
Antofagasta	358725	7389982	50	220	2	0.2
Ascotan	575136	7597754	3956	200	66.2	34.9
Ayquina	570227	7536538	3031	140	31.4	56.6
Baquadano	414749	7419946	1032	160	1.5	4.6
Calama	509841	7517409	2260	130	2.8	12.7
Camar	606276	7411224	3020	30	25.9	52.3
Chiu-Chiu	536440	7529250	2524	140	6.3	38.7
Conchi Embalse	539003	7564490	3010	170	14.5	0.1
Conchi Viejo	528514	7572609	3491	180	28.6	3.7
Cupo	570641	7554915	3600	160	81	86.8
El Tatio	601729	7526160	4320	130	129.7	123.7
Inacaliri	596588	7564208	4100	170	119.8	76.4
Lequena	535139	7605268	3320	210	61	18.7
Ojos San Pedro	568440	7568716	3800	170	58	30.0
Parshall N2	549805	7573477	3318	180	23.1	23.1
Peine	595346	7381030	2480	20	18.6	20.9
Quillagua	444822	7605629	802	250	0.4	23.0
Quinchamale	541684	7577572	3020	180	18.3	19.6
Rio Grande	585833	7495117	3250	100	58.7	17.9
SCL Plant	569278	7385349	2300	0	15.1	22.7
Sierra Gorda	467247	7468888	1616	130	0.9	26.8
Socaire	613485	7391129	3251	40	22.8	31.7
Talabre	613735	7421435	3600	40	54.4	78.1
Tal-Tal	350886	7189130	9	310	7.5	4.4
Toconao Exp.	602581	7435191	2430	40	21.3	74.3
Toconce	586111	7537991	3350	140	75.6	17.9
Tocopilla	378070	7557678	45	250	1.3	3.4

**Table S1. Comparison of average annual precipitation between gage and remotely sensed sources** from 1998-2009 from DGA gage measurements (<http://snia.dga.cl/BNAConsultas/reportes>) and one measurement from the Sociedad Chilena de Litio meteorological station (SCL Planta) with gridded precipitation from the TRMM 2B31 dataset [Bookhagen and Strecker, 2008] for the Region of Antofagasta, Chile.

Salar	Surface Area	Average Elevation	AET	Mean PET	Standard Deviation	Mean PET	AET/PET	Reference for AET estimate
	km <sup>2</sup>	m asl	m <sup>3</sup> /s	mm/year	mm/year	m <sup>3</sup> /s	%	
Salar de Atacama	2750	2313	5.6	2999	13	262	2	Mardones, 1986 / DGA, 2010
	2864	2313	22.7	2999	13	272	8	Kampf and Tyler, 2006
Salar de Pedernales	315	3356	0.58	2384	6	24	2	Johnson et al., 2010

**Table S2. Actual evapotranspiration as a fraction of potential evapotranspiration derivation for the Salar de Pedernales and Salar de Atacama.**

Zone	Area (km <sup>2</sup> )	Mean PET (m <sup>3</sup> /s)	Mean AET (m <sup>3</sup> /s)			Zone	Area (km <sup>2</sup> )	Mean PET (m <sup>3</sup> /s)	Mean AET (m <sup>3</sup> /s)			
			2%	0.5%	8%				2%	0.5%	8%	
B	1.3*	8.2E-02	6.5E-02	6.5E-02	6.5E-02	J	3.3	1.8E-01	3.6E-03	9.0E-04	1.4E-02	
	12.8*	7.8E-01	6.2E-01	6.2E-01	6.2E-01		1.2	6.4E-02	1.3E-03	3.2E-04	5.1E-03	
	1.8	1.0E-01	2.0E-03	5.1E-04	8.2E-03		34.1	1.9E+00	3.7E-02	9.3E-03	1.5E-01	
C	112.6	6.7E+00	1.3E-01	3.4E-02	5.4E-01		22.9	1.2E+00	2.5E-02	6.1E-03	9.8E-02	
	13.3	7.2E-01	1.4E-02	3.6E-03	5.8E-02		4.9	2.6E-01	5.3E-03	1.3E-03	2.1E-02	
	14.9	8.6E-01	1.7E-02	4.3E-03	6.9E-02		6.9	3.8E-01	7.6E-03	1.9E-03	3.0E-02	
D	11.8	7.7E-01	1.5E-02	3.8E-03	6.1E-02		1.8	1.0E-01	2.0E-03	5.0E-04	8.0E-03	
	22.2	1.4E+00	2.9E-02	7.2E-03	1.2E-01		33.5	1.9E+00	3.7E-02	9.3E-03	1.5E-01	
	13.5	8.8E-01	1.8E-02	4.4E-03	7.0E-02		K	81.0	5.3E+00	1.1E-01	2.6E-02	4.2E-01
	29.6	2.1E+00	4.2E-02	1.1E-02	1.7E-01		19.1	1.3E+00	2.5E-02	6.3E-03	1.0E-01	
E	3.0	2.2E-01	4.3E-03	1.1E-03	1.7E-02	141.5	9.3E+00	1.9E-01	4.6E-02	7.4E-01		
	1.9	1.1E-01	2.3E-03	5.7E-04	9.1E-03	2.1	1.1E-01	2.3E-03	5.7E-04	9.1E-03		
	2.6	1.6E-01	3.3E-03	8.2E-04	1.3E-02	1.9	1.0E-01	2.0E-03	5.1E-04	8.1E-03		
F	11.9	8.7E-01	1.7E-02	4.4E-03	7.0E-02	2.1	1.1E-01	2.3E-03	5.7E-04	9.2E-03		
	15.5	9.2E-01	1.8E-02	4.6E-03	7.3E-02	L	113.2	8.3E+00	1.7E-01	4.2E-02	6.7E-01	
G	8.3	4.9E-01	9.9E-03	2.5E-03	3.9E-02	1091.5	8.0E+01	1.6E+00	4.0E-01	6.4E+00		
	71.4	4.3E+00	8.6E-02	2.1E-02	3.4E-01	154.0	1.1E+01	2.2E-01	5.4E-02	8.6E-01		
	39.2	2.3E+00	4.5E-02	1.1E-02	1.8E-01	12.1	7.6E-01	1.5E-02	3.8E-03	6.1E-02		
	0.6	3.2E-02	6.4E-04	1.6E-04	2.6E-03	20.8	1.4E+00	2.9E-02	7.2E-03	1.1E-01		
	1.3	6.4E-02	1.3E-03	3.2E-04	5.2E-03	M	119.4	8.3E+00	1.7E-01	4.2E-02	6.7E-01	
	1.2	6.2E-02	1.2E-03	3.1E-04	5.0E-03	5.4	2.9E-01	5.7E-03	1.4E-03	2.3E-02		
	105.0	5.9E+00	1.2E-01	3.0E-02	4.7E-01	12.8	6.9E-01	1.4E-02	3.4E-03	5.5E-02		
H	58.1	4.8E+00	9.6E-02	2.4E-02	3.8E-01	115.7	6.4E+00	1.3E-01	3.2E-02	5.1E-01		
	51.8	3.0E+00	6.0E-02	1.5E-02	2.4E-01	1.0	5.2E-02	1.0E-03	2.6E-04	4.2E-03		
I	0.9	5.0E-02	1.0E-03	2.5E-04	4.0E-03	2.9	1.6E-01	3.2E-03	8.0E-04	1.3E-02		
	2.8	1.5E-01	3.0E-03	7.6E-04	1.2E-02	0.8	4.7E-02	9.4E-04	2.3E-04	3.8E-03		
	0.6	3.2E-02	6.4E-04	1.6E-04	2.5E-03	1.5	8.2E-02	1.6E-03	4.1E-04	6.5E-03		
	1.8	9.4E-02	1.9E-03	4.7E-04	7.5E-03	3.7	2.1E-01	4.2E-03	1.0E-03	1.7E-02		
	16.9	9.1E-01	1.8E-02	4.5E-03	7.3E-02	5.4	3.0E-01	6.1E-03	1.5E-03	2.4E-02		
	1.9	1.0E-01	2.0E-03	5.0E-04	8.0E-03	N	247.5	2.1E+01	4.1E-01	1.0E-01	1.6E+00	
	0.2	1.1E-02	2.2E-04	5.5E-05	8.8E-04	19.9	1.4E+00	2.8E-02	7.0E-03	1.1E-01		
	5.2	3.0E-01	6.0E-03	1.5E-03	2.4E-02	7.3	4.3E-01	8.6E-03	2.2E-03	3.4E-02		
	15.3	9.5E-01	1.9E-02	4.8E-03	7.6E-02	0.5	3.1E-02	6.1E-04	1.5E-04	2.5E-03		
	7.3	3.7E-01	7.5E-03	1.9E-03	3.0E-02	98.6	7.1E+00	1.4E-01	3.6E-02	5.7E-01		
358.8	2.5E+01	4.9E-01	1.2E-01	2.0E+00	24.4	1.6E+00	3.2E-02	7.9E-03	1.3E-01			
J	120.5	8.2E+00	1.6E-01	4.1E-02	6.6E-01	O	144.9	9.8E+00	2.0E-01	4.9E-02	7.9E-01	
	0.4	2.2E-02	4.3E-04	1.1E-04	1.7E-03	0.6	3.4E-02	6.8E-04	1.7E-04	2.7E-03		
	2.2	1.1E-01	2.2E-03	5.6E-04	9.0E-03	0.8	4.1E-02	8.2E-04	2.1E-04	3.3E-03		
	0.9	4.7E-02	9.3E-04	2.3E-04	3.7E-03	2.5	1.4E-01	2.7E-03	6.8E-04	1.1E-02		
	0.8	4.0E-02	7.9E-04	2.0E-04	3.2E-03	0.6	3.3E-02	6.6E-04	1.7E-04	2.7E-03		
	12.5	6.9E-01	1.4E-02	3.5E-03	5.5E-02							

**Table S3. Summary of land type, surface area, mean annual potential evapotranspiration (PET) and actual evapotranspiration (AET) for the discharge zone polygons in all considered watersheds.** Each lettered watershed includes all cumulative discharge zones in the smaller watersheds. Starred areas in watershed B are Miscanti and Miniques lakes where AET is assumed to be 80 % of PET.

<b>Variable</b>	<b>Abbreviation</b>	<b>Value</b>
Surface area (km <sup>2</sup> )	A	17,257
Discharge (m <sup>3</sup> /s)	ET	5.6 to 22.7
Recharge (m <sup>3</sup> /s)	GWR	1.1
Baseflow contribution (m <sup>3</sup> /s)	R	0
Groundwater Footprint (km <sup>2</sup> )	GF	87,850 to 356,120
Groundwater stress indicator	GF/A	5 to 21

**Table S4.** Groundwater footprint calculations for the SdA topographic watershed. Values are specific to SdA and variables and calculations described in *Gleeson et al.* [2012]. Discharge estimates from Mardones [1986] and *Kampf and Tyler* [2006]. We consider an R of 0 consistent with the more conservative water budget conceptualization.

<b>Description</b>	<b>Abbreviation</b>	<b>Value</b>
Areal recharge rate (mm/year)	R	2.5
Distance between surface water bodies (km)	L	10
Hydraulic conductivity (m/day)	K	1 to 10
Average vertical extent of groundwater flow system (m)	H	100 to 500
Maximum terrain rise (m)	d	3700
Constant (unitless)	m	8
log Water Table Ratio	log(WTR)	-3.6 to -5.3

**Table S5.** Water Table Ratio for the SdA topographic watershed. The average areal recharge rate is calculated based on the raster of GWR presented in Figure 3b, and the maximum terrain rise is derived from an ASTER DEM. Values approximate bulk aquifer properties for the SdA. We copy the values presented in *Gleeson et al.* [2011] for distance between surface water bodies, and more conservative estimate for the average vertical extent of the flow system.

Observation Point ID	Hydraulic Head Constraints (m asl)		Simulated initial heads (m asl)		Total change in head over simulation (m)		Reference for Hydraulic Head Constraints	Notes
	Modern	Paleo	Restrictive Case	Conductive Case	Restrictive	Conductive		
1	2309	2314	2311	2315	<1	<1	Field Measured Water Levels	Paleo Head of +5 m is estimated based on position relative to Tulan Wetlands
2	2310	2315	2311	2315	5	5	Field Measured Water Levels	Paleo Head of +5 m is estimated based on position relative to Tulan Wetlands
3	2498	2509	2335	2328	20	10	Betancourt et al., 2000 - Tulan Wetlands	Rio Tulan deposits showed a rise of ~11 m above current levels between 8.2 and 3.0 ky BP, Observed heads based on approximate location of modern spring discharge
4	2692	2702	2602	2345	25	15	Betancourt et al., 2000 - Taranje Wetlands	Tarajne paleowetland deposits are at a higher elevation than the Tulan deposits. Taranje deposits date between 15.4 and 9.0 ky BP, Observed heads based on approximate location of modern spring discharge
5	2960	2975	2964	2390	180	25	Springs at Imilac and Punta Negra Quade et al 2008 - Midpoint ground elevation between the Salar de Imilac (2970 m asl) & Salar de Punta Negra (2950 m asl)	Sdl & SdP are south of SdA, so approximate elevations are projected onto a similar elevation on east side (where the model geometry was derived), note that Imilac springs are fault controlled according to Quade, Observed heads based on approximate location of modern spring discharge
6	4210	4240	3980	3842	460	350	Cervetto (2012) Wells - LA and LAAR wells , Grosjean et al., 1995	Water Elevations in LA and LAAR series wells from Anexo G of Cervetto's thesis range from 4208-4216m, Laguna Tuyajito esimates of +20-40 m
7	4320	4345	4005	3916	460	350	Cervetto (2012) Wells - PN and PNAR wells , Grosjean et al., 1995	Water Elevations in PN series wells from Anexo G of Cervetto's thesis range from 4320-4322m, Aguas Calientes IV +25 m
8	3900	4000	3993	4008	100	100	Modern Levels - Salar de Olaroz Ground elevation is 3900m ( <a href="https://www.orocobre.com/PDF/NI%2043-101_Technical%20Report-Oloroz%20Project.pdf">https://www.orocobre.com/PDF/NI%2043-101_Technical%20Report-Oloroz%20Project.pdf</a> )	Paleo high stands of ~ 100 m (Grosjean et al., 1995; Placzek et al., 2006, 2013)

**Table S6.** Simulation results and estimates of modern and paleo hydraulic heads for specific places within the model domain. Sources of estimates and notes discussing them in 2 right-hand columns of table.