

Environmental Lithium Exposure in the North of Chile—I. Natural Water Sources

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Abstract Lithium as an essential element for human life is still a subject of controversy. However, it is accepted that it does have profound neurological effects and is a valuable treatment for bipolar disease. Generally, it occurs in barely trace amounts in groundwater with few major exceptions. One of these is the Northern area of Chile where all potable water and many of the food stuffs contain high levels of lithium; between 100 and 10,000 times higher than most rivers in North America. Inevitably, the local population has

been exposed to these levels in their drinking water for as long as the region has been populated. The present report details lithium levels in all the surface water sources of Northern Chile with comparison to that elsewhere. The implications for the local population are discussed and their situation compared to those exposed to other sources of lithium pollution.

Keywords Lithium · Water · Northern Chile

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Introduction

Lithium has become the “magic ion” [1] with major implications for mining, manufacturing, and medicine. Driven by the need for green energy, miners and manufacturers of lithium batteries have developed an insatiable appetite for new sources of lithium [2]. Similarly, medical scientists have now found lithium to be the most potent neurogenic agent in the world capable of stimulating new brain cell growth with the potential to heal nerve injury and possibly prevent Alzheimer’s disease [1].

Despite these vital needs and important clinical findings, we still know almost nothing about the effects of lithium on normal people particularly in terms of brain function. And in truth, the normal levels of lithium in the normal human body are so low they are called “ultra-trace”. Indeed, Cade [3], the Australian psychiatrist who discovered lithium as a treatment for bipolar disease in 1949, said that “mankind or at least the non manic depressive members could live perfectly happily and efficiently in a lithium free universe” [4] since the levels of lithium are normally barely measurable [5].

It is therefore not surprising that there have not been epidemiological studies of normal populations carrying high lithium loads purely from natural environmental sources. As the clearance rate of lithium is so fast (the plasma elimination half-life of a single dose of lithium is from 12 to 27 h (varies with age)) [6], such populations would have to live where everything was literally “soaking” in lithium. One such place is in the extreme north of Chile. We learned this 3 years ago after reading a somewhat obscure one page “Letter to the Editor” by Professor Roberto Zaldivar in 1980 [7], cited in a few review articles [8–10],^{1,2}. Zaldivar [7] described a series of small villages in two valleys in the very north of Chile where the lithium blood levels were exceptionally high due to elevated levels of lithium in the drinking water.

In 2009, we went to the area studied by Zaldivar to find more information about these villages and uncover, if possible, more data. The area of interest is in what is now known as Region XV and consists of three valleys all within 100 miles of the Peruvian border. The main and only city in the region is Arica and that is where we began our

investigative efforts. The University of Tarapaca, the provincial branch of the University of Chile, seemed to be a logical starting point so we invited Professor Leo Figueroa of the Department of Chemistry for dinner explaining our interest beforehand. He arrived with a book with a somewhat obscure title “The Aymara, Strategies in Human Adaptation to a Rigorous Environment” [11] and referred us to Table 1 in Chapter 3 [12]. This revealed a tabular array by location and year from 1973 to 1979 in lithium water concentrations in the same area studied by Zaldivar. Professor Figueroa, however, had never heard of Zaldivar. He explained that his work was done with the multinational Multi-Andean Health and Genetics Program (MAHGP) headed by Prof. Wm Schull of the University of Texas³. The aim of the MAHGP was to assess the methods of adaptation to the hypoxia of high altitude. The MAHGP independently discovered the high lithium levels in 1979 as part of their environmental assessments done solely in the larger context of the high altitude study. Lithium levels were measured [12] but their potential attributable effects were never specifically assessed. The same was true of a limited collaborative study done several years later by Professor Ronnie Barr from McMaster University in Canada in conjunction with scientists from the National Institute of Standards Technology in the USA. They too came to the same study areas as Zaldivar and Schull, documented the increased

³ The MAHGP’s aim was to determine the strategies involved “human adaptation to a rigorous environment”. The environment under study was the Andean altiplano and the subjects were largely the Aymara who populated that area in altitudes largely above 4,000 m in southwest Bolivia, northwest Argentina, and northwest Chile. The multinational and multidisciplinary study team assembled in 1972 involved scientists from Bolivia (Universidad de San Andres), Chile (Universidad de Chile, Universidad de Norte—now the Universidad of Tarapaca, Junta de Adelanto de Arica, and the Servicio Nacional de Salud), Ecuador and Peru (Universidad de San Marcos), Argentina (University de Juay), and the USA (Mayo Clinic, University of Michigan, and the University of Michigan and the University of Texas Health Science Centre at Houston). Scientific teams from these various institutes visited hundreds of villages and examined more than 2,200 people in the Bolivian, Argentinian, and Chilean altiplano. The examinations included detailed interviews and examinations and all findings were coded and entered into detailed questionnaire proformas. The data collected included information that could potentially shed light on the possible genetic and environmental factors responsible for the adaptation to hypoxia at high altitude. The study produced hundreds of papers most of which are described in the book which describes the multinational effort “The Aymara, Strategies in Human Adaptation to a Rigorous Environment” [11]. Mueller et al. [46] described the Chilean “study segment” that began in 1972 as a multidisciplinary study, which sought to assess the health status of the indigenous peoples of the Department of Arica in northern Chile, the Aymara, and to related disease, morphological, physiological, and biochemical variation to the wide changes in altitude of the region.

¹ Schrauzer [9] and Aral and Vecchio [10] said that the people in this area had no adverse effects from the high lithium levels but admitted there were no studies to base this upon.

² We tried to find Zaldivar to learn more about his work but to no avail. He left Chile when Pinochet came to power and effectively disappeared.

water and blood lithium levels in a small number of the affected villages, but failed to evaluate the area epidemiologically.

Later in 2009, Prof. Figueroa introduced us to Drs. Schull and Barton at the University of Texas who then provided us with original MAHGP study data. Lithium exposure data from the north of Chile have only been published in a few, small, mostly obscure reports [5, 7, 12–16]; and the remaining extensive datasets remain unpublished. Given the intense interest in lithium, we have gathered all the published information we could find and much that is unpublished to produce a comprehensive critical analysis of the lithium water level data from the north of Chile; an area which, largely, is known as the Atacama desert.⁴

Materials

Data Sources Many of the materials that make up this report are unpublished. They were produced in the context of the MAHGP from 1973 to 1979 and provided to us by Dr. Sara Ann Barton of the University of Texas⁵ and Professor Leo Figueroa of the University of Tarapaca (UTA). Two other professors, formerly with UTA, Dr. Blago Razmilic and Dr. Oscar Zumeata also contributed prominently to the dataset. A few recent data were also obtained from the Direction Gral de Aguas with the help of Professor Camilo Albina of the UTA. Additional information per source data came from discussions with Prof. Ann Marie Sancha, Director, Facultad Ciencias Fisica, University of Chile. Work by Kamiya and Ilgren

provided data in 2009.⁶ Unpublished data from Zaldivar and Barr were not available (also see table legend reference codes for Tables 1, 2, 3, 4, 5, 6, and 7 for additional sources of source material). Virtually all of the data referable to San Pedro de Atacama were produced by Dr. Hugo Alonso of the University of Antofagasta. Some of the San Pedro samples were also collected by Dr. Barton and analyzed by Dr. Barr.

⁶ This desertic landscape, located on the west coast of South America, extends for more than 3,000 km along a narrow strip from northern Peru (latitude, 58 S) to northern Chile (latitude 278 S). It owes its existence to the drying effect of the cold northward flowing Humboldt Current, the existence of air masses associated with the subtropical high known as the South Pacific Anticyclone, and to the rain shadow effect of the Andean mountain ranges, which impedes the penetration of moisture carried by the eastern trade winds. Although this desert is continuous from Peru to Chile, it is usually broken into two main components. The Peruvian Coastal Desert extends from Tumbes (ca. 58 S) to Tacna in southern Peru (ca. 188 S), and the Atacama Desert from the area of Arica in northern Chile (ca. 188 S) to Copiapo (ca. 278 S). The northern part of the Peruvian Coastal Desert consists of a wide coastal plain with shifting sands known as the Sechura Desert, some 100–150 km wide. During the terminal Pleistocene and Holocene, this coastal desert provided more favorable living conditions in the form of spring water and shallow lagoons with fresh water, as documented in Pamp a de los F6 siles in the Cupisnique Desert, of northern Peru. Further south in the Peruvian Coastal Desert, thick cloud banks form over the desert lands as a result of the cooling effect of the Humboldt. When intercepted by isolated mountains or steep coastal slopes, this cool moist air gives rise to a fog zone known as garúa in Peru and Camanchaca in Chile. This moisture allows for the development of isolated and diverse vegetation formations called lomas (small hills), where plants are adapted to condense this humidity. The lomas, comprising communities of annual and perennial plants (Herbaceae and Gramineae) and cacti, grow vigorously during a short period in the winter of the Southern Hemisphere, and attract a wide variety of animals and birds (camelids, rodents, foxes). These ephemeral “fog oases” were occupied seasonally by hunter-gatherers whose base camps were located on the nearby coast. The Atacama Desert, located between 188S and 278S, can be divided in three sections: north, central, and south. The latter, being the driest, has negligible archeological data and thus is not described herein. Flanked to the east by the high Coastal Cordillera, the littoral offers discrete bays and beaches for human habitation, experiencing a mild climate. In contrast to other Southern Hemisphere deserts (such as in Australia and southern Africa), average yearly precipitation is near zero. Thus, the supply of fresh water depends on rain events outside the desert caused by convective air masses that cross the Andean crest bringing moisture laden air across the altiplano from the Amazon Basin. This phenomenon of the austral summer known as the invierno boliviano (the Bolivian or altiplanic winter) is responsible for the radical fluctuation of superficial runoff and groundwater that typically flows from the western slopes of the Andes to the Pacific. In the northern Atacama, the runoff creates narrow and deep quebradas separated by 20–30 km of barren terrain with no vegetation at all. As rainfall gradually declines toward the south, the quebradas do not reach the ocean, but rather discharge into inland basins, such as the Pampa del Tamarugal and Salar d’Atacama in the central Atacama. The southern Atacama (248–278S) is a territory with no human habitation until recently, known as the despoblado (depopulate) d’Atacama. In sum, although terrestrial biomass production is scanty and sparse, the existing water network, a distinctive feature of the Atacama Desert, offers a predictable resource for hunter-gatherers and fishermen [47].

⁴ The north of Chile, extends from the Peruvian border 2,000 miles to the south to Copiapo. It is mostly the Atacama desert known to be the driest area on earth. The coastal shores are characterized largely by pink cliff faces that rise to heights of 500–600 m along the bottom of which are sea eroded terraces on which the few towns and cities in the area have been built. Eastward, the land rises rapidly into the precordilleras (ca 3,000 m) and ultimately to the altiplano (>4,000 m) which extends into Bolivia. Chile’s eastern frontier follows the crests of the Andean Western Cordillera (5,000–6,700 m) occasionally broken by passes leading into Bolivia or Argentina. Salars (salt flats) stud the Altiplano particularly in the south. Few small Salars are found in the study area. These are apparently unconnected to the three river systems which form the primary part of this investigation. Rivers and streams originate in the cordillera and the precordilla to the east and follow valleys or quebrada. Few watered valleys reach the sea. Overall, the majorities of settlements are in the altiplano, the precordilleras, and are associated with the few modest rivers that include for the most part Rios Camerones, Copiapo, Lluta, Loa, and San Jose. Calama is the only community of consequence in the interior; Antofagasta, Arica, Chanaral, Iquique, Taltal, and Tocopilla are all on the coast.

⁵ Barton took more than 150 samples; also had discussions at the time (1979) with the Director of the Water Department in Arica, Sr. Ruben Vellozo Retamel, Jefe Subrogante, Direction Gral de Aguas (DGA), 1a Regio, Dept. Hidrologia.

In 2009, Kamiya and Ilgren attempted to collect samples where Dr. Raul Salas believed the Camerones system originated at the start of a hydraulic basin found along several now exhausted geyser areas in the region of which were three small lakes (red, yellow, and green). Each lake was at least 100 m in diameter in close proximity to each other ca 50 km south of the Salar de Surire and displayed evidence of thermal activity and some areas of an oily sheen. The lakes drained into the nearby Rio Caritaya which shortly thereafter was blocked by an artificial dam (Tranque de Caritaya) to protect against severe seasonal storms.

Geographic Reach The geographic reach of the data presented herein extends from Lima to Santiago though the majority concern the “study area” in Region XV in the north of Chile. The few Peruvian data are mostly from the very south of the country geophysically similar to the study area in north of Chile both being part of the Atacama (see endnote 6 for a description of the contiguous nature of these areas).

Primary Study Area The primary area of interest is the study area in the province of Tarapaca, department of Arica in Region XV (formerly Region I). This area was originally studied by Zaldivar [7, 13, 14], Schull and the MAHGP [12], and Barr et al. [16]. It is of particular interest since it has been the most carefully studied. It consists of three valleys, Valle Azapa (Table 1; Fig. 1), Valle Lluta (Table 2; Fig. 1), and Valle Camerones (Table 3; Fig. 1) in which communities under study are situated as well as the Lauca Precordillera (Table 4; Fig. 2).

Other Study Areas The other study areas in the north of Chile outside Region XV were not widely sampled and include villages and riverines from Regions I (Table 5; Fig. 3) and II (Table 6; Fig. 3). San Pedro is included along with small villages, lakes, and small salars near the Salar d’Atacama (Table 7; Fig. 3; see below).

Temporal Spread The temporal spread of the data covers more than 40 years as sampling went from the late 1960s [13] to 2009 (Kamiya, Ilgren, and Figueroa, unpublished data).

Water Sources The coastal communities, for the most part, as well as the hamlets and towns of the altiplano and sierra, derive their waters from surface sources; the latter are generally rivers or streams but occasionally springs. Most villages used river and stream water in 1979. Given the scarcity of water, some coastal communities such as Iquique were obliged to obtain their water from sources far removed from the town. In such cases, water was piped for example from the springs which support Pica, a fertile oasis 80 km to the east. In other cases, more elaborate diversions were constructed particularly in the headwaters of the valley to supply critical water needs (see “Discussion” below).

Water Sampling Methods Zaldivar and Barr did not provide information on their water sampling methods. The MAHGP’s methods for taking water samples for the 1979 Trace Metals Study were described in great detail in the unpublished materials provided to us in 2009 and 2010. In brief, the water sampling included a determination of the exact locations of the raw water source(s), pumping station (s), and treatment facilities for each sampling site. Questions on each water sample sheet were also answered⁷ as were those on a second questionnaire completed for the local authorities (ENDESA, DOS, and DIEGO).⁸ Kamiya and Ilgren took samples from each of the three lakes including water, rocks and nearby bones. These were placed into clean

⁷ This included the following: (1) city, village; (2) data and hour sample collected; (3) name of the nearest village, highway; (4) place sample collected as river, channel, or water tap; (5) name of the water source from which the sample was collected; (6) name of the water sources for the village; (7) the length of time the village had been using the sources of water; (8) if water was drawn from wells, how was the water drawn; (9) if the water was piped, from what material were the pipes made; (10) if the water was treated, what was the treatment process; (11) the date and amount of the last rainfall; (12) if the snow had begun to melt and if so, when; (13) the street address, sample site and number(s).

⁸ For communities of less than 1,000 people, one sample was drawn from each of three sites: the treatment facility (if one existed, otherwise from the raw source), the school, and a public water tap. For communities of more than 1,000 people, one sample was obtained at the water treatment facility (or raw source if no treatment plant existed) and each of the secondary pumping stations. In addition, a sample was taken from one house, hotel, or public tap, identifying the pumping station location. A sample was also taken from any school in which urine samples were collected and all bottling plants were visited and a sample of the water was collected after the plant’s treatment of the city water but before the water was mixed with syrup. When taking the actual sample, the tube was first rinsed with the water to be collected and the water was then collected from rushing water of the stream or river, never from nonrunning water or from water after the water tap had been opened for at least 1 min. Sample tube numbers were recorded onto the Water Sample Data Sheet and the exact location of the sample site was noted.

Some of the questions overlapped with those listed above for the Water Sample Data Sheet but those unique to the second questionnaire included: (1) name of person interviewed; (2) address; (3) exact location of the pumping station for the drinking water supply; (4) the material used to construct the drinking water storage tanks; (5) the material used to make the pipes that carried the drinking water into the homes; (6) the exact location of the treatment facilities or the location of the raw water source if there was none; (7) the manner in which the water was treated before it was pumped into the distribution pipes; (8) the name of the treatment process; (9) if the same water source(s) had been used for the past 25 years for the city drinking water and an explanation of what those were (10) where the city obtained its drinking water, (11) if the entire city obtained its water from this primary source or were there other drinking water sources for various parts of the city; (12) if the same sources were used during all seasons of the year or if for example during the dry season, additional sources were used; (13) how long the city used these sources of drinking water; (14) where the water was obtained for agricultural/irrigation purposes and animal drinking water supplies; (15) how far into the surrounding area those drinking and animal and agricultural water sources were used.

plastic containers after washing with the water from the lakes being sampled.

Sample Analysis The analysis of samples was done at the University of Tarapaca in Arica by Professors Figueroa, Zumeata, Razmilic, Mdalic, Hrepic, and Quintana at the University of Texas by Drs. Heffernan and Marks, and the University of Antofagasta by Professors Alonso and Gutteriz. They analyzed samples atomic absorption (standard addition method). Professors Barr at the Mc Master's University and Dr. Clarke at the National Institute for Standards Technology (see [16]) used high sensitivity static mass spectrometer for sample analysis [17]; Professor Zaldivar [7, 14] used the methods of Amidsen [18] and an Eppendorf photometer for the analysis of samples.

Normal Control Samples Normal control samples were taken from several different sources. Thus, samples taken from the Rio San Jose in Valle Azapa were the so called "low lithium level" controls for comparison with Valle Lluta and Valle Camerones. Other samples taken from other parts of Chile outside Region XV such as Santiago were also considered to be low lithium level controls. Finally, samples from outside Chile from several literature sources [8–10, 16] and a few additional non-Chilean samples taken by the MAHGP for Panama and Texas (see Table 6) also served as low level controls for lithium levels. Anderson et al. [19] also analyzed 22 surface waters within the Piedmont and Coastal plains of Georgia and South Carolina and found the maximum lithium levels to be 4.6 ppb. Kzosos and Stewart [20] noted they could find little information on the concentration of lithium in drinking water but "Hot spring spas advertising benefits of their Li-rich waters at worldwide web sites are listed for New Zealand, Korea, Canada, Mexico, Italy, Chile, Czechoslovakia, Germany, France, and the USA. Within the USA, Li-rich hot spring spas are present in South Dakota, New Mexico, Wyoming, California, Colorado, Utah, and Texas. In cases where the Li concentrations are reported for such spas, values typically are on the order of about 1–10 mg L⁻¹." Neri et al. [21] also constructed a very large lithium water content database from 526 communities across Canada between 1970 and 1972 and found the levels to be generally low in accordance with most found by others.

Findings The findings of this report confirm previously published reports and once again document that the highest concentrations of lithium in the world in surface waters are found in the north of Chile. Indeed, Zaldivar [13] in 1968 and Schull et al. [12] in 1979 independently discovered these high lithium levels as neither knew of the other's work.

This report also confirms previous studies of the Rio Lluta and Rio Camerones where lithium concentrations in

each valley increased with decreasing altitude from the mountains to the sea [12, 14] in contrast to the Rio San Jose in the Valle Azapa where no significant elevations were found over the course of its length (see Tables 1, 2, and 3).

Overall, the findings from all sources at the same localities are remarkably consistent. They thus remain highly comparable over the 40-year sampling period (Tables 1, 2 and 3; Fig. 3) despite using the different analytical methods noted above.

This report extends those previously published demonstrating the existence of elevated lithium concentrations both inside and outside of Region XV in areas not reported hitherto. They also reveal apparent exceptions to the high to low gradient theory (see above) both in and outside of Region XV. Thus:

Region XV There appear to be examples where the levels of lithium at higher altitudes are not far lower than those found at the sea (Fig 1).

Hoya (Headwaters) of the Rio Lluta The Laguna Chungara and the Muestras Salina del Bofedal and Sector Misitune are situated at 4,000 m above the hoya of the Rio Lluta. Anomalous, the lithium concentrations in the Sector Misitune appeared to be higher (207,000 ppb) than any of the small salars and lagunas found around the Salar d'Atacama (Table 7; Fig. 4). By contrast, the lithium concentration in the Chucuyo stream draining the Bofedales (5 ppb) and other relatively nearby high altitude (>4,000 m) lakes (Lago Cotacotani (65–130 ppb) and Lago Parinacota (133 ppb)) appeared to have relatively low lithium levels as did some of the high altitude valleys (e.g., > 3,200 m) relatively nearby like the Lauca Precorderilla. This goes from NE to SW along the Rio Tignamar south of the lithium rich lakes just described. These were also low in lithium throughout their extent (30–157 ppb) (see Table 4; Fig 2). The lithium reading taken at the highest (ca 5,000 m) town (Visviri) was also very low (40 ppb; Fig 2).

The highest lithium levels along Valle Lluta (2,300 ppb) were found at the Chaculluta bridge at km 10. The levels remained high until the Rio Lluta entered the sea (max, 1,580). However, somewhat anomalously at km 5 (110 ppb) and Chacullata airport (40 ppb) the levels were very low though the former may have been taken after desalination took place at Desalari Inc. (see below) while the latter was from a well that was probably not connected to the Rio Lluta.

Rio San Jose and Valle Azapa The lithium concentrations throughout most of the extent of the Rio San Jose along Valle Azapa were ca 100 ppb though two readings said to be from subterranean waters at El Morro (1,050 ppb) and

Gallinazo (1,240 ppb) were anomalously high (Table 1; Fig. 1).

Hoya of the Rio Camerones Lagunas Roja, Amarilla, and Verde had very high levels up to 24,880 ppb (Kamiya, Ilgren, Figuerola (unpublished data) 2009; Zumeata in 1978 (unpublished data)) at the headwaters of the Rio Camerones above Caritaya (see Table 3; Fig. 1).

Salar d Surire The Salar d Surire is located ca 50 km south of Caritaya. Lithium concentration data do not appear to have been published for this Salar wherein lithium levels approach 120,000 ppb⁹ (Table 4; Figs. 1 and 2).

Other Quebrada in Region XV Several smaller valleys or quebrada, running mostly from mountains to the sea (Q. Sagasca–Huara, 617–945; Q. Tarapaca–Tarapaca, 772–822¹⁰; Q. Coscaya, 994–1,019; and Q. Inf Tarapaca 1,044–1,644) had relatively high (>500 ppb) lithium concentrations (Table 5; Figs 1 and 2).

Region I High lithium concentrations were found along the Rio Loa (Tatio, 2,466 ppb (near thermal spring); Puritami therm, 6,439 ppb; Rio Salado, 1,500 ppb; Calama 4,100–4,700 ppb; Quillagua, 8,000 ppb), the longest river in Chile. Indeed, the highest surface water level not found in a laguna or salar was seen in Quillagua (8,000 ppb; Fig. 3).

Region II San Pedro d'Atacama contains two rivers. The lithium concentrations in the Rio Villama (2,089–2,700 ppb) are high while those in Rio San Pedro are comparatively low (340–400 ppb; Table 7; Fig. 4). To the south of San Pedro are a series of small towns, saline-rich lakes, and small salars (Fig. 4) to the east of the Salar d'Atacama. While the lithium levels in the lakes and small salars are generally very high (Table 7), those in only two of the villages, Camar (700 ppb) and Peine (500 ppb) exceeded 500 ppb. Lithium readings for two other towns in Region II, Victoria (629 ppb) and Antofagasta (550 ppb) found high lithium levels in their tap water (Table 6; Fig. 4).

Normal Readings The “normal” control readings in the Rio San Jose along the Valle Azapa were uniformly low (ca 100 ppb) throughout its length compared with the nearby Valle Lluta and Camerones (with the few exceptions mentioned above). The few readings in the major cities outside of Region XV far to the south such as Santiago (average, 22,

0–67 ppb), Quniterro (11 ppb), La Serena (180 ppb), and Valdivia (0 ppb) were equally low. Similarly, a few lithium readings from other parts of South America such as Panama (7.4 ppb) and Bolivia (La Paz, ca 15 ppb) were low as well and a few points along southern Peru near from Lima to Tacna were the same order of magnitude as Valle Azapa (average, 110 ppb). The lithium concentrations for major US cities and rivers are overall quite low (ca 1–2 ppb) [8–10, 16] (Table 6) as are the levels for one locale in Canada (Guelph, Ontario: 2 ppb) which served as the control city for the study of Barr et al. [16].

Discussion

This report has firmly documented elevated levels of lithium in certain parts of the north of Chile. The data from nearly 40 years of measurements using different analytical techniques confirm that some surface waters, basically unconnected to Salars and any commercial mining activity, contain the highest levels of lithium in the world as first reported by Zaldivar [13].

Most of the data reported herein are from the northern part of Region XV where studies of the small populations along the Rio Lluta and Rio Camerones were initially conducted [7, 12–16]. Another study was also done in San Pedro in 1989 patterned after the design, questionnaires, and forms of the MAHGP but on a much smaller scale (Barton, Barr, Alonso, unpublished data). The San Pedro study preceded a water analysis for the rivers and salars near the Salar d'Atacama. [22].

The San Pedro data display an interesting division of lithium levels between the two rivers that flow through it, the Rio Villama being ten times on average greater than the Rio San Pedro. Lithium levels in several towns and villages to the east of the Salar d'Atacama such as Peine were not surprisingly elevated; Peine is also the town where many residents work on the Salar d'Atacama. These towns and villages, as well as many of the others not found in the Valle Lluta and Camerones could serve as potential future study areas to assess the possible effects of lithium upon human health. Such areas could include some of the villages noted above with lithium levels that exceed 500 ppb and some of the small populations that live around the Laguna misitune and the Salar d Surire (e.g., Chilcaya).

Origin of Lithium in the Surface Waters of the North of Chile “Each riverine appears to have its own chemical signature” (Schull 2011, personal communication) since the lithium elevations are often found with other elements such as arsenic, boron, caesium, strontium, and others (see Table 1 of [23] for list) in varying concentrations. However, lithium may be elevated in some areas with little elevation in other elements (e.g., in Guantocondo according to Dr. Ana

⁹ While this may not influence the river pattern lithium concentrations described in this report that is not entirely certain since, according to Salas (April 2009, personal communication) drainage is through Bolivia in the Rio Lauca.

¹⁰ At Tarapaca village, the people told Dr Barton they could not drink the water from the “river” as it was so bad.

Marie Sancha, personal communication) data not available (but also see Sancha 2005 [24]).

The chemical variation between rivers and valleys is in part a reflection of the nature of the volcanic sources from whence the elements arise. Volcanism is clearly a major source of lithium [25] hence the association of higher levels of lithium in thermal sources.¹¹ It may explain the concentration gradients noted in some valley river systems. Risacher et al. [26] thus related lithium levels to thermal spring activity and temperature (see their Fig. 26). Professor Chung Diaz of the University of Antofagasta (2009, personal communication) described the process by which lithium originates from volcanic activity in an area called the “Planetary Anomaly of Elements” that includes SW Bolivia, NW Argentina, Central South Peru, and the north of Chile. Thus, as the meteoric rainwater falls on the Andean volcanoes, the hot centers cool down and became exhausted and the volcanoes collapse. This is partly due to the fact that hot water dissolves many rocks causing the release of various minerals. Indeed, where the water falls as rain into the magma chamber, the much higher temperatures cause the dissolution of materials thus causing the minerals to be released. Since Li is very soluble, the rocks are more likely to give up Li through leaching than other minerals. Due to its mobility, it readily travels down under the Andes (like many minerals such as rubidium, caesium, and strontium). These stop in the Pre-Cordillera largely because of a change in altitude, climate, and other things. The concentrations of such minerals (e.g., including sulfate, arsenic, boron, Li, Cs, Be, and Sr) further increase through evaporation on the desert floor.

Chung Diaz (2009, personal communication) also proposed that the chemical differences between river systems may be due to the particular chemistries of the volcanoes with which they are associated. He thus stated that chemically “volcanoes can be totally unpredictable. In Japan, some volcanoes produce only sulfur not lava; in the Middle East some produce phosphate only; and some in the north of Chile produce only iron as hematite.”

As for the lithium concentration gradients, Chung Diaz (2009, personal communication) said the highest concentrations of Li in the world in this particular area was first reported by Moraga et al. (as described by [27]). The higher lithium concentrations at the coast could be due to the manner in which the volcanoes evolved in the north of Chile via oceanic subduction. Thus, the volcanoes initially arose at the ocean and, as explained by James [28], migrate progressively eastward with time. The large concentration

at the coast may therefore reflect the areas where minerals accumulate as they create geothermal gradients, collapse, and accumulate during their eastward migration.

Other Factors which Influence Lithium Concentrations in the North of Chile:

Diversion and Desalination Due to the extreme scarcity of water in the north of Chile, some areas must be fed through diversions that take water from another location, sometimes many miles away. This has been done in several areas in the north of Chile so, obviously, the chemistry of the water taken in some cities is not a reflection of where it has come at source. Many years ago, diversions were thus made to accommodate the water needs of certain areas including but not limited to Arica and Valle Azapa,^{12,13} and Iquique.¹⁴

¹² “Originally the Rio Lauca, and its major tributaries, the Rio Quiburana and Rio Sajama, drained into the Lago de Coipasa in Bolivia; however, several decades ago, with the consent of Bolivia, to meet the increased water needs of the Azapa Valley agricultural enterprises and the growing electrical requirements of Arica, some of the waters of the Lauca were diverted into the Rio San Jose that sustains the Azapa Valley... The San Jose River, for example, until the diversion of a portion of the Lauca, had very few feeder streams originating in the altiplano and was not connected to any thermal area. Thus, the quality of its water was far better than most of the other rivers, but the quantity it carried was very much less, and rarely, save in years of exceptionally heavy rain in the sierra, did its waters actually reach the Pacific” [12]. Also, Kohn [31] cf: Fig. 5 “Hydrological Pattern, Dept of Arica”.

¹³ Also, with an apparent mix of surface and subterranean waters for the city of Arica, Barton said in 1979, “the water came from three wells at the Pago de Gomez water plant and from subterranean rivers which run close to the surface of the ground at the Planta Azapa plant. At both places, the water is placed into large, closed metal tanks then pumped...”.

¹⁴ The Chintaguay springs supplies water for Pica, Matilla, Pozo Almonte, Tirana, and Huayca with a pumping station in Pica at which chlorine is added; 7 kg of chlorine every 24 h, pumping 33 L/s of water. Pica area has an interesting water history, it is said that the water in the Chintaguay springs is from underground rivers which form a pool. As these are thermal springs, this causes a salt deposit build-up problem for some vegetation, especially the citrus groves which are numerous around Pica. Every 7 or 8 years, a trench has to be dug around the roots and the salt flushed off. Before 1845, Iquique’s water supply was a spring near the ocean, but an earthquake caused it to dry up, afterwards they were getting water by boat from Arica; then in 1860, the railroad tracks were laid by an English engineering firm to haul water by stream trains from Pica. This was also the era of heavy nitrate mining for which there are obvious remains on the road between Canchones Pumping station and Oficina Sara. Since 1969, Iquique gets its water from the Canchones wells, which are four wells, 45 m deep, and pumped into five large/huge metal tanks supported at least one story in the air and piped by large conduct on top of ground to Iquique. Likewise, Pozo Almonte used to use the well water from Planta D.O.S. at Oficina Sara until 5 years ago. Also, water is piped by large conduct on top of ground from wells at Sagasca to Huara and a newly built military compound at Baquedano. Matilla uses the Chintaguay spring water 6 days a week (64.8 ppb) and 1 day a week a local spring source (72.0 ppb).

¹¹ Thermal activity is one source of sample variation—note higher readings noted from Tatio geyser and Puritama at the headwaters of the Rio Loa, Chusmisa—stream between school and plant=29 ppb; open thermal spring=278 ppb; clear thermal activity in the lagunas Verde, Roja, Amarillo; Peru Caliente run stream, 140 v 251 ppb thermal spring.

Also, some of the water from Rio Lluta has been diverted at km 5 and then desalinated (Desalari Inc) before being sent to Arica. Some villages also rely upon water trucked into their area from locations with cleaner water, e.g., Diez y Seis¹⁵ on Rio Lluta and Cuya on Rio Camerones¹⁶ from Arica.

pH and Vegetation Water pH and vegetation may also influence lithium levels and help explain the anomalous distribution of lithium concentrations nearer the coast. For example, Schull et al. [12] said that “Rio Lluta is the strongest flowing and has the largest catchment area in the region. However, it is highly acidic at its point of origin near Mount Tacora, remains so as far westward as Poconchile,¹⁷ and often reaches the Pacific still acidic. ... The result of this confluence of small rivers of quite different character is the anomaly of highly alkaline agricultural soils being irrigated by strongly acid water.¹⁸” Since there appears to be a greater uptake of lithium by plants in acidic soils [10] and plant fiber binds lithium generally, the acidification of the large fields of vegetation nearer the coast could cause large amounts of lithium to be bound to plant fiber which is, in turn, washed seaward. Schou [29] also mentions the importance of sodium and potassium for lithium absorption. To the extent these condition pH, changes in acidity will alter Na–K balance and thus lithium absorption.

Season and Temperature Lithium levels clearly change with seasonal variation in temperature in certain areas such as Lago Chungara [30]. This is due perhaps in large part to greater rainfall in the winter months with dilution of the lithium levels. Some data suggest water from snow melt may be higher than tap water (Putre, 208 ppb from snow

vs 140 ppb from “tap” water) while other data shown no difference (Socorama snow (52 ppb) vs “tap” (50 ppb)).

Hydrological Pattern Higher lithium concentrations near the coast may also reflect an increase in the number of streams that feed the rivers as they get nearer to the sea. It may also reflect a greater number of volcanic springs found in these areas as well (see Fig. 5 in Kohn [31] “Hydrological Pattern, Dept of Arica”)

Mining Schull et al. [12] suggest mining activity may significantly influence some of the lithium levels found in the Rio Lluta and Camerones saying “But it is difficult to generalize about the burdens of these rivers for each is more-or-less unique, and their natural burdens have often been altered by the spoil from mining activities in the sierra and altiplano. At various times in the past, and to a lesser extent now these mines have produced antimony, arsenic, bismuth, copper, gold, lead, manganese, molybdenum, silver, sulfur, and zinc (Salas, Kast, Montecinos, and Salas, 1966).” These mining areas [32] have probably not been active for 75 years and may not have been sufficiently large nor near enough to alter the main watersheds of the Lluta and Camerones and thus change the lithium levels found therein.

Sample Number and Depth All of the observations and conclusions of this report rely upon the stability of replication. Only eight locations were sampled more than ten times. One of these, Lago Chungara, the most frequently sampled location [$n=39$] was “off” the water shed of the two valleys of chief concern, Valle Lluta and Valle Camerones. Fig. 5 displays by location six of the remaining seven sites for which more than ten samples were taken. Three sites are from Valle Lluta (Putre, $n=25$ ¹⁹; Molinos, $n=16$; Poconchile, $n=17$) and three from Valle Camerones (Conanoxa, $n=12$; Cuya Bridge, $n=18$; Camerones entry to the sea, $n=17$). Overall, there is general consistency over time for each place with high readings being seen where expected and vice versa. The fall in lithium concentrations in 1987 for the Camerones entry to the sea may be more apparent than real since it was said to have been taken from a small “stream” entering the ocean. This and other fluctuations may be part of the normal cycle of concentrations with season, temperature, depth, and also of course with the extent to which samples are taken. The “cyclical” nature of lithium concentration change is evident from the detailed sampling survey of Lago Chungara done by Mdalivic et al. [30]. This clearly indicates that detailed study through the year at different depths reveals a certain degree of variation.

¹⁵ For example, for Diez y Seis school, Barton said they “probably mixed some, channel water with the Arica truck water to make it go further. They do not use the channel (rio) water because of the water being so ‘bad’.”

¹⁶ I found the Camarones River to be running in two streams and I took samples from each. Again “river” is misleading as I crossed the first stream on rocks only getting one boot wet about 2 in. up. Again, I believe there is the possibility of water mixing at the Cuya Carabinero station as I took a sample from the tap in the restroom (546.9 ppb) and was told this water was trucked from Arica; again, Arica’s highest value is 115.1 ppb. The water is stored in a closed metal tank.

¹⁷ Its tributaries, the Azufre and Tacora Rivers, arise in a sulfur rich area, and are also acidic (pH hovers around 2–3), as are the waters it derives from thermal springs, some of which are exceptionally rich in borax (Wright et al. 1961 and Wright and Melendez 1963, op cit [12]). Both the Azufre and Tacora Rivers are highly toxic to livestock and travelers. Near Humalpaca, these rivers are joined by the Caracarani, and further addition of the waters of the Putre and Socorama Rivers, which are alkaline and apparently serve well the terrace gardens of the farmers in the vicinity of Putre, does not significantly alter the acidity of the main river but does apparently lessen its toxicity.

¹⁸ And this concatenation may be responsible for the periodic crop failures that occur. However, archeological remains are common in the Lluta valley, suggesting that the indigenous pre-Columbian population was able to utilize the water.

¹⁹ Putre, $n=11$ from 1993 [16] and $n=5$ from 1968 [13]; Molinos, $n=9$ from 1993; Poconchile, $n=4$ from 1993 [16].

Moreover, since these “rios” or “rivers” may be, for certain months of the year, only ankle deep, changes in lithium concentration might not be unexpected when the flow increases significantly, e.g., as Andean snow melt feeds the tributaries (also see ²⁰).

Salars and Surface Waters This is not a study of salars which are basically closed basins but of surface waters that may serve as potential sources of drinking water. Some lithium data for small Salars (e.g., Salar d’Surire in Region XV; Salars de Tara, Aguas Calientes, and Laco) and saline-rich Lagunae (Lejita, Miniques, Miscanti, Chivato, Trinchera, and Ojos de Salado; Region II) are included for comparison purposes and also because they are near small villages that could possibly serve as population study areas in the future.

Normal Levels Normal lithium level in surface waters in other parts of South and North America are very low (Table 6) and data are provided to provide some perspective on the extent to which the concentrations in northern Chile are elevated. Thus, by comparison, the levels in the north of Chile are between 100 and 10,000 times higher than most rivers in North America.

The very low levels in the rest of the world are certainly consistent with the fact that lithium only exists in ultratrace amounts ($<10^{-8}$ g/g) in normal human tissues even though it is relatively abundant in rocks and minerals [5]. Since lithium is very rapidly cleared from the body, its scarcity in normal tissue clearly reflects its very low concentration in most surface waters. Barr and Clark [5] underscore this by saying that Cade [3] believed “mankind or at least the non-maniac depressive members could live perfectly happily and efficiently in a lithium free universe.”

Elevated Levels of Lithium in Surface Waters in Other Parts of the World There have been very few studies on lithium in drinking water in relation to human health. Concha et al. [33] from the Karolinska Institute identified 198 indigenous

women in the Argentine altiplano with elevated lithium concentrations in their drinking water as reflected in their urine. A follow-up study by Broberg et al. [23] from the same institute proposed that the lithium elevations were associated with possible thyroid dysfunction as reflected in potentially attributable alterations in TSH and T4. However, these were not manifest in an excess of goiter among the subjects (Vahter 2011, personal communication) nor was there any evidence of chronic lithium toxicity (i.e., weight gain, edema, gastrointestinal pain, diarrhea, tremor, or polyuria) recognized in the population under study.

Implications for Mining, Manufacturing, and Medicine Mining may cause the release of lithium into the environment and potentially affect nearby communities. Aral and Vecchio [10] said “Geochemically, lithium is a highly mobile element, therefore, the environmental and occupational health and safety risks related to lithium in brines are higher.” Nonetheless, we are not aware of any studies of communities proximate to and potentially affected by mining activities.

Manufacturing lithium products particularly the production, storage, and disposal of batteries may also cause environmental release. There are few environmental release data for lithium around battery disposal dumps and no epidemiological studies of those living around such facilities [2]. Aral and Vecchio [10] have said that “a source of lithium posing impact to the environment is spent lithium batteries”.²¹ Aral and Vecchio [10] also said “lithium in the environment comes mostly from lithium-based grease (lithium hydroxide monohydrate) in vehicles and leaching from alkaline granitic rocks”. Ilgren and Kamiya [40] studied the impact of lithium stockpiles in terms of their implications for market supply. Kszos and Stewart [20] noted a major environmental lithium

²⁰ “Channel” refers to any system man has made (cement sides, open top) or dug in the earth to direct water and “stream” for water running in natural courses; what if were running in a sufficient quantity should be called a river; but the water supply is so terribly low at this time. Obviously, the holding tanks, as I refer to them, are only used to gather a sufficient quantity of water and ensure pressure to force water through pipes into the village proper. They were of two types which I labeled “adobe” if made of rocks and a poor quality cement, which was rough in texture and intermixed with various items. As opposed to tanks made of smooth cement, with seams, much in the style used in the states. Many of the villages had two holding tanks, one usually cleaner and covered which was used for water for the people of the village and a second usually open which was used for irrigation and stock water. Springs tended to be open and running into a stream or channel or with a cement wall enclosure, without a cover and the water bubbling up from the ground into the enclosure.

²¹ Consumers routinely dispose of batteries along with other garbage in the municipal solid waste [34]. The uncontrolled discarding of such batteries has been called a ‘huge environmental threat’ by some (Antler of Call2Recycle, cited by Belford [35]). There are no data available on environmental lithium release from spent batteries (noted by Kummer speaking at the International Battery Conference, 2009 [36]). Indeed, the EC Battery Directive adopted in 2004 (2006/66) set as one of its main objectives a high level of environmental protection, performance and a general ban on landfilling in concert with Article 12 Battery Directive that mandated meeting recycling target efficiencies by 26 Sept 2010. However, according to Mulliken [37], there was only one company in the USA as of 2009 (Toxco Inc) that actually recycled lithium ion batteries. Unfortunately, the major Toxco facility exploded in 2009, making it the fifth fire recorded from that facility and no apparent environmental assessment consequential upon the accidents appears to have been filed [38]; also see Taylor [39]. Safety issues continue to be a top priority at the UN level for both chemical (spillable) and electrical (nonspillable) hazards (Wiaux 2009, personal communication). Lithium may be released environmentally in non-dedicated facilities such as by those trying to recover cobalt since they would not be tracking lithium.

release from one of the US military stockpiles described by Ilgren and Kamiya [40] that resulted in lithium water levels between 10,000 and 19,000 ppb and significant aquatic toxicity. Kszos and Stewart [20] said “we know of no other documented occurrences of lithium contamination in groundwater or industrial effluents but it seems very likely that lithium contamination could be important in areas where lithium is processed (such as in Nevada, North Carolina, and Tennessee; see [41]) or used in quantity for fabrication or production of ceramics, glass, cosmetics or other materials”. Kszos and Stewart [20] said lithium clearly had the potential to be discharged in industrial effluents as shown by Long et al. [42] at Whitestone point on Lake Huron downstream from a processing plant. Examples of commercial sources of environmental water contamination are noted by the Environmental Protection Agency Foote Mineral Superfund Site 2004.²² The European Community Regulation on Chemicals and their Safe Use [44] now requires that many of the data gaps must be filled over the next few years for those downstream in the lithium manufacturing industry.

Medically, lithium has been shown to be the most potent neuroplastic and neurogenic agent known capable of causing brain cell growth in vitro and in vivo. The potential effects of lithium on the brains of individuals chronically exposed to high levels of lithium from birth have indeed been our primary interest. In the north of Chile, individuals have been exposed not only through childhood and adulthood but also transmaternally via breast milk and transplacentally in utero. Such exposures may have had significant structural functional consequences not seen hitherto. Where the exposures have occurred over many generations, these may have conferred some adaptive value in response to the rigorous environments in which many of these people live [12]. Moreover, if lithium is highly neuroprotective, long-term environmental exposures might reduce age related degeneration and even prevent the development of disorders such as Alzheimer’s disease [1, 45].

The two reports which follow describe the environmental lithium exposures from plant and animal foods and the levels of lithium found in the bodily tissues (urine, blood, nails, and hair) from the ingestion of lithium-loaded water and diverse foodstuffs

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²² Since 1942, Foote had manufactured lithium halides in solution at their Frazer Pennsylvania facility. During a routine inspection in 1969, the Pennsylvania DER found lithium groundwater concentrations exceeded 5,000 ppb in relation to wastewater discharge from the plant. In 1987, lithium surface water readings reached 8,000 ppb in some parts of the plant quarry with evidence of offsite lithium contamination in surface waters in nearby streams as high as 780 ppb [43]. Despite much concern by the US EPA and other agencies, human biological consequences due environmental lithium releases due to military or commercial facilities have never been proven.

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