

Reports

Chlorine-36 Dating of Saline Sediments: Preliminary Results from Searles Lake, California

Abstract. *Measurements have been made of the ratios of chlorine-36 to chlorine in five halite samples from Searles Lake sediments, previously dated by carbon-14, thorium-230, and magnetostratigraphic techniques. The ages calculated from the chlorine ratios are generally concordant with those from the other methods, implying the constancy of the chlorine input ratio over the last million years.*

Saline lake deposits are of major economic and scientific significance. They are among the most important sources of sodium carbonate, sulfate, and chloride, as well as potash, lithium, and boron. During the latest Pleistocene high-water stages the lakes attracted game and early man, and their ancient shorelines therefore contain important archeological remains. The sediments deposited on the floors of the lakes are frequently sensitive indicators of climatic conditions, and studies of sediment cores from saline lakes in various parts of the world have provided valuable paleoclimatic records.

One of the serious impediments to paleoclimatic studies of saline lakes is the difficulty of dating the sediments. If sufficient carbon is preserved, they may be dated by ^{14}C methods, but only to about 5×10^4 years. Thorium-230 dating has a somewhat longer range (several hundred thousand years), but large uncertainties frequently stem from assumptions concerning the initial Th/U ratio. Paleomagnetic dating does not have an age limit, but numerous samples from continuous cores or exposed sections are required in order to identify the reversal patterns and the method provides no age control for samples younger than 7×10^5 years because there appear to have been no global magnetic reversals in this period.

Chlorine-36 has been proposed as a means of dating these deposits (1). Chloride is hydrophilic, and most chloride in a closed drainage basin will relatively quickly be transported to the terminal lake or playa. The half-life, 3.01×10^5 years, is suitable for dating late Pleistocene deposits, but, until recently, lack of sufficient analytical sensitivity prevented the application of ^{36}Cl to this purpose.

This problem has been overcome with the advent of tandem accelerator mass spectrometry (TAMS) analysis of ^{36}Cl , which is capable of measuring one ^{36}Cl atom in 10^{15} atoms of chloride (2).

A site with thoroughly studied stratigraphy and independently dated sediments is desirable for testing any new dating method. Searles Lake, in southeastern California, appears to be a suitable location for testing this method. Its sediments, which include many layers containing chloride-bearing minerals, have been studied and described (3-5). The upper sediments have been dated by ^{14}C and ^{230}Th techniques (6) and the

deeper ones by magnetostratigraphy (7). During pluvial periods in the Pleistocene, Searles Lake became part of a drainage system which extended from Mono Lake in the Sierra Nevada to Death Valley (Fig. 1). During dry periods the connections between upstream and downstream basins were severed. Alternations of these episodes produced layers of muds and evaporites in Searles Lake sediments, which constitute a detailed record of major climatic fluctuations.

The ^{36}Cl entered Searles Lake dissolved in the inflow water, along with stable chloride. This ^{36}Cl was derived from three sources: atmospheric fallout, cosmic-ray production near the earth's surface (epigene zone), and subsurface (hypogene) production by radiation from uranium, thorium, and their daughter radionuclides. The atmospheric fallout, produced by cosmic-ray spallation of ^{40}Ar and neutron activation of ^{36}Ar , has been calculated to be about 30 atoms of ^{36}Cl per square meter per second at the latitude of Searles Lake (8, 9). Epigene ^{36}Cl production results from cosmic-ray spallation of calcium and potassium and neutron activation of ^{35}Cl . Few data are available on ^{36}Cl in epigene rocks. Hypogene production is only from neutron activation of ^{35}Cl . Secular equilibrium $^{36}\text{Cl}/\text{Cl}$ ratios of subsurface waters in representative rock types range from 5×10^{-15} for sandstone to 30×10^{-15} for granite (9). Chloride is a conservative solute, and thus it may be assumed that

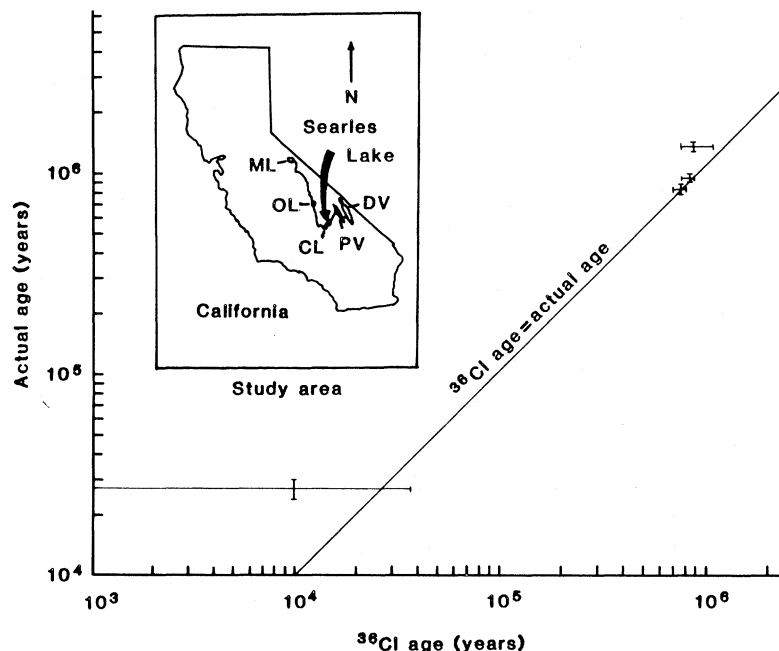


Fig. 1. A comparison of ^{36}Cl ages from Searles Lake core KM-3 with "actual" ages determined by ^{14}C , ^{230}Th , and magnetostratigraphic techniques. The study area location is indicated on the inset map. The ancient drainage pattern of the Owens River is shown. Abbreviations: ML, Mono Lake; OL, Owens Lake; CL, China Lake; PV, Panamint Valley; DV, Death Valley.

Table 1. The ^{36}Cl in Searles Lake core KM-3.

Depth (m)	$^{36}\text{Cl}/\text{Cl}$ ($\times 10^{-15}$)	Age (16) (years)	Uncorrected ^{36}Cl age (years)
14.2	55 $\pm$ 5 percent	10,000	
32.5	55 $\pm$ 6 percent	28,000	10,000 $\pm$ 27,000
190.7	8.9 $\pm$ 12 percent	800,000	802,000 $\pm$ 50,000
206.5	7.7 $\pm$ 13 percent	900,000	865,000 $\pm$ 53,000
304.3	6.6 $\pm$ 23 percent	1,300,000	922,000 $\pm$ 113,000
401.3	41.6 $\pm$ 20 percent	1,950,000	

^{36}Cl from all these sources traveled through the hydrologic system and entered Searles Lake in a time short in comparison to the half-life of ^{36}Cl .

Chlorine-36 may also be produced after deposition within the lake sediments. If the sediments remain near the land surface for a sufficient time after deposition, cosmic-ray neutrons will activate the ^{35}Cl in halite to ^{36}Cl . After hundreds of thousands of years, measurable ^{36}Cl will also be produced by neutrons arising from the decay of uranium and thorium, if sufficient concentrations of these elements are present in the surrounding sediments.

Successful sediment dating by ^{36}Cl depends on three conditions: constant or known input $^{36}\text{Cl}/\text{Cl}$ ratio, negligible cosmic-ray production after deposition, and no significant chloride translocation. The subsurface production may be calculated, and a correction introduced in the dating, if the uranium and thorium concentrations are known. We have measured the $^{36}\text{Cl}/\text{Cl}$ ratios of samples of known ages from a Searles Lake core in order to test these conditions.

Samples of halite were taken from core KM-3, a 930-m core which penetrates through the lake sediments to the basement rock (5). The top two samples are from strata dated by ^{14}C and ^{230}Th , the deeper samples at core locations between identified and dated paleomagnetic reversal boundaries. The sample depths and interpolated ages are listed in Table 1. Samples were not collected for analysis from 40 to 175 m because we lacked paleomagnetic dating control in this interval.

The samples were prepared by dissolving away about half the halite in distilled, deionized water in order to remove any exchanged or recently precipitated halite. The remaining halite was then dissolved in distilled, deionized water and the chloride precipitated as AgCl . The AgCl was purified of sulfur by repeated dissolution in ammonia and reprecipitation (9). The samples were analyzed by TAMS at the Nuclear Structure Research Laboratory, University of Rochester, according to techniques developed by Elmore *et al.* (2, 10). The

measured $^{36}\text{Cl}/\text{Cl}$ ratios are listed in Table 1.

The $^{36}\text{Cl}/\text{Cl}$ ratio of the youngest sample (14.2 m), corrected for its 10^4 -year age, was taken as the initial ratio for calculating the ages of the older samples. The similarity between the ratio of this sample, from the middle of the 10-m-thick "Upper Salt" stratum, and that of the 32.5-m-deep sample from a thin salt layer in the "Lower Salt" zone is a strong indication that postburial cosmic-ray production in the sediments is not significant. This inference is supported by calculated sedimentation rates (3-5). The $^{36}\text{Cl}/\text{Cl}$ ratio of the sample from 401.3 m is anomalously high. We are not aware of subsurface processes likely to produce a ratio this high. The most likely explanation is contamination during sample preparation. We plan to repeat this measurement in an effort to establish the origin of the high observed ratio.

The ratio of the sample from 14.2 m is interesting in its own right as an indication of the chloride budget for the basin. Smith (11) and Friedman *et al.* (12) have shown that the Upper Salt originally contained about 8×10^{11} kg of chloride, four times the present annual chloride load of the Owens River multiplied by the 2.4×10^4 year accumulation time of the Upper Salt (4). The total ^{36}Cl in the Upper Salt was thus about 48 g. The chloride in the Upper Salt has three possible sources: meteoric input, solution of chloride from rocks at depth (hypogene), and surface weathering (epigene). The meteoric source can account for only about 6×10^{10} kg of chloride (13), or 8 percent of the total. However, the calculated meteoric ^{36}Cl input (14), 14 g, is about 30 percent of the total ^{36}Cl . Even if we assume that all the remaining chloride was from hypogene sources with a typical $^{36}\text{Cl}/\text{Cl}$ ratio of 10×10^{-15} (15), only 8 g more ^{36}Cl would be accounted for. Thus at least half the observed ^{36}Cl in the Upper Salt appears to be a result of epigene processes.

These calculations are preliminary, but they illustrate the potential value of ^{36}Cl as a tracer for solutes in the hydrologic cycle. If ^{36}Cl measurements of precipitation, surface waters, and ground

waters were made, the chloride budget of the basin could be determined with confidence.

We calculated the ^{36}Cl ages for the sediments, using the basic equation

$$t = \frac{-1}{\lambda_{36}} \ln \left(\frac{\left(\frac{^{36}\text{Cl}}{\text{Cl}} \right)_{\text{measured}}}{\left(\frac{^{36}\text{Cl}}{\text{Cl}} \right)_{\text{initial}}} \right)$$

where λ_{36} is the decay constant. The ages are listed in Table 1. The initial $^{36}\text{Cl}/\text{Cl}$ ratio is assumed to be 56×10^{-15} (this is 10^{-15} higher than the ratio in the youngest sample in order to account for the 10^4 -year age of that sample). The calculated and independently determined ages are compared in Fig. 1. With the exception of the sample from 304.3 m, the ages agree within analytical uncertainty. Correction for subsurface ^{36}Cl buildup would further refine these ages if the uranium and thorium concentrations were known. A natural gamma-ray log of the hole and spot uranium and thorium analyses are available, but the correlation between the gamma readings and measured concentrations of uranium and thorium is inadequate to permit a quantitative estimate of the concentrations at the sample sites. However, the average gamma reading around 206 m is significantly higher than that around 190 m (200 compared to 80 American Petroleum Institute gamma units); this may perhaps explain the slightly young ^{36}Cl age at 206 m.

The calculated ^{36}Cl age of the sample from 304 m differs significantly from the age determined by magnetostratigraphic dating. The gamma log does not indicate high uranium concentrations. The age of the sample is at the maximum limit of the technique, and small amounts of contamination or a small uncorrected analytical background would result in large age discrepancies. Other possible explanations are that there was (i) a temporary increase in the input $^{36}\text{Cl}/\text{Cl}$ ratio or (ii) a very localized uranium concentration close to the sample, too small to be recorded on the gamma log. More ^{36}Cl measurements on this stratum, and adjoining ones, will be necessary in order to fully explain the discrepancy.

These results are preliminary but very encouraging. They imply that ^{36}Cl can provide accurate dates on continental saline sediments. They also support the hypothesis that ^{36}Cl production has been fairly constant over the last million years, a conclusion that has important implications for ^{36}Cl dating of ground water and for the geophysics of cosmic-ray modulation.

If these conclusions are confirmed, we intend to use ^{36}Cl dating to refine the chronology of Searles Lake sediments in sections of the KM-3 core that are not well dated by other methods and then to date and correlate the intervals of basin interconnection, using cores and samples from Owens, China, Panamint, and Manly (Death Valley) lakes. It should then be possible to apply the technique to saline sediments in other parts of the world.

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Serologic Evidence of Chlamydial and Mycoplasmal Pharyngitis in Adults

Abstract. In a study of 763 adult patients we found serologic evidence of infection (a fourfold increase in antibodies) with *Chlamydia trachomatis* in 20.5 percent of the patients and with *Mycoplasma pneumoniae* in 10.6 percent, but with group A streptococcus (by culture) in only 9.1 percent. Pharyngitis, the most common problem for which patients seek medical care in the United States, may be caused by nonviral, potentially treatable organisms more often than had been suspected.

Pharyngitis accounts for over 40 million visits by adults to medical facilities each year in the United States (1). It also accounts for over 100 million days of absence from the workplace annually, or more days than are lost from all strikes, work stoppages, and lockouts combined (2). In patients with a throat culture that is negative for beta-hemolytic group A streptococci, the cause of pharyngitis usually is assumed to be viral, and antibacterial treatment is withheld (3). We sought to determine whether two non-streptococcal agents potentially treatable by currently available antibacterials—*Chlamydia trachomatis* and *Mycoplasma pneumoniae*—might be playing a role in this extremely common illness. We therefore conducted a prospective 1-year study of adults seen in four general medical practices in New England.

The study group comprised 763 unselected patients with either a chief complaint or an elicited complaint of sore throat. The mean age ($\pm$ standard deviation) was 30.9 ± 10.5 years; 61 (8 percent) of the patients were 50 or older. From each patient a standardized battery of medical history and physical examination data were obtained. A double-swab throat culture was planted on separate, nonselective sheep blood agar plates that were stabbed according to routine methods (4) and incubated anaerobically to enhance beta-hemolysis. Group A streptococci were identified by beta-hemolysis and bacitracin sensitivity.

From each patient we obtained a serum specimen for measuring acute-phase antibodies. The acute-phase serum was tested for heterophil antibody by the horse-cell agglutination method or the Paul-Bunnell sheep cell agglutination method. Since we could not hope to obtain convalescent sera from all 763 patients, we created a stratified random sample that selected more heavily from patients with explicitly defined clinical findings of more severe disease, but which sampled from patients with all degrees of illness severity. A convalescent serum specimen was obtained from 166 patients approximately 6 weeks (45.6 ± 16.4 days) later (5). Acute- and convalescent-phase sera were tested for

antibodies to numerous diverse organisms. The individuals performing the tests had no knowledge of the clinical data on the patients. Not all of the paired sera could be tested for antibodies to each organism: in some cases anticomplementary activity and other technical difficulties precluded accurate testing (6).

Antibodies to streptococcus were measured with the Streptozyme test: a fourfold increase was regarded as diagnostic, and is known to be closely correlated with the measurement of antistreptolysin-O antibody (7). Antibodies to influenza viruses A and B, parainfluenza viruses 1, 2, and 3, adenovirus, and respiratory syncytial virus were measured in the Vermont State Public Health Laboratory with standard complement fixation techniques (8); a fourfold increase was regarded as diagnostic. Complement-fixing antibodies to *M. pneumoniae* (9) were measured in the laboratory of R. Chanock at the National Institutes of Health; again, a fourfold increase was regarded as diagnostic. Using the simplified microimmunofluorescent method of Wang *et al.* (10), one of us (J.S.) measured immunoglobulin G (IgG) and immunoglobulin M (IgM) antibodies to *C. trachomatis*: a fourfold increase in these antibodies (except to the A complex serotypes) was regarded as positive. Representative *Chlamydia psittaci* isolates of avian and mammalian origin were also included as slide antigens (and the results were never positive). We did not attempt to culture *M. pneumoniae* or *C. trachomatis*.

Frequency rates of different organisms, when based on the tests of the paired sera, were estimated separately for each stratum. The rates for each stratum were determined by using a logarithmic linear model (11) to reduce the sampling error associated with direct estimates based on small numbers of patients in particular strata. These rate estimates were applied to unsampled members of each stratum in determining the overall frequency of the different pathogens.

The frequency of evidence of infection with the various organisms was estimated in all patients with pharyngitis, as well