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CHLORINE-36 INVESTIGATIONS OF GROUNDWATER INFILTRATION IN THE EXPLORATORY STUDIES FACILITY AT YUCCA MOUNTAIN, NEVADA

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ABSTRACT

Chlorine-36, including the natural cosmogenic component and the component produced during atmospheric nuclear testing in the 1950's and 1960's (bomb pulse), is being used as an isotopic tracer for groundwater infiltration studies at Yucca Mountain, a potential nuclear waste repository. Rock samples have been collected systematically in the Exploratory Studies Facility (ESF), and samples were also collected from fractures, faults, and breccia zones. Isotopic ratios indicative of bomb-pulse components in the water (^{36}Cl/Cl values > 1250×10^{-15}), signifying less than 40-yr travel times from the surface, have been detected at a few locations within the Topopah Spring Tuff, the candidate host rock for the repository. The specific features associated with the high ^{36}Cl/Cl values are predominantly cooling joints and syngenetic breccias, but most of the sites are in the general vicinity of faults. The non-bomb pulse samples have ^{36}Cl/Cl values interpreted to indicate groundwater travel times of at least a few thousand to possibly several hundred thousand years. Preliminary numerical solute-travel experiments using the FEHM (Finite Element Heat and Mass transfer) code demonstrate consistency between these interpreted ages and the observed ^{36}Cl/Cl values but do not validate the interpretations.

INTRODUCTION

One of the key information needs for performance assessment of a potential high-level nuclear waste repository at Yucca Mountain, Nevada, is the quantification of ground-water infiltration and travel times in the unsaturated zone. Related to this need is the identification of preferential fluid pathways – features such as faults and fractures through which water may largely bypass the rock matrix. These needs are being addressed by chlorine-36 isotopic studies of surface samples, drill cores and cuttings, and, most recently, samples from the Exploratory Studies Facility (ESF). The ESF is an ~8-m-diameter tunnel beneath Yucca Mountain, extending as deep as the potential repository level. The tunnel exposures allow detailed characterization of the petrologic and structural settings of sample sites. Chlorine-36 investigations are part of an extensive hydrologic testing program in the ESF. The isotopic studies are fully integrated with mineralogic and textural analysis of transmissive features and with numerical modeling of groundwater infiltration.

PRINCIPLES OF CHLORINE-36 ISOTOPIC STUDIES

The interpretation of chlorine isotopic data is simple in principle and complex in application. The use of ^{36}Cl as a tracer and as a means of dating ground water is based on the production of this cosmogenic isotope in the atmosphere and its incorporation into infiltrating water. Once isolated from the atmosphere, the ^{36}Cl content of the ground water gradually decreases as a result of radioactive decay. Chloride extracted from the accessible pore spaces of subsurface samples is representative of the water that moved through the rock. Water ages, representing estimates of ground-water travel time from the surface to the underground sample sites, may be calculated from the decay constant for ^{36}Cl, the sample ^{36}Cl/Cl, the initial atmospheric ^{36}Cl/Cl, and the secular equilibrium ^{36}Cl/Cl resulting from *in situ* production by the subsurface neutron flux (from the decay of uranium and thorium isotopes) according to the standard equation of Bentley et al. [1].

The most important uncertainties affecting interpretation of the isotopic data are temporal variations in the production and deposition rates of cosmogenic ^{36}Cl relative to the deposition of stable chloride on the ground surface. The production rate of cosmogenic ^{36}Cl is controlled by variations in the earth's magnetic field strength, and the deposition of the cosmogenic isotope is influenced by climatic factors.

The deposition of stable chloride on the ground surface, which affects the $^{36}Cl/Cl$ value independent of variations in cosmogenic ^{36}Cl production, is largely a function of climate. The compound effect of climatic change includes varying eolian deposition of salt derived from salt flats and dry lake beds and changing patterns and frequency of storms from the Pacific, carrying salt of marine origin. A theoretical reconstruction of ^{36}Cl production rates for the last million years and a ~40 ky record of $^{36}Cl/Cl$ variations from regional packrat middens, dated by the ^{14}C method, both suggest that $^{36}Cl/Cl$ ratios were higher throughout most of the Pleistocene than during the last few thousand years [2].

Waters entering the subsurface during the last 40 years contain high concentrations of ^{36}Cl relative to natural background values. The elevated values are traceable primarily to global fallout from more than 70 above-ground nuclear tests conducted between 1952 and 1958 [3]. This input provides a fortuitous tracer to identify infiltrating water of very recent origin. ESF samples with $^{36}Cl/Cl$ values indicating a component of bomb-pulse ^{36}Cl are the basis for identifying fast pathways in the subsurface.

Chlorine-36 of cosmogenic origin is also produced in surficial materials and is a potential complicating factor in the interpretation of isotopic data from subsurface samples. At Yucca Mountain, Ca-rich soil calcretes are accumulation sites for cosmogenic ^{36}Cl. Our working hypothesis, subject to refinement, is that the release of ^{36}Cl from calcretes by dissolution is too small and too slow to significantly affect the isotopic signature of infiltrating water. Other potential sources of extraneous ^{36}Cl are discussed in [4].

COLLECTION AND ANALYSIS OF SAMPLES

The silicic tuffs encountered in the ESF include three main stratigraphic units: the mostly welded Tiva Canyon Tuff, the nonwelded Paintbrush Tuff, and the mostly welded Topopah Spring Tuff, in order of increasing age and depth. The PTn hydrologic unit, used in our modeling efforts, consists of the Paintbrush Tuff plus the immediately overlying and underlying nonwelded rocks of the Tiva Canyon and Topopah Spring Tuffs. Geologic samples were selected to provide a systematic representation of the bedrock and to include likely transmissive features such as faults, fractures, and breccia zones. Bedrock samples were collected at 200-m intervals along the tunnel. A large variety of features was sampled to test for the existence of preferential fluid pathways. ESF sample locations are measured inward from the north portal. For example, a location 115 m inward is designated as Station (abbreviated Sta.) 1+15. Depths from the ground surface to the ESF range from about 40 m at Sta. 2 to about 300 m at Sta. 34.

Chloride was extracted from one- to five-kg rock samples by leaching in an equal mass of deionized water for 48 hours. The Cl of interest is on the outer surfaces of particles or fractures. Poorly cohesive material was leached without further comminution, but other samples were crushed to 1- to 2-cm size fragments prior to leaching. An aliquot of the leachate was analyzed for Cl and Br by ion chromatography to estimate the contribution of Cl from construction water traced with LiBr. The remaining leachate was decanted, acidified to promote settling of particulates, and filtered. Known quantities of isotopically pure ^{35}Cl were added to samples with low Cl concentrations. Silver nitrate was added to the leachates to precipitate silver chloride, AgCl. The AgCl was purified of S by multiple cycles of dissolution in ammonium hydroxide, addition of barium nitrate to precipitate barium sulfate, followed by reprecipitation of the AgCl with nitric acid. The purified AgCl precipitates were sent to the Purdue Rare Isotope Measurement (PRIME) Laboratory for Cl isotopic analysis by accelerator mass spectrometry.

The factors and uncertainties that must be taken into account are described in [2, 4]. Isotopic compositions of samples were corrected for contamination by construction water, which has been isotopically characterized and labeled with lithium bromide to achieve a known Br/Cl ratio. Water ages, representing estimates of maximum ground-water travel time from the surface to the underground sample sites, were calculated from the corrected data assuming a maximum initial ^{36}Cl/Cl value of 1250×10^{-15} according to the standard equation of [1]. The complete data set of isotopically analyzed samples is contained in [4]. Table I lists the corrected ^{36}Cl/Cl values and calculated ages for all mineralogically characterized samples.

RESULTS AND INTERPRETATIONS

In the complete data set [4], fourteen samples (13%) have corrected ^{36}Cl/Cl ratios less than 500×10^{-15}, whereas 60 samples (57%) have values in the range of 500×10^{-15} to1250×10^{-15}. Thirty-one samples (30%) have ratios above 1250×10^{-15}. Except for two systematic (bedrock) samples, the samples with ratios above 1250×10^{-15} are from faults, fractures, or breccia zones. Our working hypothesis is that samples with ^{36}Cl/Cl ratios above 1250×10^{-15} contain a component of bomb-pulse ^{36}Cl and therefore indicate the presence of water less than 40 years old. Data from packrat middens and theoretical calculations of past cosmogenic ^{36}Cl production rates suggest that cosmogenic input was higher in the past but possibly not high enough to produce infiltration-water ^{36}Cl/Cl ratios above 1250×10^{-15}.

Most bomb-pulse samples are from the vicinities of fault zones. Only a few bomb-pulse samples are directly from fault traces; these include several breccia samples from the Bow Ridge fault zone (E008, E011) and a breccia from the Sundance fault (E175). Most Topopah Spring samples with bomb-pulse values are associated with syngenetic features, mainly cooling joints and breccias, that formed while the pyroclastic deposit was cooling and do not extend into overlying units. This result implies that paths of rapid movement of water from the surface must be more complex than individual throughgoing fault traces. In some locations, fault traces locally follow pre-existing syngenetic features. For example, the Sundance fault follows a syngenetic breccia zone at the ESF level.

The calculation of water ages/travel times from the ^{36}Cl/Cl data is conceptual-model-dependent. For ESF samples with ^{36}Cl/Cl less than 1250×10^{-15}, four alternative interpretations were developed as possible explanations of the observed values:

Interpretation 1: Modern water. Travel times for water in the unsaturated zone are assumed to be sufficiently fast that an initial ratio of 500×10^{-15} (present cosmogenic background value) can be assumed. Chloride in the samples represents a mixture of waters traveling along various flow paths and for different periods of time. Samples with ^{36}Cl/Cl ratios above $\sim 500 \times 10^{-15}$ contain at least a small component of bomb-pulse water, and therefore almost all samples from the ESF received infiltration during the last 40 years.

TABLE I

Structural Settings, $^{36}Cl/Cl$ Values, Water Ages, and Secondary Mineralogy of ESF Sample Sites

Sample[1]	Station	Sampled feature[2]	Corrected $^{36}Cl/Cl$ $\times 10^{-15}$	Water age (upper limit, ky)[3]	Calcite	Opal[4]	Clay/Mord.[5]	Clay/Mord. 2 or more[6]	Feldspar±Cr. Silica±Fe-Ti oxides[7]	Transported Particulates[8]	Mn minerals	Other mineral(s)[9]
E001	1+98	fault breccia	518±17[10]	396	-	-	●	-	●	●	●	-
E008	1+99.8	fault breccia	2132±139	●	●	-	●	-	●	●	●	-
E010	1+99.8	fault breccia	722±51	245	-	-	●	-	?	●	●	-
E011	1+99.8	fault breccia	2424±160	●	●	●	?	?	?	●	●	-
E007	2+3	bedrock	519±14	394	-	-	-	-	-	-	-	-
E073	5+04	fracture	463±16	446	-	-	●	-	-	-	●	-
E028	12+44	cooling jts.	2636±90	●	-	-	-	-	●	●	-	●
E029	13+00	bedrock	632±24	305	-	-	-	-	●	-	-	-
E030	13+67	cooling jts.	1613±93	●	-	-	-	-	●	-	-	-
E031	14+00	shear zone	2352±213	●	-	-	-	-	●	-	-	-
E033	14+41	fault breccia	853±44	170	●	●	-	-	●	-	-	-
E035	15+05	fracture	619±43	314	●	●	●	-	●	-	●	-
E036	16+12	cooling jt.	385±35	532	●	●	-	-	●	-	-	●
E037	16+19	fracture	975±36	110	-	-	●	-	-	●	-	-
E038	17+00	bedrock	632±123	305	-	-	-	-	●	-	●	-
E040	18+96	broken rock	1642±57	●	●	-	●	-	●	-	-	-
E041	19+00	bedrock	755±26	225	-	-	-	-	●	-	●	-
E042	19+31	breccia	3044±119	●	●	-	-	-	-	-	-	-
E044	19+42	breccia	2298±74	●	●	-	-	-	●	-	-	-
E045	21+00	bedrock	791±32	204	-	-	-	-	●	-	-	-
E046	22+71	fractures	865±37	164	●	-	-	-	●	-	-	-
E047	23+00	bedrock	648±33	294	-	-	-	-	-	-	-	-
E050	24+40	fault breccia	2582±98	●	●	-	-	-	●	-	-	-
E020	24+68	fracture	804±55	197	●	●	-	-	●	-	-	-
E052	26+79	shear zone	2034±66	●	●	-	-	-	●	-	-	-
E141	29+00	bedrock	924±51	134	-	-	-	-	●	-	-	-
E142	29+21	fracture	581±25	343	●	-	-	-	-	-	-	-
E144	29+73	cooling jt.	816±32	190	●	-	-	-	-	-	-	-
E149	31+64	cooling jt.	633±33	304	●	-	●	-	-	-	●	-
E150	33+00	fr. bedrock	1340±56	●	-	-	-	-	-	-	●	-
E152	34+28	fr. bedrock	4064±321	●	●	-	-	-	-	-	-	-
E153	34+32	cooling jts.	3305±205	●	●	-	-	-	-	●	●	-

TABLE I (cont.)

Structural Settings, ^{36}Cl/Cl Values, Water Ages, and Secondary Mineralogy of ESF Sample Sites

Sample[1]	Station	Sampled feature[2]	Corrected ^{36}Cl/Cl $\times 10^{-15}$	Water age (upper limit, ky)[3]	Calcite	Opal[4]	Clay/Mord.[5]	Clay/Mord. 2 or more[6]	Feldspar±Cr. Silica±Fe-Ti oxides[7]	Transported particulates[8]	Mn minerals	Other mineral(s)[9]
E154	34+71	cooling jts.	3769±114	(●)	●	-	-	●	-	●	●	-
E155	35+00	bedrock	988±56	105	-	-	●	-	●	●	●	-
E157	35+03	cooling jts.	1339±83	(●)	●	-	-	-	-	-	●	-
E158	35+08	cooling jts.	2579±172	(●)	●	-	-	●	-	-	●	-
E160	35+45	cooling jts.	3533±199	●	(●)[11]	-	-	●	●	●	●	-
E161	35+58	cooling jt.	2141±78	(●)	●	-	-	●	●	-	●	-
E175	35+93	fault breccia	2833±234	(●)	(●)[11]	-	-	●	●	●	●	-

[1]Samples were divided into separate splits for isotopic and mineralogic analysis. Mineralogic data were also recorded for the sample sites In cases where more than one split of a sample was measured for chlorine isotopic ratios, the value reported in this table is the highest value obtained.

[2]Abbreviations: cooling jt. = cooling joint; fr. bedrock = fractured bedrock.

[3]The black dot symbol in this column denotes samples containing a component of "bomb-pulse" chlorine inferred to be less than 40 years old. Parentheses around the symbol indicate that bomb-pulse values were measured in a sample or sub-sample closely associated with the mineralogically characterized sample.

[4]As used here, opal is transparent, colorless to light-colored, and typically fluoresces yellow-green in short-wave UV light. X-ray diffraction analysis of selected samples indicates opal-A.

[5]This category includes clay and/or mordenite.

[6]An entry in this column indicates the presence of two or more distinct deposits of different colors.

[7]This category includes minerals inferred to be of early to late syngenetic origin. Reported occurrences in this category are limited to minerals in growth position on the rock surfaces. Cr. Silica = crystalline silica, including quartz, chalcedony, cristobalite, tridymite, opal-CT.

[8]This category includes physically transported particulates, mostly silt- and sand-size material. Deposits of clay-size material are not included here.

[9]This category includes fluorite and unidentified minerals.

[10]1-σ standard deviation.

[11]Calcite was not present in the aliquot for mineralogic characterization but was observed in fractures at the collection site (E160) or in fractures adjacent to the fault (E175).

Interpretation 2: Holocene to late Pleistocene water, with travel times <50 ky. The travel times for most waters are likely to have been more than 40 years. If ground-water travel times to the ESF level were less than 50 ky, changes in the initial $^{36}Cl/Cl$ ratios due to radioactive decay would be negligible. The range of $^{36}Cl/Cl$ values below the bomb-pulse threshold would reflect temporal variations in the cosmogenic input function and mixing of waters with different input ratios. Samples with ratios between 500×10^{-15} and 1200×10^{-15} would have lower age limits (because of the no-decay assumption) of about 0 to 15 ky, based on reconstructed cosmogenic input values and packrat midden data for this time period [4].

Interpretation 3: Mid- to late Pleistocene water, with travel times >50 ky. The travel times for most water at the ESF level, other than bomb-pulse samples, may be as much as several hundred thousand years. Initial $^{36}Cl/Cl$ ratios for most of the infiltrating water were much higher than present-day values, and the initial ratios of older samples have been reduced due to radioactive decay. Upper limits for water travel time may be calculated assuming a maximum initial $^{36}Cl/Cl$ value of 1250×10^{-15} (see Table I). Calculated upper age limits for non-bomb-pulse samples in the complete data set [4] average about 280 ky and range up to 670 ky.

Interpretation 4: Water ages/travel times indeterminate. This alternative is included to account for factors that have not yet been fully evaluated. One such factor is the contribution of ^{36}Cl from cosmic irradiation of soil calcite. This source is unlikely to have a significant effect on the measured values of subsurface samples [4].

Independent data are required to help assess which of these interpretations are most applicable. As shown below, Interpretations 2 and 3 are most compatible with modeling results. They also imply maximum water ages comparable to U-series ages of calcites and opals from the ESF [5].

FLOW AND TRANSPORT MODELING

A series of numerical modeling experiments using the FEHM (Finite Element Heat and Mass Transfer) code [6] was run to assess the extent to which the various interpretations of ^{36}Cl data are compatible with current understanding of the hydrogeologic framework of Yucca Mountain and with current estimates of hydrologic parameters. The goal of the modeling study was to simulate chloride transport in the unsaturated zone at Yucca Mountain with a specific emphasis on interpretation and analysis of $^{36}Cl/Cl$ ratios measured in the ESF. Details of the simulations and input parameters are given in [4].

The conceptual model for ^{36}Cl transport is that it enters the unsaturated zone with infiltrating water and migrates downward either in the matrix or fractures of the variably welded tuff units. The distribution of ^{36}Cl between fractures and matrix depends on the flux rate as well as the unit in which the flow occurs. Higher flow rates increase the potential for sustained fracture flow in all units. The welded units have lower matrix permeabilities than the nonwelded units and hence fracture flow is more readily generated and sustained in those units. The nonwelded units have higher matrix permeabilities, making fracture flow less likely. The bulk permeability of fractures in nonwelded units is low due to lower fracture density, and the potential for water to flow from fractures into the matrix is higher in nonwelded units due to higher moisture gradients in that direction than in welded units.

The nonwelded Paintbrush Tuff hydrologic unit (PTn) is expected to decrease the velocity of downward moving water because most, if not all, flow occurs in the matrix. However, because bomb-pulse ^{36}Cl values have been measured in ESF samples below the PTn, there must be a mechanism for fast flow through the unit. Fast flow could occur if (1) the matrix is bypassed by a fraction of flow moving quickly through fractures in the PTn, or (2) the matrix saturation of the PTn is locally high, allowing fracture flow to occur.

Simulations of $^{36}Cl/Cl$ values in the unsaturated zone indicate that infiltration rates between 1 and 5 mm/yr are more consistent with the measured values in ESF samples than infiltration rates on the order of 0.1 mm/yr. Using current hydrologic material parameters, simulations using infiltration rates of 0.1 mm/yr yield $^{36}Cl/Cl$ values significantly less than 500×10^{-15} throughout the ESF level. Infiltration rates of 1 and 5 mm/yr lead to simulated $^{36}Cl/Cl$ ratios between 500×10^{-15} and 800×10^{-15}, consistent with measured values of most non-bomb-pulse samples. A cumulative breakthrough curve for Sta. 35+00 at a 1 mm/yr infiltration rate and unenhanced fracture permeability shows travel times on the order of 10^4 to 10^5 yr for most water (fracture and matrix) arriving at the ESF.

The simulations support the hypothesis that bomb-pulse $^{36}Cl/Cl$ ratios in the ESF reflect fast pathways associated with faults or other features of increased fracture permeability in the PTn. For simulations performed with PTn bulk fracture permeability increased above "base-case" values and infiltration rates of 1 to 5 mm/yr, small amounts of water containing bomb-pulse ^{36}Cl arrived at the ESF level within the required 40 years. At an infiltration rate of 0.1 mm/yr, bomb-pulse arrivals could not be simulated at the ESF level for any of the cases examined.

Using preliminary estimates of the spatially distributed infiltration flux [7] ranging from 0 to 30 mm/yr and averaging about 4 mm/yr, three-dimensional site-scale simulations projected a distribution of $^{36}Cl/Cl$ values consistent with ESF measurements. Under areas predicted to have no infiltration, simulated values are very low. However, some lateral flow brings younger water to units below the Tiva Canyon Tuff in some of those zones.

MINERALOGIC ASSOCIATIONS

Mineralogic studies of samples collected for isotopic analysis document the geochemical context of preferential fluid pathways. Studies of void-filling minerals in surficial pedogenic deposits and in the subsurface [e.g., 5, 8] suggest that the most common secondary minerals deposited during the last few hundred thousand years are calcite, opal, and clays. Mineralogic data for samples in this study show that calcite is usually present at sample sites that have received infiltration during the last 40 years (Table I), with 15 of 19 (79%) "bomb-pulse" values from samples containing calcite. By comparison, calcite is present in only 8 of the 20 (40%) samples with ages greater than 40 years, or 8 out of 12 (67%) if bedrock samples are excluded.

Amorphous opal (opal-A) is much less common in the samples than calcite and is associated with only one bomb-pulse sample from the Bow Ridge fault zone (E011). This finding is significant because U-series geochronology studies in the ESF, intended like the ^{36}Cl studies to identify temporal patterns of infiltration, have concentrated on the dating of opal associated with calcite [5]. However, the mineralogic data for the ^{36}Cl study suggest a possibility that opal deposition is not associated with pathways of very recent infiltration or perhaps that pathways of relatively rapid infiltration are distinctive geochemical and hydrologic environments not conducive to opal deposition. Conceptual models of infiltration based on data from calcite-opal assemblages may not be directly applicable to fast-path infiltration.

CONCLUSIONS

The data and modeling results in this study are compatible with a conceptual model of flow in which bomb-pulse $^{36}Cl/Cl$ values occur under zones of low alluvial cover, indicating that water readily enters the fractured Tiva Canyon Tuff and is transported into the underlying Paintbrush Tuff which acts as a barrier to downward movement. In the vicinity of faults or fracture zones, water rapidly traverses the Paintbrush Tuff by a combination of fracture and matrix flow and enters the Topopah Spring Tuff where downward flow occurs along fractures. The Topopah Spring Tuff is sufficiently fractured that flow is not restricted to fault traces. Infiltrating water that moves downward by routes other than fast pathways requires at least a few thousand years to reach the ESF level. Mineralogic data suggest that some geochemical distinctions exist between fast pathways and pathways without enhanced fault/fracture permeability.

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