Final Report for

DETERMINATION OF WATER MOVEMENT IN THE UNSATURATED ZONE
AT YUCCA MOUNTAIN USING CHLORIDE, BROMIDE, AND CHLORINE ISOTOPES
AS ENVIRONMENTAL TRACERS

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INTRODUCTION

This report, prepared by Hydro Geo Chem staff for Los Alamos National Laboratory, summarizes work conducted by the company under Subcontract 9-XG1-N3993-1. The ultimate objective of this work is to characterize the movement of subsurface water in the vicinity of Yucca Mountain, Nevada. Data produced under this contract is to be used by the U.S. Department of Energy in its Yucca Mountain Site Characterization Project (YMP) to help determine hydrologic flows that may affect the performance of a potential nuclear waste repository. The data may be used in the licensing proceedings, and certain quality assurance procedures have thus been required. The work has focussed on measuring the distribution of environmental tracers—chlorine-36, chlorine, and bromine—and on evaluating the depth to which these conservative solutes have percolated in the unsaturated zone at Yucca Mountain.

The following discussion summarizes progress made on the tasks outlined in the original Scope of Work. Details of this work and all data acquired by Hydro Geo Chem for this subcontract have been systematically organized in logbooks and laboratory notebooks (Appendices A and B). These documents have been structured to make it easy to trace the analytical history of a sample, from time of receipt to the final analytical results. The current status of this work and its relevance for the Yucca Mountain Project have been described in two publications co-authored by Hydro Geo Chem staff (Appendices C and D):

- Fabryka-Martin, J.T., S.J. Wightman, W.J. Murphy, M.P. Wickham, M.W. Caffee, G.J. Nimz, J.R. Southon, P. Sharma (1994). Distribution of chlorine-36 in the unsaturated zone at Yucca Mountain: an indicator of fast transport paths. Proc., FOCUS '93: Site Characterization and Model Validation, (American Nuclear Society: La Grange Park, Illinois), pp. 58-68.
- Fabryka-Martin, J., G.S. Bodvarsson, S. Wightman, W. Murphy, M. Wickham, M. Caffee, G. Nimz, J. Southon, and P. Sharma (1994). Distribution of chlorine-36 in the unsaturated zone at Yucca Mountain: an indicator of fast transport paths, p. 93 in: M.A. Lanphere, G.B. Dalrymple, and B.D. Turrin (eds.), Abstracts of the Eighth International Conference on Geochronology, Cosmochronology, and Isotope Geology (ICOG-8), U.S. Geological Survey Circular 1107.

COMPLETION OF TASKS IN THE SCOPE OF WORK

TASK 1: CHLORINE-36 ANALYSES OF SOILS AND ROCKS

- 1.0 Analyse soil and drillhole cuttings for chloride and chlorine-36, as well as for other constituents useful for interpreting the chlorine-36 data in terms of separating the meteoric component of chloride from the rock component.
- 1.1 Develop and document a routine technical procedure for processing and analyzing samples for chlorine-36 and other constituents as necessary to achieve the objective of task 2.0.

As of the end of the contract period, the following routine technical procedures had been prepared or revised by Hydro Geo Chem employees and had been issued by LANL/YMP as controlled documents (i.e., these have been incorporated into the LANL/YMP Quality Assurance Program):

LANL-INC-DP-87, R2	Identification, storage and handling of samples at Hydro Geo Chem
LANL-INC-DP-88, R0	Collection of soil samples for analysis of moisture content, bulk density, halides and chlorine isotopes
LANL-INC-DP-89, R0	Procedure for sieving soil and rock samples
LANL-INC-DP-90, R1	Measurement of moisture content of soil samples
LANL-INC-DP-92, R1	Sample leaching to extract soluble chloride and bromide
LANL-INC-DP-94, R1	Using ion chromatography to determine chloride and bromide concentrations
LANL-INC-DP-95, R1	Preparation of samples for chlorine-36 analysis
LANL-INC-DP-96, R0	Measurement of bulk density of soil samples
LANL-INC-DP-97, R0	Preparation of carrier solution for chlorine-36 samples

These documents have governed nearly all of the work conducted by Hydro Geo Chem in the processing and analysis of YMP samples. Data generated following these procedures have been systematically recorded in several logbooks (see Appendices 1 and 2).

- 1.2 Process and analyze on the order of forty samples of soil and drill-hole cuttings for chloride, chlorine-36 and other constituents as determined in task 2.0

As of the end of the contract period, Hydro Geo Chem employees had processed and analyzed samples as follows:

- 22 soil samples and 92 rock samples (including replicates) for chlorine-36, docu-

mented in the logbook for chlorine isotope results, TWS-INC-7-04-92-04

- 9 samples for stable chlorine isotopes, documented in TWS-INC-7-04-92-04
- Approximately 160 rock samples and 70 soil samples for chloride (as well as bromide in most cases), documented in ion chromatography logbooks TWS-INC-7-05-92-01, TWS-INC-11-92-03, and TWS-CST-01-94-10.

1.3 Process and analyze samples to monitor background contributions of chlorine-36

As of the end of the contract period, Hydro Geo Chem had processed and analyzed 23 samples to monitor background contributions of ^{36}Cl . Two of these were analyzed in duplicate. Results of these analyses are documented in the chlorine isotope logbook (TWS-INC-7-04-92-04, Appendix Table F-3). The main purpose of these background samples has been to serve as quality checks, prepared concurrently with each suite of environmental samples in order to verify the lack of any significant contamination. All of these blanks have had $^{36}\text{Cl}/\text{Cl}$ ratios less than or equal to 2×10^{-14} ; and most ratios have been less than 1×10^{-14} , generally well below the ratios of the samples they accompanied.

A second purpose of the background samples has been to check for the possible presence of high levels of chlorine-36 in the LANL facilities intended for sample storage and processing after relocation of operations from Tucson to Los Alamos in May 1994. Results of these analyses are also documented in the chlorine isotope logbook (TWS-INC-7-04-92-04, Appendix Table F-3). The measurements indicated the need to take precautions against contamination of samples in the boxcar (TA-48, RC-36) in which some of them are now being stored at LANL. All of these samples are stored in double containers, i.e., the sample bags themselves are contained in a second bag or box. In addition, the results for the chemical laboratory (Room 406) set aside for this work at TA-48, RC-1, were sufficiently high that the principal investigator decided to limit sample processing activities in this room to ion chromatography analyses, and to conduct sample processing for chlorine-36 in another building containing clean room facilities previously shown by her to be free of chlorine-36 contamination.

1.4 Develop a procedure to ensure the reproducibility of results

Hydro Geo Chem has routinely evaluated the reliability and reproducibility of ^{36}Cl , chloride and bromide results by analysis of blanks, standards, and replicate samples. The results of these quality-check analyses are documented in the chlorine isotope logbook (TWS-INC-7-04-92-04) and in the ion chromatography logbooks (TWS-INC-7-05-92-01, TWS-INC-11-92-03 and LA-CST-03-94-03).

TASK 2: INTERPRETATION OF CHLORINE-36 DATA

- 2.0 Develop a procedure for interpreting the chlorine-36 data in terms of separating the meteoric component of chloride from the rock component.

Apparent water residence times based upon the measured chlorine-36 signal in a rock sample may be older than the true ages due to the effect of dilution of the meteoric signal by older chloride released from the rock during the drilling process. Thus, a procedure is needed whereby the extent of dilution by rock chloride can be estimated and corrected for. Hydro Geo Chem has investigated two approaches for separating the meteoric component of chloride from the rock component. The more promising approach involves the use of chloride/bromide (Cl/Br) ratios as an indicator of the extent of mixing of these two sources. This approach is described in detail in the Focus '93 publication (Appendix C). A second approach uses stable chlorine isotopes as a mixing indicator, but results-to-date suggest that the isotopic ratio of the meteoric end-member may be too variable for this approach to work (documentation is contained in chlorine isotope logbook, TWS-INC-7-04-92-04).

In 1993, using Paradox software, Hydro Geo Chem designed and began to build a data base of analytical results to facilitate the task of identifying end-member Cl/Br and $^{36}\text{Cl}/\text{Cl}$ values and applying the mixing model to adjust ^{36}Cl -based age estimates. Because the data base tracks the receipt and numerous analyses associated with over 700 samples, this work is still ongoing.

TASK 3: CHLORINE-36 ANALYSES OF GROUNDWATERS

- 3.0 Analyse water samples from the saturated zone and from perched water zones from the YMP region for chlorine-36 and chlorine, to be used for determining the age of the water.

As of the end of the contract period, 39 water samples had been collected by the USGS for chlorine-36 analysis and forwarded to Hydro Geo Chem for analysis.

Documentation of the receipt and subsequent history of these samples is contained in the sample logbook TWS-INC-7-02-92-03. Documentation of ^{36}Cl analyses for 22 of these samples is contained in the chlorine isotope logbook (TWS-INC-7-04-92-04).

Documentation of chloride and bromide analyses for 27 of these samples is found in the laboratory notebook (TWS-INC-7-02-92-02) or the ion chromatography logbooks (TWS-INC-7-05-92-01, TWS-INC-11-92-03, or TWS-CST-01-94-10). Analyses of the remaining samples will be analyzed if and when so instructed by the Principal Investigator; some of these are duplicates or known to have been contaminated at the time of collection and hence will probably be discarded without analysis.

TASK 4: SAMPLE COLLECTION

4.0 Participate in collecting samples for analysis.

A Hydro Geo Chem employee participated in the collection of soil samples for this project in February 1992. These samples were collected near Test Cell C in Area 25 of the Nevada Test Site and were intended to test the hypothesis that high levels of chlorine-36 observed in cuttings from borehole N55 at Yucca Mountain may have been caused by contamination of the drillrig when it was previously used to drill holes adjacent to Test Cell C.

Hydro Geo Chem staff drafted a set of criteria to be followed by YMP personnel during the collection of ream-bit cuttings for this project during drilling operations. Otherwise, there has been no call for the direct involvement of Hydro Geo Chem staff in site selection or sample collection of cuttings or water samples because such activities are generally controlled by other YMP participants.

TASK 5: SAMPLE STORAGE

5.0 Arrange for storage of samples in a facility protected from weather as long as necessary.

Sample quantities and hence sample storage requirements for this project have not been inconsequential. At the present time, the sample inventory consists of about 700 samples with a total estimated weight on the order of 18 metric tons. From the beginning of the contract until May 1994, Hydro Geo Chem rented two storage lockers in Tucson for sample storage. An early activity on this contract was to conduct a thorough inventory of the samples, and to dispose of several tons of rocks that had been acquired under the previous contract and that were no longer relevant to the project. In May 1994, Hydro Geo Chem conducted an inventory of samples onsite in preparation for shipping all of these to Los Alamos where they are being stored in sheds and a boxcar (TA-48:RC-36, RC-40 and RC-89).

TASK 6: REPORTING REQUIREMENTS

6.0 Submit quarterly and annual progress reports to Los Alamos National Laboratory outlining the progress of work.

At the request of the Principal Investigator and to be consistent with the monthly reporting requirement of the YMP program, Hydro Geo Chem has filed informal monthly

letter reports summarizing the work accomplished during each month (Appendix E). These reports have generally been very short, with details provided in the laboratory notebooks and logbooks (Appendix B). Gaps in this record generally occurred when the Principal Investigator called a halt to Hydro Geo Chem activities, e.g., during review of the governing study plan and quality assurance procedures.

TASK 7: QUALITY ASSURANCE REQUIREMENTS

- 7.0 Perform all work under a Quality Assurance (QA) Program designed by the contractor to demonstrate the quality of the data generated under this contract and approved by the Los Alamos Yucca Mountain Project (YMP) office. Work cannot start until such approval has been obtained. Alternatively, perform all work under the QA Program established by the Los Alamos YMP office.

Hydro Geo Chem has performed all work for this contract under the Los Alamos YMP QA program. The effectiveness with which the company has implemented these requirements during the contract period has been documented in Los Alamos Audit Reports LANL-AR-91-14 (dated 11/26/91), LANL-AR-92-11 (dated 3/01/93), LANL-AR-93-11 (dated 1/18/94). Only five deficiencies, all easily corrected, were identified during these audits.

APPENDIX A

ORGANIZATION OF DATA ACQUIRED BY HYDRO GEO CHEM FOR SUBCONTRACT 9-XG1-N3993-1

All data acquired by Hydro Geo Chem for this subcontract have been systematically organized in logbooks and laboratory notebooks. These documents have been structured to make it easy to trace the analytical history of a sample, from time of receipt to the final analytical results.

1. *Sample logbooks* are used to track the receipt of the original sample, the withdrawal of individual aliquots for analysis, and the final disposition of the original sample. There are separate logbooks depending upon the nature of the sample: rock cuttings, soils, waters, or drillcore. These logbooks provide traceability for each sample to a unique SMF barcode ID, and assign a unique laboratory ID and aliquot ID following LANL-INC-DP-87.
2. *Ion chromatography logbooks* are used to document sample leaching following LANL-INC-DP-92, and analysis of leachates and water samples following LANL-INC-DP-94. Any sample sieving that may be required following LANL-INC-DP-89 is also documented in these logbooks.
3. The *chlorine-36 logbook* is used to document activities associated with the analysis of samples for chlorine-36, beginning with the preparation of silver chloride following LANL-INC-DP-95, submission of the silver chloride to an outside laboratory for chlorine-36 analysis, and receipt of the analytical results. Sample submission and receipt of results for stable chlorine isotopic analyses are also included in this logbook.
4. *Laboratory notebooks* are used primarily to document troubleshooting activities, and scoping, prototype or research and development activities. These notebooks may also be used to provide details as appropriate on deviations and possible sources of uncertainty associated with activities governed by detailed technical procedures (DPs). Very few data associated with activities governed by DPs are reported in these notebooks, once the first ion chromatography logbook was initiated.

APPENDIX B

NOTEBOOKS AND LOGBOOKS MAINTAINED BY HYDRO GEO CHEM
FOR SUBCONTRACT 9-XG1-N3993-1

NOTEBOOK TYPE AND ID	NOTEBOOK TITLE	DATE STARTED
SAMPLE LOGBOOKS		
TWS-INC-7-10-91-04	Logbook #1 for rock samples	02-12-92
TWS-INC-02-93-05	Logbook #2 for rock samples	02-10-93
TWS-INC-7-03-92-05	Logbook for soil samples	03-16-92
TWS-INC-7-02-92-03	Logbook for water and carrier salt samples	02-14-92
TWS-INC-09-93-02	Logbook for drillcore samples	09-10-93
TWS-INC-7-02-92-04	Logbook binder (correspondence related to sample acquisition and transfer)	03-03-92
LABORATORY NOTEBOOKS		
TWS-INC-7-02-92-02	Hydro Geo Chem laboratory notebook #1	02-14-92
TWS-INC-01-93-03	Hydro Geo Chem laboratory notebook #2	12-01-92
LA-CST-03-94-03	Hydro Geo Chem laboratory notebook #3	03-01-94
ION CHROMATOGRAPHY LOGBOOKS		
TWS-INC-7-05-92-01	Ion chromatography logbook #1	06-01-92
TWS-INC-11-92-03	Ion chromatography logbook #2	11-11-92
TWS-CST-01-94-10	Ion chromatography logbook #3	01-28-94
CHLORINE-36 LOGBOOK		
TWS-INC-7-04-92-04	Chlorine-36 logbook #1	05-01-92

APPENDIX C

PUBLISHED CONFERENCE PAPER: FOCUS '93 CONFERENCE

Fabryka-Martin, J.T., S.J. Wightman, W.J. Murphy, M.P. Wickham, M.W. Caffee, G.J. Nimz, J.R. Southon, P. Sharma (1994). Distribution of chlorine-36 in the unsaturated zone at Yucca Mountain: an indicator of fast transport paths. Proc., FOCUS '93: Site Characterization and Model Validation, (American Nuclear Society: La Grange Park, Illinois), pp. 58-68.

DISTRIBUTION OF CHLORINE-36 IN THE UNSATURATED ZONE AT YUCCA MOUNTAIN: AN INDICATOR OF FAST TRANSPORT PATHS

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ABSTRACT

The $^{36}\text{Cl}/\text{Cl}$ ratios for Cl extracted from soil and ream-bit cuttings are being used to provide information on characteristics of water movement through the unsaturated zone at Yucca Mountain. Deep penetration of the bomb-pulse signal in alluvium provides evidence for the variable effectiveness of alluvium and associated vegetation in attenuating infiltration, possibly as a result of different runoff characteristics of the watersheds. Fast transport of water via fractures through the Tiva Canyon welded unit is indicated by detection of elevated levels of ^{36}Cl in the underlying Paintbrush nonwelded unit in 3 of 5 boreholes. Preliminary interpretation of ^{36}Cl data for one deep borehole indicates average residence times of 5 to 7 $\times 10^5$ yr in the deeper Topopah Spring welded units, and 1 to 3 $\times 10^5$ yr in the underlying Calico Hills nonwelded unit.

I. INTRODUCTION

Yucca Mountain, Nevada, is under investigation as a potential site for a high-level radioactive waste repository. Because groundwater constitutes the most likely medium for transporting radionuclides from the repository to the accessible environment, determining the nature and rate of water movement through the unsaturated zone at Yucca Mountain is a critical task for assessing the future performance of a nuclear waste repository. The task is complicated by the extremely low moisture fluxes, the great thickness of the unsaturated zone, the heterogeneity of the rock units, and uncertainties about historical and future climatic variations. The lack of long-term direct measurements of infiltration requires proxy indicators of water movements through the unsaturated zone in order to estimate historical infiltration patterns.

In the present study, the $^{36}\text{Cl}/\text{Cl}$ ratio for Cl extracted from soils and drill cuttings samples is being used to provide information on water movement through the unsaturated zone at Yucca Mountain. Chlorine-36 is produced by cosmic-ray secondaries interacting with argon isotopes and by neutron-activation of ^{35}Cl and

enters the hydrologic cycle via dry and wet atmospheric fallout. When Cl at the surface is carried underground by infiltration, the radioactive decay of ^{36}Cl in the Cl can theoretically be used to time the rate of water movement in the deep subsurface. Where the residence time of meteoric water (and hence of the meteoric ^{36}Cl) in the rock matrix is on the same order of magnitude as the ^{36}Cl half-life (3.0×10^5 yr), then it may be appropriate to use the decay of the meteoric ^{36}Cl to derive these residence time estimates; the applicable range of this approach is on the order of 5×10^4 to 1×10^6 yr. Chlorine-36 also occurs at concentrations up to 2 to 3 orders of magnitude above natural levels as a result of global fallout from high-yield nuclear weapons tests conducted primarily at the Pacific Proving Grounds between 1952 and 1958.¹⁻² This latter signal can be used to test for the presence of fast transport paths, e.g. fractures and faults, to understand processes affecting the rate of water movement in shallow soils, and to estimate present-day infiltration rates. These data are part of the input for developing and evaluating the validity of models of ground-water movement at the candidate repository.

II. HYDROGEOLOGY OF STUDY AREA

Yucca Mountain is located in the northern Mojave Desert and lies 150 km northwest of Las Vegas in southern Nevada. The landform is composed of alternating layers of interbedded welded and nonwelded tuffs that have been uplifted and tilted, fractured and faulted. About 500 to 750 meters of unsaturated rock are present. Generalized conceptual models of moisture movement in the unsaturated zone beneath Yucca Mountain have been developed.³⁻⁴ Wittwer et al. provide a review of subsequent site-scale models for the unsaturated zone, highlighting the principal findings of these efforts with respect to flow paths.⁵ Regarding the parameters and underlying assumptions of these models, some of the key questions that are being evaluated in the present study are as follows:

(a) Of the precipitation falling on Yucca Mountain (~ 17 cm yr^{-1}),⁶ what proportion escapes evapotranspiration and moves into the unsaturated zone to become net

ACKNOWLEDGMENTS

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infiltration? How variable is this proportion as a result of the highly variable topography and variations in geologic units cropping out at the surface?

(b) If the influx of infiltration following a given precipitation event is large, e.g. in areas of concentrated runoff, is there evidence that fracture flow may be initiated in the upper welded unit, the Tiva Canyon (TCw)? If so, can such fracture flow be sustained to the base of this unit into the Paintbrush nonwelded (PTn) unit? Or, alternatively, does moisture in the fractures move into the TCw matrix by capillary forces before it reaches the PTn?

(c) Is there evidence that lateral flow occurs at contacts between welded and nonwelded units, such as the TCw and the underlying nonwelded PTn, due to possible capillary or permeability barriers occurring between these two units? Does lateral flow occur within the nonwelded units because of the significant difference between vertical and horizontal hydraulic conductivities?

(d) Is there evidence that infiltrating water reaches the Calico Hills nonwelded (CHn) unit via fractures or faults in the Topopah Spring welded (TSw) unit?

III. EXPERIMENTAL DESIGN

A number of YMP studies are designed to test these and other aspects of the conceptual model by independent approaches: moisture profiling, physical properties, fracture characteristics, climate and water-budget modeling. The best indicator of long-term water movement is the spatial distribution of water ages, or residence times, in the subsurface. Radiometric methods, which are based on measurements of atmospheric tritium, ^{14}C , and ^{36}Cl carried underground by percolating water, provide the most direct means for obtaining estimates of residence time as a function of location. These methods also intrinsically average many large-scale geologic heterogeneities as well as short- and long-term climatic, vegetal and geomorphic variations. Detection of bomb-pulse levels of these radionuclides provides valuable data on the types of hydrogeologic situations in which episodic fracture flow may be occurring.

A. Natural ^{36}Cl as a water tracer

The technique chosen to estimate the water travel time through the unsaturated zone in this study is the measurement of the $^{36}\text{Cl}/\text{Cl}$ ratio in soluble Cl carried into the subsurface by meteoric water and now present in pore water or on the surfaces of mineral grains. Cl is deposited globally both in precipitation and in dry fallout. The source of most of the Cl in this fallout is sea salt lofted into the troposphere by surface winds. A very small fraction of Cl atoms in the fallout consists of ^{36}Cl of cosmogenic origin.

The half-life of ^{36}Cl (3×10^5 yr) makes this nuclide an appropriate radiometric tool for estimating infiltration estimates, assuming matrix flow and given the travel times based on hydrologic modeling.⁴ If the net liquid flux through the Topopah Spring Member matrix is assumed

to be 0.2 mm yr^{-1} downward,³ with an average volumetric moisture content of 10%, then estimates of ground-water travel time through this unit exceed 1×10^5 yr. Although unambiguous interpretation of the data is not possible, published analyses of $^{36}\text{Cl}/\text{Cl}$ ratios in cuttings from borehole USW UZ-1 showed the efficacy of this approach.⁷ Ratios measured for depths of 311 and 372 m were considerably less than the modern ratio, suggesting significant ^{36}Cl decay and hence long residence times. Upper age limits of 3 and 8×10^5 yr are calculated for the meteoric Cl content of these samples, respectively, using the model described in section IV.D.1. These residence times correspond to lower bounds on long-term average water velocities of 0.9 and 0.5 mm yr^{-1} at these depths. (Note that shorter residence times, and hence faster velocities, would be calculated if the data could be corrected for dilution of the meteoric ^{36}Cl signal by rock Cl in these samples, using the model described in section IV.D.2. However, the Cl/Br data needed to make such a correction are not available.)

The average infiltration rate can be a useful tool for the investigation of recharge at Yucca Mountain in that it provides an estimation of the net effect of all processes affecting infiltration and subsurface moisture and solute transport. It can, however, be misused if the limitations are not recognized and understood. For example, an estimate of the long-term infiltration rate based upon the decay of the meteoric ^{36}Cl signal does not necessarily reflect present-day infiltration rates. Processes that affect the net infiltration rate include short- and long-term variations in climatic conditions, chloride and ^{36}Cl compositions of meteoric water and dry fallout, vegetation, and geomorphology of surface drainages. A description of the limitations of this approach are discussed in more detail in section V of this paper.

B. Bomb-pulse ^{36}Cl as a water tracer

Infiltration rates in arid soils have been evaluated using bomb-pulse ^{36}Cl as a water tracer in several studies in the past.⁸⁻¹² Only a negligible proportion of the ^{36}Cl measured in these studies was of cosmogenic origin; the isotopic composition of the samples was dominated by bomb-pulse ^{36}Cl . Norris et al. used measurements of bomb-pulse ^{36}Cl in soil samples from two locations near Yucca Mountain to characterize the infiltration of precipitation during the past quarter century and to examine the differences in surficial hydrologic infiltration between the two locations.⁸ The data were interpreted to yield an estimate of the downward liquid infiltration rate at Yucca Wash, located to the east of Yucca Mountain, of 1.8 mm yr^{-1} averaged over 35 years.

C. Sample collection, processing and analysis

Thus far, over 700 soil, rock and groundwater samples have been collected from the Yucca Mountain area for ^{36}Cl analysis as part of site-characterization activities; of these, about 90 have been analyzed (plus

Figure 1. Map showing the location of sampling sites discussed in this paper. (Not shown: VH-1 and JF-3)

numerous replicates), for which locations are shown on Figure 1. Soil samples were collected from pits in Midway Valley. Groundwater samples were collected from the saturated zone by USGS investigators as part of other YMP activities. Rock samples were collected during dry-drilling operations as ream-bit cuttings. Five of the holes were shallow infiltration monitoring boreholes (N-series holes) which extended below the PTn into the top of the TSw. Of these, USW UZ-N11 is located on Mile-High Mesa; USW UZ-N37 is in the bottom of Wren Wash; and USW UZ-N54 is in the bottom of WT2 Wash. USW UZ-N53 and -N55 are adjacent to N54, but on the toes of the ridges that bound the wash. Relative to N54, N53 is 54 m south and 3.0 m higher in elevation, while N55 is 71 m north and 8.1 m higher in elevation. Another suite of analyzed samples, extending from the surface into the Calico Hills Formation, is from UE25 UZ-16, at the mouth of WT2 Wash about 200 m NE of N54.

For soils and ream cuttings, a batch extraction method has been used in which leachable halides are extracted by combining the sample with deionized water, then decanting the solution. Sample size requirements range from as little as 100 g in the case of shallow soil samples

Table 1. Data base for estimate of meteoric $^{36}\text{Cl}/\text{Cl}$ ratio at Yucca Mountain

Field site	LANL ID	Depth (m)	mg Cl /kg	Cl/Br Ratio	Measured $^{36}\text{Cl}/\text{Cl}$ $\times 10^{-15}$
Soil samples from Midway Valley, this study					
MWV-P6	ST-78	3.1	119	210	458 $\pm$ 12
	ST-79	2.75	148	207	462 $\pm$ 7
MWV-P13	ST-80	3.0	36	214	459 $\pm$ 15
	ST-81	2.55	67	203	475 $\pm$ 14
MWV-P23	ST-84	2.6	555	222	483 $\pm$ 11
	ST-85	2.3	401	225	459 $\pm$ 11
MWV-P24	ST-86	2.9	264	223	466 $\pm$ 11
	ST-87	2.5	147	261	524 $\pm$ 10
NPTP-16	ST-88	2.4	335	183	466 $\pm$ 9
	ST-89	2.2	330	192	468 $\pm$ 13
Soil samples from Norris et al. ⁸					
Yucca Wash	Y1	1.8	974	n.m.	455 $\pm$ 64
Exploratory	E2	2.1	504	n.m.	531 $\pm$ 41
Shaft site	E1	2.8	638	n.m.	557 $\pm$ 67
Groundwater sample from Norris et al. ⁷					
J13	—	—	n.m.	n.m.	531 $\pm$ 41
Groundwater samples from this study					
JF3	W004	—	n.m.	n.m.	511 $\pm$ 63
VH-1	W006	—	9	111	540 $\pm$ 11
Alluvial samples from this study					
UZ-N54	R006	4.9	206	195	526 $\pm$ 22
	R007	5.9	148	230	501 $\pm$ 21
	R008	7.3	102	247	507 $\pm$ 14
UNWEIGHTED AVERAGE					493 $\pm$ 34
n.m. - not measured					
Data are preliminary and have not yet been thoroughly reviewed.					

intended to be analyzed solely for Cl/Br ratios, to as much as 15 or more kg of rock material to be analyzed for Cl isotopes. Cl and Br are assayed in the leachates using ion chromatography. (Br data are used in section IV.D.2 to correct measured $^{36}\text{Cl}/\text{Cl}$ ratios for dilution of meteoric Cl by rock Cl.)

Processing of the water and leachate samples for analysis of $^{36}\text{Cl}/\text{Cl}$ ratios involves the addition of silver nitrate to the samples in order to precipitate silver chloride as target material. The $^{36}\text{Cl}/\text{Cl}$ ratios are then measured by accelerator mass spectrometry (AMS)¹³ at one of three laboratories: University of Rochester, Purdue University, and Lawrence Livermore National Laboratory. Frequent submission of blanks and replicates together with interlaboratory comparisons confirm the reproducibility and reliability of these results.

Results are presented for the Midway Valley soils (Table 1), shallow boreholes and UZ-16 (Table 2). These represent only a subset of the total data base insofar as only those Cl concentrations and Cl/Br ratios associated with ^{36}Cl analyses are shown.

Table 2. Halide and ^{36}Cl analyses of borehole cuttings

Sample ID	Top of interval (ft)	mg Cl /kg rock	Cl/Br Ratio	Measured $^{36}\text{Cl}/\text{Cl}$ $\times 10^{-15}$	Ther./ Mech. Unit
USW UZ-N11					
R149	15	n.m.	n.m.	1000 $\pm$ 50	TCw
R152	31	6	113	696 $\pm$ 23	PTn
R154	41	7	129	923 $\pm$ 13	PTn
R157	55	3	964	1082 $\pm$ 37	PTn
R159	65	3	281	13060 $\pm$ 250	PTn
R162	80	4	216	10100 $\pm$ 700	PTn
USW UZ-N37					
R95	5	3	36	1350 $\pm$ 38	UO
R98	11	58	205	1230 $\pm$ 17	UO
R101	20	4	213	1158 $\pm$ 24	UO
R103	24	n.m.	n.m.	1112 $\pm$ 29	UO
R107	30	5	110	35 $\pm$ 64	UO
R112	36	5	260	161 $\pm$ 6	TCw
R285	110	n.m.	n.m.	504 $\pm$ 22	PTn
R288	125	4	78	253 $\pm$ 10	PTn
R293	148	4	423	327 $\pm$ 6	PTn
R298	174	6	226	847 $\pm$ 12	PTn
R301	189	5	n.m.	575 $\pm$ 124	PTn
R307	217	4	41	850 $\pm$ 105	PTn
R312	242	7	844	269 $\pm$ 6	PTn
Note: PTn/TSw1 contact @ 256.7 ft					
USW UZ-N53					
R349	124	6	376	2006 $\pm$ 69	TCw
R351	134	n.m.	n.m.	2050 $\pm$ 90	TCw
R353	144	3	248	4561 $\pm$ 130	PTn
R361	183	6	126	2369 $\pm$ 34	PTn
R364	198	3	168	710 $\pm$ 36	PTn
R366	208	10	301	522 $\pm$ 17	PTn
Note: PTn/TSw1 contact @ 230.7 ft					
USW UZ-N54					
R002	5	9	80	3719 $\pm$ 175	UO
R003	7	62	180	1098 $\pm$ 33	UO
R004	10	84	166	687 $\pm$ 35	UO
R005	12	152	208	556 $\pm$ 21	UO
R006	15	206	195	526 $\pm$ 22	UO
R007	17	148	230	501 $\pm$ 21	UO
R008	22	102	247	507 $\pm$ 14	UO
R033	135	10	300	277 $\pm$ 19	PTn
R034	140	9	267	149 $\pm$ 22	PTn
R117	165	8	60	402 $\pm$ 9	PTn
R121	184	7	222	480 $\pm$ 16	PTn
R125	204	9	81	474 $\pm$ 49	PTn
R128	218	2	93	352 $\pm$ 13	PTn
R130	228	2	124	332 $\pm$ 16	PTn
Note: PTn/TSw1 contact @ 230.5 ft					
USW UZ-N55					
R035	0	2	192	1747 $\pm$ 25	UO
R040	9	3	225	5565 $\pm$ 220	TCw
R072	165	4	136	14740 $\pm$ 640	PTn
R073	170	4	143	11010 $\pm$ 190	PTn
R074	174	4	180	27040 $\pm$ 330	PTn

Sample ID	Top of interval (ft)	mg Cl /kg rock	Cl/Br Ratio	Measured $^{36}\text{Cl}/\text{Cl}$ $\times 10^{-15}$	Ther./ Mech. Unit
USW UZ-N55 (continued)					
R075	180	2	131	13157 $\pm$ 184	PTn
R078	189	8	300	1413 $\pm$ 57	PTn
R081	203	4	126	22300 $\pm$ 345	PTn
R084	218	8	161	17168 $\pm$ 1202	PTn
R087	232	5	196	6580 $\pm$ 364	PTn
R088	237	6	109	8203 $\pm$ 140	PTn
R089	242	3	109	9187 $\pm$ 220	PTn
R090	247	2	194	10480 $\pm$ 180	TSw1
R091	252	2	229	17050 $\pm$ 230	TSw1
Note: PTn/TSw1 contact @ 245.4 ft					
UE25 UZ-16					
R163	0	8	147	2280 $\pm$ 150	Fill
R165	7	58	170	615 $\pm$ 32	UO
R167	12	123	193	526 $\pm$ 16	UO
R168	14	172	198	500 $\pm$ 15	UO
R169	16	158	192	528 $\pm$ 24	UO
R170	17	104	191	750 $\pm$ 35	UO
R173	22	56	197	601 $\pm$ 26	UO
R174	24	25	179	507 $\pm$ 26	UO
R177	30	25	207	572 $\pm$ 22	UO
R178	39	7	187	1030 $\pm$ 60	UO/TCw
R182	143	1	194	106 $\pm$ 40	PTn
R183	153	1	96	346 $\pm$ 27	PTn
R184	158	1	67	329 $\pm$ 24	PTn
R185	163	1	67	338 $\pm$ 20	PTn
R186	181	2	93	306 $\pm$ 21	PTn
R187	219	2	97	434 $\pm$ 26	PTn
R205	669	1	195	145 $\pm$ 15	TSw2
R219	1090	2	213	123 $\pm$ 7	TSw2
R220	1110	3	315	117 $\pm$ 8	TSw2
R221	1122	1	157	150 $\pm$ 9	TSw3
R224	1166	1	n.m.	272 $\pm$ 16	CHn1
R225	1170	1	142	363 $\pm$ 34	CHn1
n.m. - not measured					
Notes:					
1. Thermal/mechanical units: ^{5,15} UO - Undifferentiated overburden; TCw - Moderately to densely welded Tiva Canyon; PTn - Non to moderately welded and bedded zones of Tiva Canyon, Yucca Mountain, Pah Canyon and Topopah Spring members; TSw1 - Moderately to densely welded Topopah Spring member; TSw2 - Densely welded Topopah Spring, potential repository horizon; TSw3 - Lower vitrophyric zone, Topopah Spring; CHn1 - Partially welded to nonwelded and bedded zones of Topopah Spring member, tuffaceous beds of Calico Hills Formation					
2. Preliminary estimates of unit contacts provided by D.C. Buesch. ¹⁴					
3. In some cases, Cl and Br concentrations were measured on different sample aliquots than those used to obtain ^{36}Cl analyses. Where more than one ^{36}Cl analysis is available for a given sample, a weighted average ratio is reported.					
4. These data are preliminary and have not yet been thoroughly reviewed.					

IV. INTERPRETATION

A. Meteoric background Cl/Br and $^{36}\text{Cl}/\text{Cl}$ ratios

The meteoric Cl/Br ratio is estimated to be 177 ± 56 , based on analyses of 40 soil samples from Midway Valley plus 67 alluvial samples from boreholes UZ-16 and N37, N54 and N55. The large variability is attributable to analytical difficulties in measuring the extremely low Br concentrations in some of these samples. No systematic variability with depth or location is apparent in these data. The meteoric $^{36}\text{Cl}/\text{Cl}$ ratio is estimated to be $4.9 \pm 0.3 \times 10^{-13}$ based on an unweighted average of 19 soil, alluvial and water samples (Table 1).

B. Infiltration processes in alluvium

Several approaches have been used to derive information about shallow infiltration processes from Cl and ^{36}Cl data. Those discussed below include: (1) the Cl mass balance age, based upon the time required to accumulate the mass of Cl measured in the profile; (2) qualitative information based upon the shape and magnitude of the bomb-pulse profile and the depth to the center of mass of the ^{36}Cl bomb pulse.

1. *Cl mass balance ages.* Stable Cl concentration profiles in alluvium have been widely used to estimate water transport rates in the unsaturated zone.^{9,11,16-19} The travel time of Cl in the profile, assumed to represent the travel time of water, is evaluated according to

$$t = \frac{\sum_i (\text{Cl}_i z_i \rho_b)}{P \text{Cl}_p} \quad (1)$$

where Cl_i is the Cl concentration in interval i (g Cl g^{-1}), z_i is the thickness of interval i (m), ρ_b is bulk density (g m^{-3}), P is mean annual precipitation rate (m yr^{-1}), and Cl_p is the mean Cl concentration in precipitation (g Cl m^{-3}), including the contribution from dry fallout. The summation term $\sum_i (\text{Cl}_i z_i \rho_b)$ is the cumulative total mass of Cl from the surface to depth z_i (g Cl m^{-2}). The parameter Cl_p may be estimated by dividing the natural ^{36}Cl fallout at the site by the product of mean annual precipitation and the natural (prebomb) $^{36}\text{Cl}/\text{Cl}$ ratio for that site.⁹⁻¹⁰ The approach accounts for loss of water by evapotranspiration but assumes that Cl acts conservatively. For the vicinity of Yucca Mountain, the estimated Cl accumulation rate ($P \text{Cl}_p$) is $\sim 106 \text{ mg m}^{-2} \text{ yr}^{-1}$, with a corresponding concentration in precipitation of 0.62 mg L^{-1} for Cl_p . This estimate is based on an average annual precipitation rate of 17 cm yr^{-1} ,⁶ a prebomb $^{36}\text{Cl}/\text{Cl}$ ratio of 4.9×10^{-13} (this study), and a natural ^{36}Cl fallout rate of $28 \text{ atoms } ^{36}\text{Cl m}^{-2} \text{ s}^{-1}$.

Figure 2 shows Cl mass balance ages for the alluvial profiles in N37, N54 and UZ-16. The maximum Cl accumulation age at the base of the alluvium in N37 (11 m, 36.5 ft) is 230 years. In contrast, age profiles for

UZ-16 and N54 are similar to one another, attaining maximum ages of $\sim 1000 \text{ yr}$ at depths of 7.6 m (25.5 ft). Clearly, based on the Cl mass balance ages, the average downward rate of infiltration is significantly slower in the latter two holes than in the former.

2. *Bomb-pulse ^{36}Cl .* Figure 3 plots the measured $^{36}\text{Cl}/\text{Cl}$ and bomb-pulse ^{36}Cl profiles for N37 and N54. The integrals of the bomb-pulse ^{36}Cl peaks in these two profiles are 2.3 and $2.1 \times 10^{12} \text{ atoms m}^{-2}$, respectively, agreeing with the expected integral for total global fallout at the site.^{9,11} There is no evidence for lateral transport of Cl into or out of the profile, nor for contamination by ^{36}Cl fallout from local Nevada Test Site activities. The center of mass for the bomb-pulse ^{36}Cl profile lies within the depth intervals of 3.0 to 4.6 m (10 to 15 ft) for N37 and 2.1 to 4.6 m (7 to 10 ft) for N54, still within the zone of evapotranspiration (ET). This distribution suggests that only a negligible proportion of precipitation over the past 35 years has infiltrated below the ET zone. However, the depth of penetration is still considerably greater than that measured at other arid Southwestern sites (e.g., at Yucca Wash, the peak occurred at a depth of 0.5 m ⁸), which may be evidence for contribution from runoff in the washes.

The monotonic decrease from a peak value in the $^{36}\text{Cl}/\text{Cl}$ profile in N54 is a shape typically observed in other soil studies, but that for N37 is odd in that the ratio remains fairly constant over a 6-m (20-ft) interval. A possible explanation is that the profile was disturbed by a recent and extreme runoff event in this wash, that not only smeared out the ^{36}Cl profile but also removed much of the accumulated Cl, such that the relatively young Cl mass balance age may be a recent phenomenon. One possible hypothesis that could account for this difference would be if, over the past 35 years, a greater contribution

Figure 2. Chloride mass balance ages in N37, N54, and UZ-16, based upon the quantity of chloride accumulated in these alluvial profiles using eqn (1) and assuming an accumulation rate of $106 \text{ mg m}^{-2} \text{ yr}^{-1}$. Sample depths for UZ-16 have been adjusted by subtracting the presence of 6 ft of fill.

of runoff (with negligible Cl and ^{36}Cl content) has entered the alluvium at N37 in Wren Wash, perhaps as a consequence of its larger catchment area and higher average precipitation rate, as compared to that contributing to the alluvium at N54 and UZ-16 in WT2 Wash.

The UZ-16 alluvial profile (Table 2) does not show a distinct bomb-pulse peak. The highest $^{36}\text{Cl}/\text{Cl}$ ratio is seen in the shallowmost sample, which consists entirely of fill material used to build up the drill pad. Presumably, the bomb-pulse lies within intervals that have not yet been analyzed, e.g., the unanalyzed intervals at 5.8 to 6.7 m (4.0 to 4.9 m, corrected for the 1.8-m cover of fill material) are bracketed by intervals with ^{36}Cl concentrations that are slightly elevated above background. A bomb-pulse signal is also detected at the alluvial/TCw contact (sample R178). This distribution suggests a possible fast transport path through the alluvium to this point or, perhaps more likely, that the alluvium may be bypassed altogether as a result of lateral transport of water along the alluvial/TCw contact, fed by infiltration along sideslopes where alluvial cover is thin or absent.

C. Evidence for fast transport of water into PTn

Possible evidence for fast transport of water into the PTn is shown by detection of elevated levels of ^{36}Cl in N11, N37, and N53 in samples from this nonwelded unit. Transport through fractures in the overlying TCw is a possible explanation and is supported by neutron logging of N53 which has shown changes in moisture content down to a depth of 12 m, well into the TCw.²⁰ No evidence of bomb-pulse is seen in the PTn in N54 and UZ-16, despite their proximity to N53 with its sizable ^{36}Cl signal, suggesting that the occurrence of fracture flow is

Figure 3. Profiles of $^{36}\text{Cl}/\text{Cl}$ and calculated bomb-pulse ^{36}Cl concentrations in boreholes N37 and N54. Data from Table 2; $^{36}\text{Cl}/\text{Cl}$ ratios for N37 samples that have not been analyzed have been estimated by averaging results from adjacent intervals.

heterogeneous and perhaps related to the depth of the overlying alluvial cover. Alternatively, the possibility of contamination during drilling cannot be ruled out for these holes. Extensive testing has verified that no contamination occurs during sample preparation and analysis in the laboratory, but the $^{36}\text{Cl}/\text{Cl}$ ratios measured in the N55 samples (Table 2) are considerably higher than can be explained by global fallout of ^{36}Cl .

D. Residence time estimates

For rock samples in which bomb-pulse ^{36}Cl is absent, ^{36}Cl and Cl/Br data from drillhole samples can be used to establish limits on water movement rates in the unsaturated zone based upon two approaches. The first approach, called the Limiting Model, uses the measured $^{36}\text{Cl}/\text{Cl}$ ratio to estimate an upper limit for the water residence time, or the lower limit for the rate of water movement. In this case, the rate of water movement could be faster, but not slower, than that indicated by the measured $^{36}\text{Cl}/\text{Cl}$ ratio. The second approach, called the Dilution Model, corrects the measured $^{36}\text{Cl}/\text{Cl}$ ratio for dilution of the meteoric Cl by rock Cl. These two approaches are described below, with results of model calculations reported in Table 3 for borehole samples.

1. *The Limiting Model.* In the simplest case, the measured $^{36}\text{Cl}/\text{Cl}$ ratios can be used to estimate upper limits for average water residence time at the sampled depths, and hence lower limits for the net downward velocities or fluxes to those depths. The linear downward velocity is defined as vertical depth divided by ^{36}Cl age and thus implies the assumption that water moves vertically through the matrix. The linear velocity can be used to estimate the Darcy velocity or Darcy flux by multiplying the velocity by the volumetric water content of the intervening rock units; this conversion of results may not be straightforward where water contents vary considerably from one unit to another.

These ^{36}Cl -based age estimates assume that the initial meteoric $^{36}\text{Cl}/\text{Cl}$ ratio is known and constant during the period of interest. For old water, the buildup of ^{36}Cl as a result of the subsurface neutron flux must also be taken into account. The secular equilibrium $^{36}\text{Cl}/\text{Cl}$ ratio is on the order of 2×10^{-14} . An age estimate is obtained by applying the standard equation for radioactive decay with correction for ingrowth,¹

$$t = -\frac{1}{\lambda_{36}} \ln \left[\frac{(R_x)_{36} - (R_r)_{36}}{(R_0)_{36} - (R_r)_{36}} \right] \quad (2)$$

where λ_{36} is the decay constant for ^{36}Cl , $2.30 \times 10^{-6} \text{ yr}^{-1}$, and $(R_x)_{36}$, $(R_r)_{36}$, and $(R_0)_{36}$ are the measured, rock, and initial meteoric $^{36}\text{Cl}/\text{Cl}$ ratios, respectively.

2. *The Dilution Model.* A more realistic estimate of residence time can be obtained by correcting the measured $^{36}\text{Cl}/\text{Cl}$ ratio for dilution of the meteoric

component by rock Cl, much of which may have been introduced into the sample as a consequence of drilling and crushing in the laboratory. Rock Cl generally has a $^{36}\text{Cl}/\text{Cl}$ value that is more than an order of magnitude lower than that of meteoric Cl. It is hypothesized that the proportion of rock Cl may be estimated by assuming two-component mixing between meteoric and rock Cl. Each of these end-members is assumed to have a characteristic and constant Cl/Br ratio, and the rock component is also assumed to have a characteristic and constant $^{36}\text{Cl}/\text{Cl}$ ratio.

According to this dilution model, the measured Cl/Br and $^{36}\text{Cl}/\text{Cl}$ ratios, $(R_x)_i$, are determined from the generic mixing equation,

$$(R_x)_i = f_m (R_m)_i + f_r (R_r)_i \quad (3)$$

where R_m and R_r are ratios for meteoric and rock Cl, respectively, and f_m and f_r are the relative proportions of Cl derived from meteoric and rock sources, respectively, and are assumed to sum to 1. The subscript i refers to the type of ratio: $^{36}\text{Cl}/\text{Cl}$ for $i = 36$, CB for $i = \text{Cl}/\text{Br}$. The value of f_m and hence f_r can be estimated from the measured Cl/Br ratios by solving the mixing equation for f_m ,

$$f_m = \frac{(R_x)_{CB} - (R_r)_{CB}}{(R_m)_{CB} - (R_r)_{CB}} \quad (4)$$

Once a value of f_m has been estimated for a given sample, the meteoric $^{36}\text{Cl}/\text{Cl}$ ratio, $(R_m)_{36}$, is estimated from the mixing equation,

$$(R_m)_{36} = \frac{(R_x)_{36} - f_r (R_r)_{36}}{f_m} \quad (5)$$

The corrected meteoric $^{36}\text{Cl}/\text{Cl}$ ratio is then substituted for $(R_x)_{36}$ in eqn (2) to derive a residence time estimate. Figure 4 illustrates the effect of this correction on the residence time estimate as a function of measured $^{36}\text{Cl}/\text{Cl}$ ratio and extent of dilution by rock Cl.

3. *Residence time estimates derived from models.* Application of these two models to ^{36}Cl data from UZ-16 and N54 suggests average residence times of 1 to 3 x 10⁵ yr in the PTn, 5 to 7 x 10⁵ yr in the deeper TSw2 and TSw3, and 1 to 3 x 10⁵ yr in the CHn (Table 3). Based upon propagation of errors, these residence time estimates may have uncertainties as great as 50%. Major problems in the interpretation include uncertainties in the meteoric and rock Cl/Br ratios, and analytical problems with Br determinations that could be causing erroneous Cl/Br ratios for specific samples. Also, it is troublesome that Cl/Br ratios in many of the PTn samples are considerably less than the meteoric average, suggesting that the two-component mixing model with constant end-member values may oversimplify the geochemistry of these halides in this hydrogeologic environment.

Table 3. Residence time estimates based on measured and corrected $^{36}\text{Cl}/\text{Cl}$ ratios

Sample ID	Cl/Br Ratio	Est f_m	$^{36}\text{Cl}/\text{Cl}$ ratio $\times 10^{-15}$		Residence time (10^3 yr), based on:		Ther./ Mech. Unit
			Meas.	Corr.	Meas. ratio	Corr. ratio	
UE25 UZ-16							
R182	194	0.97	106	109	747	732	PTn
R183	96	1.00	346	346	168	168	PTn
R184	67	1.00	329	329	191	191	PTn
R185	67	1.00	338	338	179	179	PTn
R186	93	1.00	306	306	225	225	PTn
R187	97	1.00	434	434	64	64	PTn
R205	195	0.96	145	150	584	568	TSw2
R219	213	0.92	123	132	668	633	TSw2
R220	315	0.68	117	163	694	526	TSw3
R221	157	1.00	150	150	567	567	TSw3
R224	133	1.00	272	272	280	280	CHn1
R225	142	1.00	363	363	146	146	CHn1
USW UZ-N37							
R112	260	0.81	167	201	514	424	TCw
R285	n.m.	1.00	504	504	< 50	< 50	PTn
R288	78	1.00	253	253	314	314	PTn
R293	423	0.42	327	750	194	Modern	PTn
R298	226	0.89	847	949	Modern	Modern	PTn
R301	n.m.	1.00	575	575	Modern	Modern	PTn
R303	41	1.00	850	850	Modern	Modern	PTn
R312	844	< 0.1	269	2510	285	Modern	PTn
USW UZ-N54							
R033	300	0.71	277	306	271	123	PTn
R034	267	0.79	149	194	571	469	PTn
R117	60	1.00	402	402	99	99	PTn
R121	222	0.90	480	532	18	< 50	PTn
R125	81	1.00	367	367	141	141	PTn
R128	93	1.00	352	352	160	160	PTn
R130	124	1.00	332	332	187	187	PTn

Notes on derivation of calculated ages:

1. Assumed end-member values: Rock Cl/Br = 600; Meteoric Cl/Br = 180; Rock $^{36}\text{Cl}/\text{Cl}$ = 20 x 10⁻¹⁵; Meteoric $^{36}\text{Cl}/\text{Cl}$ = 500 x 10⁻¹⁵
2. f_m calculated using Eqn 4. Where measured Cl/Br ratio < 180, assumed f_m = 1.0
3. Corrected $^{36}\text{Cl}/\text{Cl}$ calculated using Eqn 5. Where corrected $^{36}\text{Cl}/\text{Cl}$ > 500 x 10⁻¹⁵, assumed age is modern (post-bomb)
4. Residence times calculated using Eqn 2.
5. Residence time estimates are subject to change as the $^{36}\text{Cl}/\text{Cl}$ and Cl/Br data base is updated for end-member ratios.

Nonetheless, although estimates for individual samples may be suspect, one cannot help but be struck by the irregular profile of ages with depth in any given borehole. The irregularity suggests a highly heterogeneous flow system, with multiple flow paths

contributing to moisture present at any given point. This inevitable mixing highlights the problematic issue of the value of attempting to define a single age for water and suggests that hydrologic models that consider only one-dimensional processes such as vertical infiltration would be inappropriate for this system. Rather, two-dimensional or even three-dimensional representations that can account for such processes as dispersion and lateral flow are necessary.

A second observation is that the apparently younger ages observed in the CHn samples compared with the TSw2 and TSw3 samples in UZ-16 would imply that it is inconsistent to interpret the ^{36}Cl -based ages for the CHn samples in terms of vertically downward velocity estimates, i.e., that the water in the CHn at this location may not derive predominantly from matrix flow through the overlying TSw, but rather by way of some faster transport path. One possibility is downward fluid flow through the probable significant fault zone penetrated by UZ-16 at ~351 to 364 m (1150 to 1195 ft),¹⁴ although hydrologic modeling by Wittwer et al.²¹ indicated that such a fault zone would be unlikely to enhance infiltration to this depth. A more likely pathway would be lateral transport down-dip from the direction of the Solitario Canyon Fault Zone.

V. LIMITATIONS OF APPROACH

The interpretation of data as described in the previous section must recognize the limitations of the technique as well as the simplifying assumptions implicit in the models.

Effective dating range. The $^{36}\text{Cl}/\text{Cl}$ analyses can provide useful information for unsaturated zone studies only if an unambiguous bomb-pulse signal is detected, or if an unambiguous decay of the meteoric signal can be shown.

Figure 4. Calculated residence time as a function of measured $^{36}\text{Cl}/\text{Cl}$ ratio and extent of dilution of meteoric chloride by rock chloride. Plot assumes $^{36}\text{Cl}/\text{Cl}$ ratios of 5×10^{-13} and 2×10^{-14} for meteoric and rock end-members, respectively; and Cl/Br ratios of 170 and 600 for these end-members.

The method is insensitive if the hydrologic response time is greater than forty or less than about 5×10^4 yr.

Flow field. Models for estimating infiltration rates based upon the depth of bomb-pulse ^{36}Cl or upon the accumulation of Cl in soil profiles assume one-dimensional, vertically downward, piston-type flow at steady state.^{19,22-23} However, as long as the ^{36}Cl and chloride reside within the zone of evapotranspiration, root extraction of water subjects them to a highly nonuniform velocity field.²⁴ The presence of lateral flow, preferential flowpaths, and upward gradients may also invalidate these models. A similar limitation applies to the estimation of linear velocities and fluxes based on residence times calculated from ^{36}Cl decay. In addition, the existing moisture distribution may be a relic of past climatic conditions significantly different from those of the present-day, thereby invalidating the assumption of steady-state.

Constancy of end-member values. A premise of data interpretation for this study is that the meteoric $^{36}\text{Cl}/\text{Cl}$ ratio has been constant throughout the Quaternary Period, i.e., that ^{36}Cl production rates in the atmosphere and the rate of fallout of stable Cl at the site have been constant. Both of these assumptions are likely to be invalid to some extent; what is not known is the magnitude of the deviations from constancy. Phillips et al.²⁵ have suggested that ^{36}Cl production rates may have been as much as 40% greater 20,000 years ago.

The model presented earlier also assumes that Cl/Br ratios of the meteoric and rock end-members are constant values. However, end-member ratios for the tuffs may vary among the various lithologic units, or as a function of the extent of hydrothermal alteration of a given unit or subunit. The meteoric ratio may have been different in the past at Yucca Mountain under different climatic regimes when different storm tracks may have been dominant because the Cl/Br ratio varies as a function of distance from the oceanic source. It is also possible that marine aerosols are not the important determinant of meteoric Cl deposition rates at Yucca Mountain,²⁶ in which case other factors may govern its accession rate and variability in halide ratios.

Differential transport rates for Cl and water. Interpretation of the ^{36}Cl data in terms of the rate of water movement through the unsaturated zone requires the consideration of processes that may differentiate Cl movement from water movement. Theoretically, the rate of movement of a tracer introduced into the unsaturated zone, even a perfectly conservative tracer, will lag behind that of the water front because of dilution of the tracer with the initial water content. Hence, tracer movement will approximate water movement only if the initial water content is negligible or if changes in moisture content are sufficiently small, i.e., it will lag behind water movement under sudden recharge events. Another aspect of Cl movement that is important for hydrologic modeling and that is not

negligible arises from the nonvolatile character of these anions. Water movement through the unsaturated zone may occur in downward pulses through the matrix in the liquid phase, followed by upward movement in the vapor phase, but Cl ions move only in the liquid phase. Hence, while the net flux might actually be upwards, the ^{36}Cl data can only provide an estimate of the downward component. In addition, the ^{36}Cl decay data will probably reflect an age that results from mixing of Cl from more than one pulse. Cl may also be concentrated by plant root activity, which could retard the rate of Cl movement relative to that of water.

Propagation of uncertainties. The large uncertainty in the end-member Cl/Br ratios, together with analytical problems in measuring Cl/Br ratios in some of these samples, causes uncertainties as great as 50% for the residence time calculations.

Contamination. The $^{36}\text{Cl}/\text{Cl}$ profile measured for N55 (Table 2) shows values considerably higher than can be explained by global fallout of ^{36}Cl . Until the source of these elevated ^{36}Cl signals can be identified, the ^{36}Cl results in the other N-holes are also suspect. Tests now being conducted to evaluate alternative sources include comparisons of ^{36}Cl contents of drillcore to those measured in ream cuttings, ^{36}Cl profiles in soil collected from the vicinity of a suspected local source, and tritium analyses of moisture extracted from drillcore.

VI. SUMMARY AND CONCLUSIONS

About 90 soil, water and rock samples have been analyzed for ^{36}Cl as part of the Yucca Mountain Site Characterization Project. From a subset of these, the background meteoric $^{36}\text{Cl}/\text{Cl}$ ratio is estimated to be $4.9 \pm 0.3 \times 10^{-13}$. Additional samples have been analyzed for Cl and Br concentrations in order to determine background Cl/Br ratios in meteoric water. Based on 107 samples of local soil, alluvium and groundwater, the meteoric Cl/Br ratio is estimated to be 177 ± 56 . The large variability is attributable to analytical difficulties in measuring the extremely low Br concentrations. This parameter is needed in order to correct measured $^{36}\text{Cl}/\text{Cl}$ ratios in rock samples for dilution of the meteoric component by rock Cl before calculating residence times from these data.

Cl concentrations and the $^{36}\text{Cl}/\text{Cl}$ ratio in alluvium penetrated by drillholes are being used to provide information on characteristics of water movement through the unsaturated zone at Yucca Mountain. Bomb-pulse ^{36}Cl is detected in the alluvium in 2 holes to depths of 4.6 and 7.9 m; below those depths, it is present in the alluvium only at background levels. This deep penetration of moisture provides evidence for the variable effectiveness of alluvium and associated vegetation in attenuating infiltration, possibly as a result of different runoff characteristics of the watersheds. Accumulation of Cl in these alluvial profiles also provides evidence that

infiltration rates below the zone of evapotranspiration are negligibly small under these conditions. However, an elevated ^{36}Cl signal at the base of the alluvium in UZ-16 may indicate lateral flow along the alluvial/TCw contact, fed by infiltration along sideslopes where alluvial cover is thin or absent.

Possible evidence for fast transport of water via fractures through the welded TCw is shown by detection of elevated levels of ^{36}Cl in the underlying nonwelded PTn in three of five boreholes (N11, N37 and N53). However, because ^{36}Cl levels in another borehole, N55, were higher than could be accounted for by global fallout, the possibility that elevated levels in any of these holes may also be attributable to contamination cannot as yet be ruled out. No evidence for bomb-pulse in the PTn is seen in UZ-16 and N54, which indicates that fracture transport through the TCw might be irregular in occurrence.

Preliminary interpretation of ^{36}Cl data for boreholes UZ-16 indicates apparent average residence times of 1 to 3×10^5 yr in the PTn, 5 to 7×10^5 yr in the deeper welded TSw2 and TSw3, and 1 to 3×10^5 yr in the nonwelded CHn. The apparently younger ages observed in the CHn samples compared with the TSw samples suggest that the water in the CHn at this location may not derive predominantly from matrix flow through the overlying TSw, but rather that a faster transport path may be involved such as lateral transport downdip from the direction of the Solitario Canyon Fault Zone.

These residence time estimates may be a useful tool for the investigation of recharge at Yucca Mountain in that they reflect the net effect of all processes affecting infiltration and subsurface moisture and solute transport. They can, however, be misused if the limitations are not recognized and understood. For example, long-term infiltration rates based upon these residence time estimates may not necessarily reflect present-day infiltration rates. Processes that affect the net infiltration rate include short- and long-term variations in climatic conditions, Cl and ^{36}Cl compositions of meteoric water and dry fallout, vegetation, and the ever-changing geomorphology of surface drainages.

The emerging picture from these data is that of a highly heterogeneous flow system, with multiple flow paths contributing to the moisture present at any given point. This inevitable mixing highlights the problematic issue of the value of attempting to define a single age for water. In any case, the ^{36}Cl data must be examined in concert with other independent lines of evidence—such as neutron moisture logs, lithologic logs, hydraulic parameters, and other isotopic and geochemical data—in order to ensure that the interpretation is consistent with all available information.

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APPENDIX E

LIST OF LETTER REPORTS SUBMITTED TO LANL
UNDER SUBCONTRACT 9-XG1-N3993-1

Date of letter	Reporting period	# pages
08/16/90	May through July 1990	1
01/15/91	October through December 1990	3
05/15/91	March through April 1991	1
12/05/91	May through October 1990	1
01/06/92	November 1991	1
02/25/92	December 1991 to January 1992	1
04/01/92	February 1992	1
04/24/92	March 1992	1
05/25/92	April 1992	1
07/21/92	June 1992	3
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04/30/93	April 1993	2
05/28/93	May 1993	2
06/23/93	Calendar Year 1992	5
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10/01/93	September 1993	2
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