Chlorine 36 in Great Basin waters: Revisited

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Abstract. Inspired by recently published chloride balance chronologies of closed-basin lakes in the Great Basin, researchers of the early 1960s attempted to estimate residence times of chloride in these lakes using ^{36}Cl. Unfortunately, the analytical methods of the period were not capable of measuring the ^{36}Cl levels found in these waters. About 20 years after the early research, advances in accelerator mass spectrometry permitted ^{36}Cl measurement at the required sensitivity. In this study we follow up on those pioneering efforts by remeasuring and reevaluating the ^{36}Cl content at several of the previously studied sites, focusing on Mono Lake in eastern California. Our data show that in general the streams in the region have $^{36}Cl/Cl$ ratios similar to those expected in present-day atmospheric fallout, but that the terminal lakes into which the streams flow have much lower ratios. These lower ratios could result from either a very long (>1 million years) residence time of the chloride in the basin sinks or from subsurface influx of low-^{36}Cl chloride. In the case of Mono Lake, a mass balance model based on the ^{36}Cl data and on independent estimates of chloride fluxes and reservoirs indicates major subsurface chloride input, presumably from volcanic sources, and an accumulation time in the range of 100–450 kyr. The upper bound of this range is similar to the timing of a shift from long-term humid to arid climate in the region and may indicate that hydrological closure of the basin was triggered by this event.

Introduction

In a historic paper published 40 years ago, *Davis and Schaeffer* [1955] suggested many of the principles now routinely employed in terrestrial cosmogenic radionuclide studies. Among their suggestions was that the amount of ^{36}Cl in the waters of closed-basin lakes could be used to estimate the accumulation time of the chloride in the lake waters. Apparently inspired by the ^{14}C and chloride balance studies of *Broecker and Orr* [1958] and *Broecker and Walton* [1959], *Bonner et al.* [1961] followed up on the suggestion of *Davis and Schaeffer* [1955] and reported measurements of ^{36}Cl in chloride from the Great Salt Lake and several lakes and streams in the western Great Basin, especially Mono Lake. Unfortunately, the screen wall counting apparatus that was the only method available for ^{36}Cl measurement at that time was not sensitive enough to quantify the ^{36}Cl in the natural waters sampled. The only sample in which the ^{36}Cl activity could actually be measured was one from the Truckee River that clearly contained excess ^{36}Cl due to fallout

from atmospheric nuclear weapons testing [*Bonner et al.*, 1961].

Approximately 20 years after the attempt by *Bonner et al.* [1961], the studies that they had envisioned became possible through the increase in analytical sensitivity offered by accelerator mass spectrometry (AMS) [*Elmore et al.*, 1979; *Elmore and Phillips*, 1987]. In this paper we report ^{36}Cl measurements by AMS on samples from the western Great Basin and from the Great Salt Lake equivalent to those that they attempted to measure by beta counting. Most of the basins we (and *Bonner et al.* [1961]) sampled have very limited data on ^{36}Cl in possible input sources and on subsurface hydrogeology. We therefore focus our interpretation on data from the Mono Lake Basin in eastern California (Figure 1), which is one of the more thoroughly studied basins. We draw on the chloride balance studies and groundwater modeling of *Rogers* [1993] and *Rogers and Dreiss* [this issue (a, b)] to provide parameters for a simple ^{36}Cl budget model that is used to estimate the chloride accumulation time within the basin. A description of the geologic setting and groundwater geology of the Mono Basin can be found in the paper by *Rogers and Dreiss* [this issue (a)].

Chlorine 36 Measurements and Systematics

Sample Collection and Analysis

Water samples for ^{36}Cl analyses were collected from a variety of sources in the far western Great Basin during 1983 and 1984. Locations are shown in Figure 1. Samples from high–chloride concentration waters were collected by direct precipitation of AgCl from acidified water using addition of $AgNO_3$. Chloride was collected from dilute waters (less than 20 ppm Cl) by concentration on Dowex 1-X8 anion exchange resin. The resin was prepared by eluting with 5 or more bed volumes of 2 M $NaNO_3$, followed by a 2-M sodium acetate elution. The sample chloride trapped on the resin was eluted with $NaNO_3$
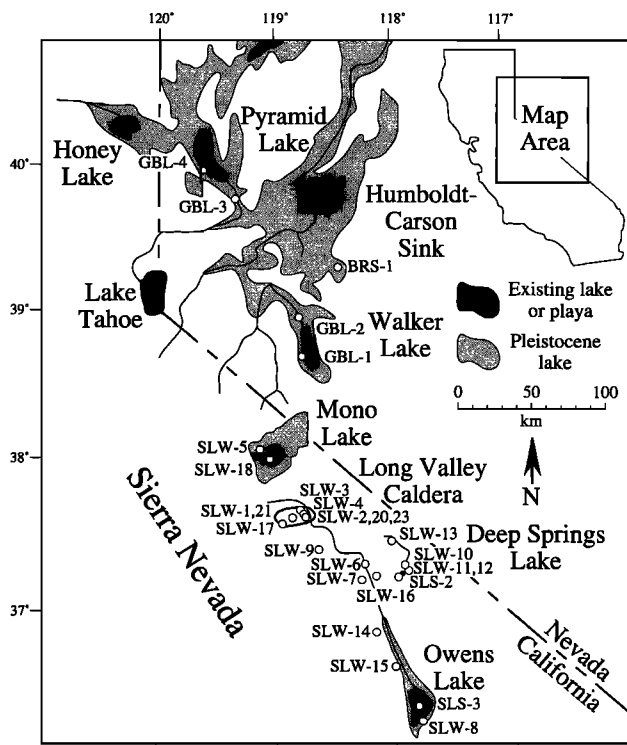
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Figure 1. Locations of water samples for ^{36}Cl analyses, listed in Table 1.

and precipitated by addition of $AgNO_3$. The ^{36}Cl/Cl ratio of the AgCl targets was analyzed by accelerator mass spectrometry at the University of Rochester using the method of *Elmore et al.* [1979]. The data are reported in Table 1, which includes data on measured tritium ratios (1 TR = 1 ^{3}H atom in 10^{18} ^{1}H atoms) in several samples. These tritium data can be used to help evaluate the possibility of nuclear weapons–produced ^{36}Cl in the samples [*Phillips et al.*, 1988]. Further information on sample locations or collection and processing methods can be found in the work by *Jannik* [1989].

Comparison of our data in Table 1 with those from *Bonner et al.* [1961] demonstrates the advance in analytical capability offered by AMS. With the exception of the bomb-affected Truckee River sample, the highest value measured by *Bonner et al.* [1961] was 0.013 ± 0.006 dpm/(g Cl) (equivalent to a ^{36}Cl/Cl ratio of 175(±80) × 10^{-15}) for Pyramid Lake, for which locality the AMS measurement was 162(±18) × 10^{-15} (sample GBL-4) (dpm denotes disintegrations per minute). However, the highest in absolute magnitude was the sample from "Utah Salt Flat" which yielded a value of −0.015 ± 0.004 dpm/(g Cl), equivalent to −200(±54) × 10^{-15} ^{36}Cl/Cl. (The negative values indicate higher beta counts in the blanks of *Bonner et al.* [1961], which were derived from salt dome halite, than in the sample materials.) This degree of error indicates that the effective sensitivity of the screen wall counters available to *Bonner et al.* [1969] was thus no better than a ^{36}Cl/Cl ratio of about 200 × 10^{-15}.

Spatial and Temporal Patterns of ^{36}Cl/Cl

Bonner et al. [1961] measured a ^{36}Cl activity of 0.13 ± 0.02 dpm/(g Cl) (1750(±270) × 10^{-15} ^{36}Cl/Cl) in the Truckee River, compared to a 1984 value of 694(±92) × 10^{-15} (sample

GBL-3). This difference can presumably be attributed to the bomb ^{36}Cl fallout peak having passed through the hydrological system [*Bentley et al.*, 1982]. By 1984 the bomb pulse had fallen to less than 0.5% of its 1960 maximum and was close to pre-bomb background levels [*Elmore et al.*, 1982; *Synal et al.*, 1990]. The 1984 value for the Truckee River is similar to that from other, smaller catchments. The Francis Lake sample (SLW-9) was collected in large part to test the magnitude of the bomb ^{36}Cl influence. It was taken from a small (~3 km^2), steep, high-elevation (3600 m) watershed near the Sierra crest and should thus have been flushed of bomb fallout soon after the ^{36}Cl pulse had passed through the atmosphere. The similarity of the ^{36}Cl/Cl ratio from this catchment, 732 × 10^{-15}, to the Truckee River and other larger drainages indicates that only minor bomb influence was persisting. The inference that these relatively high ^{36}Cl/Cl ratios are representative of natural deposition is also supported by the high value (1100 × 10^{-15}) for a snowmelt sample (SLW-17) collected from a roof in the town of Mammoth Lakes in 1984.

One significant regularity observed in most of the waters is that the ^{36}Cl/Cl ratio in the terminal lakes and playas is typically much lower than that in the influent streams (e.g., Owens River versus Owens Lake, Walker River versus Walker Lake, Truckee River versus Pyramid Lake). This discrepancy can be attributed either to very long residence time of chloride in the terminal basins (several half-lives, or of the order of 1 million years would be required) or to sources of low-^{36}Cl/Cl ratio salt within the hydrological sinks but not the surface water tributaries. The only exception to this regularity is the Deep Springs Valley playa lake (SLS-2). Deep Springs Valley is the smallest of the closed basins examined and appears to contain a relatively small inventory of chloride within and below the playa lake. There are no apparent sources of geothermal or sedimentary chloride, and chloride may be lost rapidly by eolian deflation or groundwater leakage to nearby valleys at lower elevation. The residence time of chloride in the terminal playa is apparently short, and the ^{36}Cl/Cl ratio is thus close to that of the meteoric input.

The Great Salt Lake was of particular interest to the early workers and was the first lake they attempted to "date" [*Davis and Schaeffer*, 1955]. We were able to obtain prenuclear testing samples from the lake: BYU-658 was collected by J. E. Talmage, president of the University of Utah from 1894 to 1897, on or before April 16, 1891, and LDS60-2278 (from the Latter Day Saints Church Museum) was collected during or prior to 1943. The average ^{36}Cl/Cl ratio of these samples, 30 × 10^{-15}, is far below the detection limit of screen wall counters and is in fact the lowest from any of the terminal sinks sampled. This low ratio is probably due to the large proportion of Great Salt Lake chloride derived from springs that discharge high concentrations of ancient sedimentary salt [*Feth*, 1959]. The two bedded salt samples were from a core drilled on the Southern Pacific railroad causeway across the northern end of the lake. They are from the middle and top of the most massive salt layer known in the subsurface at Great Salt Lake. On the basis of extrapolation of ^{14}C ages with depth in the upper part of the cores, the salt is tentatively estimated to have been deposited about 125 ka (D. R. Currey, personal communication, 1985). If the ^{36}Cl/Cl ratio at the time of salt deposition can be assumed to be the same as in the historical lake (30 × 10^{-15}) and the difference attributed to radioactive decay, then the combined core measurements yield an age estimate for the salt deposition of 106 ± 70 ka (including uncertainty in both the lake and

Table 1. Ratios of ^{36}Cl/Cl, Chloride Concentrations, and Tritium Ratios (Atoms ^{3}H/10^{18} H) for Natural Water and Salt Samples From the Eastern Sierra Nevada Region

Sample	Number	^{36}Cl/10^{15}Cl	Bonner et al. [1961] ^{36}Cl/10^{15} Cl	Cl$^-$, ppm	Tritium, TR
Western Great Basin: Meteoric Waters					
Owens River drainage					
Francis Lake	SLW-9	732 ± 48		0.83	
Sawmill Creek	SLW-14	508 ± 35		1.02	25.6 ± 0.8
Snowmelt, Mammoth Lakes	SLW-17	1,098 ± 91			
Owens River	SLW-15	426 ± 93		9.88	15.8 ± 0.4
Deep Springs Valley drainage					
Birch Creek	SLW-10	391 ± 76		6.22	6.96 ± 0.24
North Fork Crooked Creek	SLW-13	204 ± 61		1.52	44
Corral Springs	SLW-11	774 ± 63		6.64	37
Artesian well	SLW-12	1,095 ± 98		4.56	12
Deep Springs Lake	SLS-2	749 ± 142			
Other drainages					
Walker River	GBL-2	1,120 ± 55			
Truckee River	GBL-3	694 ± 92	1,750 ± 270		13.1 ± 0.4
Western Great Basin: Terminal Lakes and Playas					
Owens Lake	SLS-3	79 ± 9			
Walker Lake	GBL-1	88 ± 20			
Pyramid Lake	GBL-4	162 ± 18	175 ± 80		
Eightmile Flat	BRS-1	181 ± 12	−110 ± 40		
Mono Lake	SLW-18	99 ± 11	−150 ± 70	18,000	
Western Great Basin: Geothermal Sources					
Long Valley					
Casa Diablo Spring	SLW-1	35 ± 7		353	0.35 ± 0.09
Casa Diablo well	SLW-21	17 ± 4		275	
Unnamed spring, Whitmore area	SLW-2	42 ± 11		181	0.50 ± 1.0
Little Hot Creek	SLW-3	16 ± 4		214	0.16 ± 0.10
Hot Creek	SLW-4	9 ± 3		228	0.23 ± 0.10
Big Alkali Spring	SLW-20	14 ± 3		144	
Soda Flat Spring	SLW-23	23 ± 14		290	
Owens Valley					
Keough hot springs	SLW-7	35 ± 7		185	0.44 ± 0.09
Warm Springs	SLW-16	262 ± 46		16	0.56 ± 0.12
Dirty Socks well	SLW-8	5 ± 6		1,470	1.30 ± 0.11
Bishop artesian well	SLW-6	86 ± 20		22.4	2.05 ± 0.13
Mono Basin					
DeChambeau hot well	SLW-5	99 ± 19		361	0.12 ± 0.9
Eastern Great Basin					
Great Salt Lake					
Halite precipitated from lake water	LDS60-2278	31 ± 6	0 ± 130		
	BYU-658	29 ± 6			
Bedded salt, 67.0 m	BONN-C4	23 ± 5			
Bedded salt, 76.6 m	BONN-C3	24 ± 6			

Water samples collected by authors. Data for similar ^{36}Cl Samples Measured by *Bonner et al.* [1961], converted to units of ^{36}Cl/10^{15} Cl, are shown for comparison.

core ^{36}Cl/Cl measurements), which is consistent with the earlier ^{14}C extrapolation.

Chloride Residence Time From ^{36}Cl

Chlorine 36 is a radioactive (half-life 301 ± 4 kyr) isotope of chlorine that is produced in the atmosphere and at the surface of the Earth by the action of cosmic rays, and in the subsurface, by radiogenic neutrons [*Bentley et al.*, 1986]. Atmospheric and land surface production is generally much higher than in the subsurface, making ^{36}Cl a useful tracer for the origins of salt in hydrological systems. Measurement of ^{36}Cl in closed-basin lakes can thus provide constraints for the salt budget in addition to those from a simple mass balance on chloride. There are generally two primary potential sources for chloride and ^{36}Cl: surface inflow (a combination of atmospheric deposition and weathering-released land surface production) and subsurface (sedimentary, geothermal, or volcanic) input. Once chlo-

ride enters the lake system, the ratio of ^{36}Cl to stable chloride will be influenced by three processes: radioactive decay, production of ^{36}Cl in the lake water by cosmic radiation, and production of ^{36}Cl in the sediments by radiogenic neutrons. In addition, the budgets of both ^{36}Cl and stable chloride will be influenced by eolian deflation and subsurface seepage leaving the basin. Both of these processes remove ^{36}Cl and stable chloride at the same rate. Under steady state inputs, the evolution of both the stable chloride inventory and the ^{36}Cl/Cl ratio in the basin can be simulated with a simple analytical model.

The mass balance of chloride in the lake basin can be expressed by the differential equation:

$$\frac{dM_{Cl}}{dt} = i_{Cl} - lM_{Cl} \tag{1}$$

where M_{Cl} is the total chloride inventory (kilograms), i_{Cl} is the chloride input rate (kilograms per year), l is the chloride or ^{36}Cl fractional loss due to eolian export or groundwater leakage (fraction per year), and t is time (years). Similarly, the mass balance of ^{36}Cl is given by

$$\frac{dM_{36}}{dt} = i_{36} + \bar{P}M_{Cl} - (\lambda + l)M_{36} \qquad (2)$$

where M_{36} is the total ^{36}Cl inventory (kilograms), i_{36} is the ^{36}Cl input rate (kilograms per year), $\bar{P}$ is the average ^{36}Cl production rate in the lake water and sediment due to cosmic rays and radiogenic neutrons (atoms ^{36}Cl per kilogram chloride per year), and λ is the decay constant for ^{36}Cl (2.3 × 10^{-6}/yr). This formulation assumes that the input and production of ^{36}Cl are constant with time and thus does not account for introduction of bomb ^{36}Cl or past variations in cosmic ray flux.

Equation (1) can be solved, subject to the initial condition $M_{Cl} = 0$ at $t = 0$, to yield

$$M_{Cl} = (i_{Cl}/l) (1 - e^{-lt}) \qquad (3)$$

Equation (3) can be substituted into (2) (for M_{Cl}) and equation (2) solved using the initial condition $M_{36} = 0$ at $t = 0$ to give

$$M_{36} = \frac{i_{36}l + i_{Cl}\bar{P}}{l(\lambda + l)} (1 - e^{-(\lambda+l)t}) + \frac{i_{Cl}\bar{P}}{l\lambda} (e^{-(\lambda+l)t} - e^{-lt}) \qquad (4)$$

Dividing (4) by (3) gives the evolution of the ^{36}Cl ratio with time:

$$R_{36} = \frac{M_{36}}{M_{Cl}} = (1 - e^{-lt})^{-1}\left[\left(\frac{l}{\lambda + l} \frac{i_{36}}{i_{Cl}} + \frac{\bar{P}}{(\lambda + l)} \right)(1 - e^{-(\lambda+l)t}) \right.$$
$$\left. + \frac{\bar{P}}{\lambda} (e^{-(\lambda+l)t} - e^{-lt}) \right] \qquad (5)$$

Under the implicit assumption of long-term constancy in the processes and fluxes involved, the input of ^{36}Cl can be treated as a simple addition of the surface (meteoric) and subsurface (volcanic) components:

$$i_{36} = i_{Cl,s}R_s + i_{Cl,ss}R_{ss} \qquad (6)$$

where $i_{Cl,s}$ is the surface (meteoric) chloride inflow, $i_{Cl,ss}$ is the subsurface (geothermal or volcanic) chloride inflow, R_s is the $^{36}Cl/Cl$ ratio in the surface (meteoric) inflow, and R_{ss} is the $^{36}Cl/Cl$ ratio in the subsurface (geothermal or volcanic) inflow. Using the relation $i_{Cl} = i_{Cl,s} + i_{Cl,ss}$, (3) can be rearranged and substituted into (6) to give

$$i_{36} = i_{Cl,s}R_s + \left(\frac{lM_{Cl}}{1 - e^{-lt}} - i_{Cl,s} \right) R_{ss} \qquad (7)$$

The average $^{36}Cl/Cl$ ratio of the inflow can then be obtained by substitution of (3) and (7):

$$\frac{i_{36}}{i_{Cl}} = R_{ss} + \frac{(R_s - R_{ss})i_{Cl,s}}{lM_{Cl}} (1 - e^{-lt}) \qquad (8)$$

Finally, (8) can be substituted into (5) to give the combined ^{36}Cl and Cl model used to evaluate the salt accumulation time in the Mono Basin:

$$R_l = (1 - e^{-lt})^{-1}\left\{ \frac{l}{\lambda + l} \left[R_{ss} + \frac{(R_s - R_{ss})i_{Cl,s}}{lM_{Cl}} (1 - e^{-lt}) \right. \right.$$
$$\left. \left. + \frac{\bar{P}}{l} \right](1 - e^{-(\lambda+l)t}) + \frac{\bar{P}}{\lambda} (e^{-(\lambda+l)t} - e^{-lt}) \right\} \qquad (9)$$

where R_l is the $^{36}Cl/Cl$ ratio in the mixed lake/subsurface reservoir. This model has a common basis with models previously developed by Bonner et al. [1961], Paul et al. [1986], and Kaufman et al. [1990], but in addition to the processes considered by those models it explicitly includes the effects of subsurface leakage and eolian deflation on the ^{36}Cl and Cl budgets.

Mono Lake Study

In this section we focus, as an example, on one of the more thoroughly studied lake systems in the Great Basin, Mono Lake. We first assess the chloride budget of the basin. This essentially constitutes an updating of previous conventional chloride budget studies by Broecker and Walton [1959] and Scholl et al. [1967]. We then apply to Mono Lake the ^{36}Cl model described above and compare the results to the conventional chloride budget.

Geological evidence places some constraints on the hydrological closure time of the basin. The structural depression of the basin has developed over the past 3–4 Myr [Gilbert et al., 1968; Christensen et al., 1969]. Geophysical evidence, summarized by Rogers and Dreiss [this issue (a)], indicates approximately 1.5 km of lacustrine sediment in the basin center. The 760-ka Bishop Tuff is found there at about 0.4 km depth. Linear extrapolation of age with depth therefore indicates lacustrine deposition was initiated approximately 2.5–3.0 Ma. Presumably, the lake overflowed at first. The beginning of quasi-permanent closed hydrological conditions (and hence, of chloride accumulation) has not been dated, but 1 Ma would appear to be a reasonable guess for the minimum period of closure.

Conventional Basin Chloride Budget

In setting up a chloride budget for Mono Basin, we consider the basin boundary as the limit of our system, but focus on chloride dissolved in the lake and saline groundwater. We assume that beyond the lake margin, soil and sediment storage of chloride within the basin is minimal and that chloride redistribution by runoff is efficient on the timescales involved (i.e., 100 kyr). The sources of chloride input to the basin hydrologic system are rainfall, aerosols, volcanism, hot springs, and rock weathering. There is no evidence of evaporite deposits [Gilbert et al., 1968; Lajoie, 1968; Christensen et al., 1969; Axtell, 1972; K. R. Lajoie, personal communication, 1990], and limited data on chloride contents of nonevaporite minerals suggest that rock weathering is not an important chloride source [Feth, 1981]. Input from volcanoes and hot springs may be significant in Mono Basin over geologic time, but we have not estimated it in this section.

Lake and Groundwater Chloride Mass

From data of Los Angeles Department of Water and Power [1987] the volume and chloride concentration of Mono Lake in 1940 were 5.36 × 10^9 m^3 and 11,057 mg/L, respectively (Table 2). The total chloride mass in the lake was 5.93 × 10^{10} kg; the average for nine analyses from 1940 to 1980 is 5.91 × 10^{10} kg.

Table 2. Chloride Budget Components

Source/Sink	Flux	Chloride Content	Chloride Yield	Notation
Rainfall	7.27×10^8 m³/yr	0.3–0.5 mg/L	$2.4–3.6 \times 10^5$ kg/yr	$i_{Cl,s}$
Deflation	$1.1–3.4 \times 10^7$ kg/yr	11,000 ppm	$1.3–3.8 \times 10^5$ kg/yr	$-dM_{Cl,eolian}/dt$

Reservoir	Volume	Chloride Content	Chloride Mass	Notation
Mono Lake (1940)	5.36×10^9 m³	11,057 mg/L	5.91×10^{10} kg	M_l
Groundwater	6.0×10^{10} m³	4,310 mg/L	2.6×10^{11} kg	M_{ss}

The approximate volume of basin fill included within the saline groundwater outline of Figure 4 of *Rogers and Dreiss* [this issue (a)] is 1.5×10^{11} m³. Assuming an average porosity of 0.4, the volume of saline groundwater is 6.0×10^{10} m³. The Geothermal Resources International well spontaneous-potential (SP) log and model results [*Rogers and Dreiss*, this issue (a)] suggest a maximum saline groundwater concentration of 18,000 ppm total dissolved solids (TDS). Postulating that the groundwater composition is a dilution of lake water, the chloride concentration would be 4310 mg/L, for a total groundwater chloride mass of 2.6×10^{11} kg. The total chloride in the lake plus groundwater is 3.2×10^{11} kg; only 18% is in the lake. These figures are sensitive to choices of porosity, total saline groundwater volume, and concentration and assume the saline groundwater is well mixed. The groundwater concentration may be higher, as suggested by the Paoha Island hot springs [*Rogers and Dreiss*, this issue (a)]. The GRI well establishes only a lower limit on the maximum groundwater salinity, because it may have sampled within the saline-fresh interface.

Estimation of Chloride Sources and Sinks at Mono Lake

Seventy-five percent of the precipitation in Mono Basin occurs between October and March, mostly as snow in the Sierra Nevada [*Vorster*, 1985]. Sierra Nevada snow sampled by *Feth et al.* [1964b] had a mean chloride concentration of 0.5 mg/L. Mean values for the Sierra crest and east slope, more appropriate for Mono Basin, were 0.48 and 0.47 mg/L. The advantage of snow samples is that they are bulk precipitation samples and include dry fallout [*Feth et al.*, 1964b]. *Whitehead and Feth* [1961] reported a Cl concentration of 0.2 mg/L for Saddlebag Lake, on the Sierra Nevada crest in Mono Basin. *Feth et al.* [1964a] noted that for seven lakes in the Sierra Nevada, the Cl concentration was only 60% of that in the related snow, a difference they attributed to Cl adsorption on kaolinite. This implies that the bulk input of Cl at Saddlebag Lake is 0.33 mg/L. Given a total rainfall of 7.27×10^8 m³/yr [*Rogers and Dreiss*, this issue (a)], at spatially uniform concentrations of 0.33 or 0.5 mg/L, we estimate the annual chloride loading of Mono Basin by precipitation and aerosols as 2.4×10^5 to 3.6×10^5 kg/yr (Table 2).

The chloride sinks are deflation and basin leakage. Leakage can occur by surface overflow of the basin during pluvial high stands and by groundwater outflow through the bedrock. Mono Basin and Owens Valley are separated by bedrock at depth, so there is thought to be little groundwater underflow between them [W. R. Danskin, personal communication, 1989]. Lake Russell has overflowed in the past, although *Benson et al.* [1990] indicate that it has not spilled since 36 ka. For hydrodynamic reasons, described by *Rogers and Dreiss* [this

issue (b)], we do not think that overflows flushed significant proportions of chloride out of the basin.

Since Los Angeles began exporting water from Mono Basin in 1940, there has been substantial aerosol removal from the playa surface [*Cahill and Gill*, 1987]. Entrainment of spray from the lake surface is a minor component of the dust [*Kusko and Cahill*, 1984]. *Cahill and Gill* [1987] estimated that one 3-hour storm can deflate 85,000 kg of dust from the playa. This equates to the removal of only a few microns of the playa surface. Dust episodes of this magnitude occur 3.3% of the time at Mono Basin [*Cahill and Gill*, 1987].

Using *Cahill and Gill's* [1987] statistics, we calculate that 3.4 $\times 10^7$ kg/yr of dust is removed from the playa. From data of *Kusko and Cahill* [1984], chloride is about 1.1% of the dust, by weight. The calculated amount of chloride removed from the basin by deflation is therefore 3.8×10^5 kg/yr. This is probably the largest deflation rate in the basin's history and is nearly the same as the rate of Cl input by precipitation (Table 2).

We can only speculate on past deflation rates in Mono Basin. The current chloride deflation rate probably represents an extreme, owing to the large area of lake bed exposed in the early 1980s. Over geologic time, such a rate might apply only to periods of rapid lake level decline. Further, all the deflated dust may not leave the basin. Consideration of the pattern of Holocene lake level fluctuations of Mono Lake [*Stine*, 1990], and of Searles Lake fluctuations during the Pleistocene [*Jannik et al.*, 1991], suggests that lake regression has occurred no more than about one third of the time. A choice of one-third the current deflation rate gives a value of 1.3×10^5 kg/yr of chloride removal from the basin, but this probably represents a maximal estimate since most regressions have almost certainly not been as abrupt as that caused by the historic diversion. For the ^{36}Cl model discussed below, we therefore arbitrarily lower this estimate by an additional 30% to obtain an estimated long-term average chloride deflation rate of 8.4×10^4 kg/yr.

Chloride Accumulation Time

For comparison with previous salt budget studies and the ^{36}Cl results in the following section, we can combine the sources and sinks to estimate the time required to accumulate the chloride in Mono Basin's surface water and groundwater. The total solute mass in the lake and sublake sediments is 1.39 $\times 10^{12}$ kg, which corresponds to a total chloride mass of 3.19 $\times 10^{11}$ kg. For a precipitation concentration of 0.5 mg/L and one third of the current Cl deflation rate, the net input rate is 2.4×10^5 kg/yr. These assumptions are selected to produce a minimum accumulation time estimate. Dividing the inventory of 3.19×10^{11} kg by this net input rate yields an accumulation time for the chloride of about 1.3 Myr. This is about 40% of the estimated 3 Myr age of the lake. Counting only the input from

Table 3. Specified Model Parameter Distributions, Response Coefficients (C_R), and Normalized Standard Deviations (σ_N) for Input Parameters

Symbol	Parameter	Distribution	C_R	σ_N
	Input Parameters			
M_{ss}	subsurface chloride inventory	$(2.6 \pm 1.12) \times 10^{11}$ kg	+1.57	0.71
$i_{Cl,s}$	surface chloride input rate	$(3.56 \pm 1.99) \times 10^5$ kg yr^{-1}	−1.33	0.60
R_s	surface input ^{36}Cl/Cl ratio	$(750 \pm 250) \times 10^{15}$	−1.00	0.80
l	loss constant	$(2.5 \pm 2.0) \times 10^{-7}$ yr^{-1}	−1.00	1.00
R_l	lake ^{36}Cl/Cl	$(100 \pm 10) \times 10^{-15}$	+0.32	1.00
R_{ss}	subsurface equilibrium ^{36}Cl/Cl ratio	$(22 \pm 12) \times 10^{-15}$	−0.21	0.96
λ	^{36}Cl decay constant	$(2.30 \pm 0.03) \times 10^{-6}$	−0.11	0.96
P_{ss}	subsurface ^{36}Cl production rate	$(9.0 \pm 4.6) \times 10^5$ atoms ^{36}Cl (kg Cl)$^{-1}$ yr^{-1}	+0.08	1.07
P_l	lake water ^{36}Cl production rate	$(3.9 \pm 0.5) \times 10^6$ atoms ^{36}Cl (kg Cl)$^{-1}$ yr^{-1}	0.00	0.99
M_l	lake chloride inventory	$(5.9 \pm 0.3) \times 10^{10}$ kg	0.00	1.00
	Output Parameters			
$i_{Cl,ss}$	subsurface chloride input rate	$(3.2 \pm 2.3) \times 10^6$ kg yr^{-1}	−2.00	0.43
R_i	average input ^{36}Cl/Cl ratio	$(95 \pm 36) \times 10^{-15}$	+1.30	0.95
i_{Cl}	total chloride input rate	$(3.6 \pm 2.5) \times 10^6$ kg yr^{-1}	−1.12	0.44
R_∞	^{36}Cl/Cl ratio as $t \to \infty$	$(80 \pm 14) \times 10^{-15}$	+1.02	0.94

Distributions reported for the output parameters are experimentally derived from the Monte Carlo results, and the output response coefficients and normalized standard deviations are calculated relative to the experimental standard deviations.

rainfall at 0.5 mg/L, with no other sources or sinks, gives an accumulation time of 0.9 Myr.

Parameters for Mono Lake ^{36}Cl Budget Model

The average ^{36}Cl/Cl ratio of the samples representing meteoric water (samples SLW-9 through 14, SLW-17, SLS-2, and GBL-2 and 3) in Table 1 is 708($\pm$295) $\times$ 10^{15} (the Owens River analysis was excluded from this average because much of its chloride is derived from Long Valley hot springs [*Smith*, 1976]). The Long Valley geothermal sources are of interest to this study because the thermal water there circulates through aquifers of origin and mineralogy similar to the detrital sediments surrounding Mono Lake (a combination of Quaternary rhyolitic volcanics and Sierra Nevada batholith granodiorite). The average ^{36}Cl/Cl ratio of these sources, 22($\pm$12) $\times$ 10^{-15} (samples SLW-1 through 4 and 20 through 25), is probably similar to that of volcanic or geothermal sources in the Mono Basin that are not influenced by the lake water. The low tritium ratios and high chloride concentrations of these samples indicate that enhancement of the ^{36}Cl content by nuclear weapons fallout is not likely. As discussed by *Rogers and Dreiss* [this issue (a)], the DeChambeau hot well (SLW-5) probably draws sublacustrine saline water from the fringe of the saline wedge beneath the lake, at a depth of about 300 m. The ^{36}Cl/Cl ratio of the well water, 99($\pm$19) $\times$ 10^{-15}, is essentially identical to the lake water (SLW-18), 99($\pm$11) $\times$ 10^{-15}, supporting the conclusion of *Rogers and Dreiss* [this issue (a)] that the lake water and sublacustrine brine are well mixed on a relatively short timescale. The Mono Lake and sublacustrine aquifer ^{36}Cl/Cl ratio is much lower than that of the meteoric water. This characteristic is shared by all of the terminal lakes and playas measured, with the exception of Deep Springs Lake.

Monte Carlo Analysis

In order to calculate the chloride residence time in the Mono Basin it is necessary to estimate values for all of the parameters in (9). Many of these parameters cannot be specified with any certainty. This problem was addressed by Monte Carlo analysis of the equation. All input parameters were assumed to be independent and normally distributed. A mean and one standard deviation were specified for each parameter, based on data discussed above, and are given in Table 3. The sources for those related to the chloride budget are discussed above. The values for the surface inflow ^{36}Cl/Cl ratio (R_s) and the subsurface ^{36}Cl/Cl input ratio (R_{ss}) were taken from the data averages for those categories in Table 1, as described above. One thousand Monte Carlo trials were attempted, with the actual value for each parameter in each simulation being randomly selected from the normal distribution specified for that parameter. Each simulation was accomplished by generating a value for R_l (on the left-hand side of (9)) and values for all of the independent parameters on the right-hand side of the equation and then implicitly solving the equation for time so that the right-hand side equaled the left.

The loss constant term, l (which can potentially describe both eolian loss and groundwater seepage), was calculated from the formula

$$l = \frac{-dM_{Cl,eolian}/dt}{M_{Cl}} \tag{10}$$

where $M_{Cl,eolian}$ is the chloride removed by deflation. On the basis of data discussed above, the long-term average rate of eolian chloride loss was estimated to be 8.4($\pm$3.6) $\times$ 10^4 kg/yr. Both the loss rate and the chloride inventory are treated as random variables; hence the value for l given in Table 3 is calculated from the statistics of the multiple simulations. Although the loss estimate is based on eolian deflation, since no groundwater seepage is known to occur, the sensitivity estimate for this parameter (resulting from the Monte Carlo simulations) would apply equally to eolian or seepage loss.

The average ^{36}Cl production term in (9), $\bar{P}$, is a weighted combination of the surface (cosmic ray) and subsurface (radiogenic neutron) ^{36}Cl production:

$$\bar{P} = F_l P_l + F_{ss} P_{ss} \tag{11}$$

where F_l is the fraction of the total chloride inventory residing in the lake, F_{ss} is the fraction of the total chloride inventory

residing in the subsurface $(=M_{ss}/(M_{ss} + M_l)$, where M_{ss} is the subsurface chloride inventory and M_l the lake chloride inventory), and P_l and P_{ss} are the ^{36}Cl production rates in the lake and subsurface, respectively.

The lake water production of ^{36}Cl (P_l) given in Table 3 was based on cosmic ray neutron counting data in water from *Montgomery and Tobey* [1949] and *Swetnick* [1954], adjusted for the elevation of Mono Lake. The calculation accounts for the observed depth dependence of cosmic ray neutrons in water. The subsurface production rate (P_{ss}) was calculated by assuming that the measured geothermal water $^{36}Cl/Cl$ ratio $(22(\pm 12) \times 10^{-15})$ was in secular equilibrium with the subsurface neutron flux. However, P_{ss} was generated independently of R_{ss} since the rock type supplying the volcanic input need not be identical to the rock type hosting the brine aquifer. The average ^{36}Cl production rate was calculated for each Monte Carlo realization based on the four independently generated variables: P_l, P_{ss}, M_l, and M_{ss}.

Monte Carlo Simulation Results

One thousand Monte Carlo trials were attempted. Of these, 891 were successful. The remainder were discarded because either they failed to converge or they required negative chloride inflows. The histogram of accumulation times calculated by the simulations is shown in Figure 2a. The frequency distribution of the accumulation times is approximately lognormal and is strongly skewed toward short times when it is transformed into linear time coordinates. The mean and one standard deviation of the distribution are 211 ± 247 kyr, the mode is 90 kyr, and the median, plus or minus one transformed standard deviation, is 137(+364/−54) kyr.

Various additional parameters that can be calculated from the model output are listed in Table 3. These include the subsurface chloride influx $(i_{Cl,ss})$, the total chloride influx (i_{Cl}), the mass-weighted average $^{36}Cl/Cl$ input ratio (R_i), and the $^{36}Cl/Cl$ ratio as time goes to infinity (R_∞). Notably, the mean model outputs indicate that subsurface chloride input constitutes 90% of the total input and that the initial input $^{36}Cl/Cl$ ratio is close to the measured lake ratio. The mean subsurface chloride input calculated by the model, 3.2×10^6 kg/yr, is high, but is comparable to the total chloride discharge from the Long Valley hot springs of about 5.0×10^6 kg/yr that can be calculated from data in the work by *Smith* [1976]. Both continuous [*Welhan et al.*, 1988; *Oxburgh et al.*, 1991; *Bischoff et al.*, 1993] and episodic [*Broecker et al.*, 1988] inflow of geothermal waters, probably related to volcanism and not related to the lacustrine thermohaline circulation, have been reported.

Regardless of what measure of the Monte Carlo results is employed, the calculated accumulation time based on the combined chloride and ^{36}Cl balances is substantially shorter than the approximately 1 Myr accumulation time derived from the simple chloride mass balance. The key parameter in dictating this short accumulation time is the low measured $^{36}Cl/Cl$ ratio of the lake water (99×10^{-15}) compared to the surface input ratio (750×10^{-15}). This low observed ratio could be explained by two scenarios: very long residence time (leading to a low lake ratio by radioactive decay) or a large influx of subsurface chloride having a low ratio (leading to a low lake ratio by mixing). The estimated total chloride inventory is not sufficient to permit a very long accumulation time, and thus the model produces solutions with major subsurface influx.

We suggest four possible scenarios to resolve the conflict between the apparent long (1–3 Myr) closure time of the
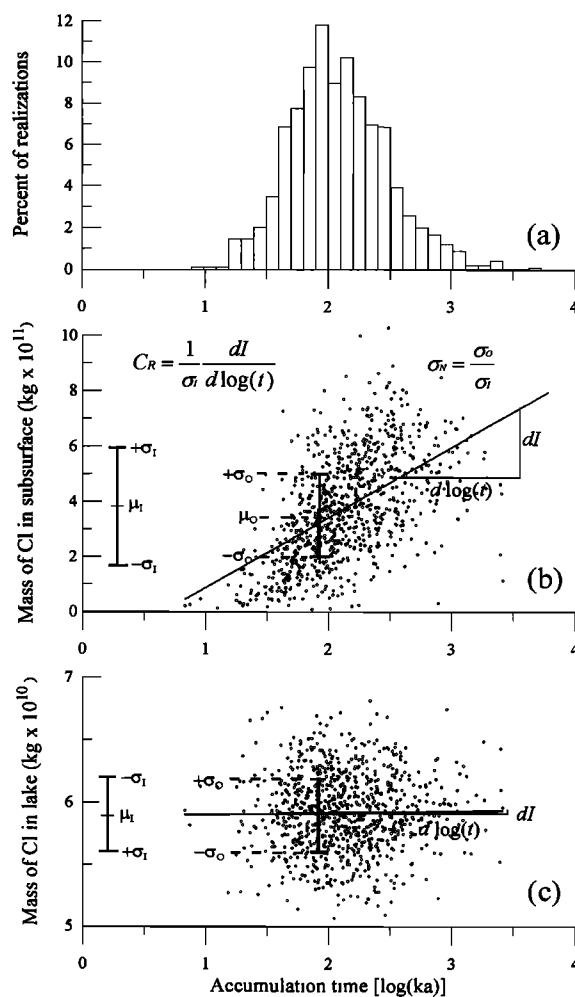

Figure 2. Results of Monte Carlo simulations of Cl and ^{36}Cl accumulation time in Mono Lake. (a) Histogram of accumulation times calculated by the simulations. (b) Plot of input values of subsurface chloride mass (M_{ss}) for the Monte Carlo simulations versus the output accumulation time. The method of calculating the response coefficient (C_R) and normalized output standard deviations (σ_N) is illustrated. This input parameter has a significant influence on the model output. (c) Same as Figure 2b, except that lake water chloride mass (M_l) is plotted against accumulation time. This input parameter exerts little influence on the model output.

lacustrine system and the relatively short (100–450 kyr) most probable result of the Monte Carlo model: (1) the basin was flushed relatively recently, (2) long-term changes in climate occurred, (3) the conceptual model underlying (9) is not adequate, and (4) the parameters used in the Monte Carlo simulations were misestimated. These are discussed in more detail below.

The first scenario involves basin flushing. This scenario is to some degree plausible. The period between 100 and 200 ka included the penultimate glacial maximum, which is a time that the lake could plausibly have overflowed. The difficulty with the scenario is that numerical simulations of the lake-aquifer system [*Rogers and Dreiss*, this issue (b)] indicate that during periods of rising lake level the saline water body in the aquifer subsides, drawing dilute water in above it, and therefore during high lake episodes there is little solute exchange between the

lake and the aquifer. These simulations thus indicate that it would be very difficult to flush the basin of chloride by sustained overflow.

The second scenario involves long-term climate change. Mono Lake has overflowed its divide and spilled to the Owens River, but this happened well prior to the last glacial maximum [Benson et al., 1990]. This apparently highly episodic timing of overflows gives support to the scenario, described above, in which the lake has been essentially closed for at least the past 1 Myr, except for brief overflows that probably would not have flushed the basin of solutes. However, the climate may have been variable over periods longer than the 100 kyr typical of glacial/interglacial cycles. Evidence discussed by Smith [1984] and Jannik et al. [1991] indicates that from 1.5 Ma (and perhaps earlier) to about 600 ka the Owens River lacustrine system was significantly wetter than during any subsequent time. Hydrological conditions during the several most recent glacial/interglacial cycles may thus not be an accurate guide to earlier conditions. These wetter conditions may have delayed the establishment of nearly continuous hydrological closure, and thus the beginning of solute accumulation, until about 600 ka.

The third scenario concerns conceptual model inadequacy. The most probable departure of the actual system from that embodied in (9) is with regard to the initial condition. The model assumes that the basin starts out free of both chloride and ^{36}Cl. In reality, the basin may have contained a substantial geothermal brine reservoir at the time of closure. Formation of a saline lake after closure could then have initiated the thermohaline mixing process and permitted the ^{36}Cl/Cl ratio to evolve to its present low value over a long time period without continuous major geothermal chloride input.

The fourth scenario concerns parameter estimation. The Monte Carlo simulations do not preclude long accumulation times; rather, they merely indicate that given the parameter distributions estimated, long accumulation times are not probable. It is possible that misestimation of critical parameters has biased the output of the model. In order to further examine this possibility, we have calculated "response coefficients" for the input parameters. These were calculated by regressing the input parameters against the log of the calculated (output) time, as illustrated in Figures 2b and 2c. The response coefficient, C_R, was defined as

$$C_R = \frac{1}{\sigma_I} \frac{dI}{d \log t} \qquad (12)$$

where I is an input parameter and σ_I is the input standard deviation assigned to that parameter.

In essence, this coefficient is a measure of the correlation between the value chosen for the input parameter and the value of accumulation time calculated by the model. Parameters that exert little influence on the output of the model will have response coefficients close to zero (Figure 2c); those that strongly affect the output of the model will have coefficients with absolute value greater than one (Figure 2b). Positive response coefficients indicate that increasing parameter values are associated with increasing calculated accumulation times; negative values indicate that increasing parameter values are associated with decreasing calculated accumulation times. In addition, we have calculated normalized standard deviations for the input parameters by dividing the calculated standard deviation for the inputs that resulted in outputs near the modal value of the output time by the input standard deviation for that parameter. Here, normalized standard deviations near

one indicate little more than random association between input and output, whereas values close to zero would indicate a near-deterministic influence of the parameter.

Examination of Table 2 (and Figure 2b) shows that the parameter having the greatest influence on the outcome of the model (i.e., the largest magnitude response coefficient) is the subsurface chloride inventory, with large inventories being associated with long accumulation times. Other parameters that have significant, but lesser, influence on the model output are the surface (meteoric) chloride influx (smaller influxes being associated with longer accumulation times), the meteoric ^{36}Cl/Cl ratio (lower ratios being associated with longer accumulation times), and the loss constant (lower rates of loss being associated with longer accumulation times).

Further examination of the input estimates for these four parameters indicates that the subsurface chloride inventory is the most likely to have been misestimated to such an extent that the output of the model would be markedly affected. The evaluation of this inventory is sensitive to values of sediment porosity, saline groundwater volume, and concentration. The chosen concentration of 18,000 ppm is possibly only a lower limit. The overall extent of saline groundwater is defined only by three deep wells, and perhaps by the deep-sourced Navy Beach and Paoha Island hot springs [Rogers and Dreiss, this issue (a)]. Because the greatest basin fill thickness occurs west of Paoha Island, the uncertainty in interface location there could be especially significant. The response analysis of the ^{36}Cl model indicates that if the mean of the subsurface chloride inventory estimate were increased by 40%, the calculated chloride accumulation time would increase by approximately a factor of 4, bringing it into better agreement with the possible closure-triggering climatic event at about 600 ka.

The possibility of chloride loss from the basin by groundwater leakage is covered by the loss term l in (9). This term will describe any loss from the basin that does not fractionate ^{36}Cl from stable chloride (which groundwater advection certainly would not). So the possibility of additional groundwater loss can be evaluated by examining Monte Carlo simulations that have high values of l. Paradoxically, these seem to be associated with younger rather than older accumulation times. Because the chloride input to the lake/groundwater system is dominated by subsurface sources, the loss term preferentially removes stable chloride, and hence less time is required to achieve the final ratio.

Conclusions

The use of ^{36}Cl to estimate chloride accumulation histories was first suggested and attempted 40 years ago by Davis and Schaeffer [1955], but could not be accomplished at that time owing to the insufficiently sensitive analytical methods then available. We have repeated measurements attempted by these early workers on Great Basin lake waters and playa salts, using AMS. The samples from rivers and streams yielded ^{36}Cl/Cl ratios in the range expected for meteoric waters. The terminal lakes and playas (with the exception of Deep Springs Valley), however, contained ^{36}Cl/Cl ratios substantially lower than their surface water inflows. We have performed a more detailed analysis of the ^{36}Cl budget at Mono Lake in order to investigate the reasons for the lower lacustrine ratios.

As elsewhere, the ^{36}Cl/Cl ratios measured in the Mono Lake and underlying brine, about 99×10^{-15}, are much lower than the ratio in inflowing meteoric water, about 750×10^{-15}, but

are higher than would be dictated by equilibrium with the surrounding rock. This lower than meteoric ratio cannot be explained by radioactive decay, since the total chloride inventory is not large enough to be consistent with accumulation over a sufficiently long time interval. Monte Carlo simulations employing a simple chloride and ^{36}Cl mass balance model indicate that the low ratios are best explained by mixing with a large inflow of subsurface (probably volcanic) chloride. The most probable accumulation times, according to the modeling, range from 100 to 450 kyr.

This accumulation time estimate is less than either the 3–4 Myr structural age of the basin or an approximate 1 Ma lake closure estimate based on simple chloride mass balance. The simplest reconciliation with both the geological history of the basin and the regional climatic history is that quasi-permanent hydrological closure was initiated by the onset of a prolonged period of relatively arid climate at about 600 ka. The somewhat shorter period of closure from the ^{36}Cl accumulation model can probably be explained by a preexisting brine reservoir (most likely geothermal) at the time of closure or by underestimation of the mass of chloride in the present brine reservoir by about 40%, or both together. In any case, the ^{36}Cl data require that there be substantial subsurface input of chloride, probably from geothermal sources associated with volcanism.

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