AIIM

Association for Information and Image Management

1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

Centimeter

Inches

MANUFACTURED TO AIIM STANDARDS
BY APPLIED IMAGE, INC.

1 of 2

CONFIDENTIAL

4510 '91 MAY 29 A10:37

**END-OF-CONTRACT REPORT
FOR
CONTRACT # 9-XDD-6329F-1
BETWEEN
THE REGENTS OF THE UNIVERSITY OF CALIFORNIA
(LOS ALAMOS NATIONAL LABORATORY)
AND
HYDRO GEO CHEM**

TWS-INC-7-05-91-10

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Prepared for:

Los Alamos National Laboratory
Los Alamos, New Mexico

Prepared by:

Hydro Geo Chem
1430 North Sixth Avenue
Tucson, Arizona 85705

May 17, 1991

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

CONTENTS

Introduction	1
Summary of Work Performed	1
Scope of Work Reviewed	2
Task 1	2
Task 2	2
Task 3	3
Task 4	3
Task 5	3
Task 6	3
Task 7	4
Task 8	4
Task 9	4
Appendix A: Formal Reports, Letter Reports, and Letters Accompanying Invoices	
Appendix B: Publications	
Appendix C: Sample Inventory	

INTRODUCTION

This document is an end-of-contract report, prepared by Hydro Geo Chem for Los Alamos National Laboratory under contract number 9-XDD-6329F-1. The ultimate goal of this work is to characterize historical infiltration and unsaturated flow in the Yucca Mountain area of the Nevada Test Site. Work on this contract has focused on using chloride, bromide, stable chlorine isotopes, and chlorine-36 distributions to evaluate the depth of infiltration in the unsaturated zone. Effort in support of this work has included developing analytical procedures, exploring ways in which to separate the meteoric component from the rock component, and meeting quality assurance requirements.

SUMMARY OF WORK PERFORMED

Samples analyzed during the course of this contract were collected from the UZN-43 borehole, in the interval from 6 to 41 feet below land surface, and from the UZ-1 borehole, from a depth of 97 feet to 1270 feet. Samples were also collected from various locations within the U12G Tunnel. The Tunnel is a horizontal shaft in the side of Rainier Mesa, with approximately 1300 feet of overburden. Additional samples were collected from Trench 14 to help identify the origin of the carbonate veins.

The scope of work proposed in the original Request for Proposal included laboratory measurement of chloride from soil and rock samples to help determine solute transport by water, stable chlorine isotope analyses of chloride leached from soil and rock samples to distinguish chloride of meteoric origin from chloride originating from other sources, and chlorine-36 analyses to investigate the rate and mechanism of chloride and water transport. Also included in the scope of work were field surveys and data collection at Yucca Mountain and the surrounding vicinity to identify areas of infiltration, hydrologic evaluation to guide field surveys and sampling, and interpretation of data in terms of solute transport in the vadose zone.

Progress on this contract has been sporadic although most of the tasks listed in the original scope of work have been addressed. For whatever reasons, funding has not been continuous; at times, as much as nine months have passed between completion of one fiscal year and availability of the following year's money. As a result, work on the contract has tended to proceed in spurts and consequently, less has been accomplished than originally anticipated. In addition, access to the Yucca Mountain site or to samples from the site has been limited.

As might be expected, the direction of the project has changed several times as new information has become available. While this has provided us with valuable insight in terms of defining future scope and direction of the project, it has slowed progress. For example, it was originally believed that total chloride was the parameter of interest in estimating infiltration. As a result, time, energy, and money were devoted to the task of designing a rock melting apparatus to determine total chloride content. Later, it was discovered that soluble chloride and bromide were more appropriate indicators of infiltration and a step-leaching procedure was designed to gather this information.

Initially work on this contract focused on the UZ-1 borehole samples. Then, considerable time, at LANL's direction, was put into exploring the chlorine-36 distribution in the U12G Tunnel. When Apache Leap samples were anticipated, it was thought that the focus of the project would shift to those samples which presumably were not contaminated during the drilling process. However, subsequent evaluation indicated that the Apache Leap site was far too wet for its samples to be useful as substitutes for the Yucca Mountain samples.

SCOPE OF WORK REVIEWED

The nine tasks addressed in the original scope of work are listed below with accompanying discussion regarding how they have been addressed in the work performed.

Task 1 Review of available geologic and hydrogeologic information concerning Yucca Mountain and vicinity.

This task has been partially addressed, during both the previous contract and the early stages of this contract. However, additional work involving a more extensive review and compilation of geologic and hydrogeologic information would be useful.

Task 2 Review and preparation of report on all available information on chloride and chlorine-36 measurements performed to date on soil and rock samples at the Yucca Mountain site.

Much of the previous chloride and chlorine-36 measurements performed on soil and rock samples from Yucca Mountain were done by Hydro Geo Chem. Those data were formally presented in the final report prepared by Hydro Geo Chem for LANL under contract 9-X14-J7419-1.

Task 3 Planning of experiments and development of procedures for the exploratory shaft project.

All of the experimental and developmental work that has been performed on this contract is for the express purpose of evaluating infiltration in the unsaturated zone. As such, it is applicable to all samples whether collected from a trench, a deep borehole, or the exploratory shaft. However, specific procedures outlining the details for collecting samples in the exploratory shaft have not been prepared. It is not possible to define such procedures until additional information regarding the specifics of the exploratory shaft is made available.

Task 4 Analyses of chloride from core material to measure the distribution of chloride with sample depth.

The distribution with depth of chloride and bromide in both the UZ-1 and the UZN-43 boreholes have been determined using cuttings from those boreholes.

Task 5 Analyses of stable chlorine isotopes in soil and rock to distinguish chloride of meteoric origin from chloride of other potential origins.

The ratio of chloride to bromide (Cl/Br) has been evaluated as a tool to distinguish the meteoric component of chloride from the rock component. The Cl/Br ratio was chosen over the stable chlorine isotope ratio because it a less time-consuming and more cost-effective approach. It has proven itself to be useful in this endeavor; however, additional work is needed to refine the procedure. Work has begun comparing the effectiveness and sensitivity of the two approaches.

Task 6 Analyses of chlorine-36 to determine age of chloride at Yucca Mountain and to investigate the rate of moisture and chloride transport.

Chlorine-36 has been analyzed at several depths throughout the UZ-1 borehole. Qualitative results indicate meteoric chloride in the shallow soil environment and chlorine-36 in the deeper profile approaching cosmogenic levels. However, isolated occurrences of chlorine-36 bomb-pulse at depth indicate a relatively rapid transport mechanism connecting the shallow soil environment with the deep profile.

Task 7 Field survey of Yucca Mountain and vicinity to identify areas of infiltration.

It has not been possible to conduct a field survey of Yucca Mountain and vicinity, due to a history of denied access to the Nevada Test Site.

Task 8 Hydrologic interpretation of the data in terms of flow and solute transport.

Due to the intermittent progress on this project and the amount of time devoted to developing procedures, insufficient data have been collected to support a quantitative interpretation of flow and solute transport. However, a brief and on-going qualitative interpretation of flow and solute transport has been possible based on the depth of infiltration of the chlorine-36 bomb-pulse.

Task 9 Formal reporting of information to LANL.

Copies of several reports prepared during this contract have been included in Appendix A as part of this document. These reports document analytical results and provide suggestions for future work. They are listed below with a brief abstract summarizing their content.

Letter report, September 29, 1988: This letter report summarized work performed during fiscal year 1988. The Dionex Ion Chromatography System was purchased and set up for routine chloride analysis. Additional design, construction, and installation work was performed on the induction furnace. UZ-1 and UZN-43 samples were leached and analyzed to establish the depth profile of leachable chloride. In addition, work with the shatterbox was begun in order to look at the effects of particle size on leachable chloride.

Letter report, July 31, 1989: This letter report summarized work performed during the first ten months of fiscal year 1989. Information included in this report is also included in the following formal report.

Formal report, April 2, 1990: This formal report summarized work performed during fiscal year 1989. Developmental work included devising a technique to analyze chloride and bromide simultaneously using ion chromatography and creating the step-leaching procedure to

identify the rock component of leachable halides. Analytical work included determining chloride and bromide profiles for the UZ-1 borehole, estimating the Cl/Br ratio for both meteoric and rock end members, estimating the cosmogenic $^{36}\text{Cl}/\text{Cl}$ ratio, correcting chloride content to reflect only the meteoric input, evaluating the significance of grain-size on leachable halides, and looking at chloride, bromide, and chlorine-36 distributions in the U12G Tunnel and Trench 14.

Letter report, October 9, 1990: This letter summarized work performed during fiscal year 1990. The chlorine-36 sample preparation procedure was modified to improve yields. Modifications include use of a centrifuge to separate the AgCl precipitate from the aqueous phase and acidification to reprecipitate the AgCl. Borehole samples were collected from Apache Leap, although the increased moisture in the profile makes their use questionable. A complete sample inventory was conducted, including samples from both Yucca Mountain and Apache Leap. A preliminary draft, quality assurance document was prepared outlining sample collection, technical, and analytical procedures.

Letter report, January 15, 1991: This letter accompanied the invoice for October through December of 1990. Several UZ-1 borehole samples were analyzed for stable chlorine isotopes with results showing a good correlation with the Cl/Br ratio. Initial developmental work with the column leaching set-up indicated it to be a favorable approach for large-volume leaching although additional work is suggested.

Prior to 1988, work on the project was sporadic and was focused primarily on defining analytical requirements and obtaining the necessary equipment. For these reasons, sufficient data were not obtained during that period of time to warrant a formal report. Selected letters that were written throughout the contract to accompany monthly invoices have been included in Appendix A to the extent that they provide a brief summary of ongoing work. One publication has been produced based on the work performed under this contract. That paper, with Edward Norris as principal author, is included in Appendix B of this document.

A complete sample inventory, listing samples from both Yucca Mountain and Apache Leap, has been included in Appendix C.

APPENDIX A:

Formal Reports, Letter Reports, and Letters Accompanying Invoices

**CHARACTERIZATION OF HISTORICAL INFILTRATION
IN THE UNSATURATED ZONE AT THE NEVADA TEST SITE
USING CHLORIDE, BROMIDE, AND CHLORINE-36
AS ENVIRONMENTAL TRACERS**

Prepared for:

Los Alamos National Laboratory
Los Alamos, New Mexico
Contract No. 9-XDD-6329F-1

Prepared by:

Hydro Geo Chem, Inc.
1430 N. Sixth Avenue
Tucson, Arizona 85705

April 2, 1990

TABLE OF CONTENTS

INTRODUCTION	1
BACKGROUND	5
Chloride as an Environmental Tracer	5
Chlorine-36 as an Environmental Tracer	6
Use of Bromide to Determine Meteoric and Rock End Members	8
EXPERIMENTAL PROCEDURES	10
Step-wise Leaching Experiment	10
Effects of Sieving	11
Shatterbox Experiment	11
Analytical Procedures	12
ANALYTICAL RESULTS	13
Halide Analyses of Sieved Samples	13
Estimation of Meteoric Halide Ratio	13
Step-wise Leaching Experiment	18
Shatterbox Experiment	18
Correction for Meteoric Chloride	23
Chlorine-36 Analyses	23
Step-wise Leaching for Chlorine-36	25
Chlorine-36 in the U12G Tunnel	26
Trench 14 Samples	30
DISCUSSION OF RESULTS	31
Estimation of End Members	31
Use of Chlorine-36 to Determine Extent of Percolation	32
SUGGESTIONS FOR FUTURE WORK	36
Estimation of End Members	36
Use of Chlorine-36 to Determine Extent of Percolation	38
CONCLUSION	39
REFERENCES	41
APPENDIX A: PROCESSING OF ROCK SAMPLES FOR CHLORIDE, BROMIDE, AND CHLORINE-36 ANALYSES	
APPENDIX B: ANALYTICAL DERIVATIONS	

INTRODUCTION

This document is an annual report, prepared by Hydro Geo Chem for Los Alamos National Laboratory under contract number 9-XDD-6329F-1. The ultimate goal of this work is to characterize historical infiltration and unsaturated flow in the Yucca Mountain area of the Nevada Test Site. Figure 1 shows the location of Yucca Mountain at the Nevada Test Site. Current work focuses on the use of chloride, bromide, and chlorine-36 distributions to evaluate the depth to which percolation of these conservative solutes, presumably effective environmental tracers of infiltrating water, has occurred in the unsaturated zone.

Yucca Mountain is currently under consideration as a permanent repository for disposal of nuclear wastes. The area is underlain by a thick sequence of ash-flow and bedded tuffs, ranging in thickness from 1800 to 3050 meters. Tuff is a complex rock type that can potentially show substantial variation in chemical composition, mineralogy, and texture. Samples collected from an exploratory borehole at Yucca Mountain, however, showed only limited variations in chemical composition (Waters and Carroll, 1981).

The proposed repository would be located at a depth of about 1000 feet below land surface in the Topopah Spring Member of the Paintbrush Tuff. The Topopah Spring Member, approximately 1200 feet thick, is subdivided into 24 lithologic intervals. It ranges from slightly welded at its base to densely welded in the interior, and then to nonwelded at the top of the Member (Waters and Carroll, 1981). Depth to water is approximately 1600 feet in the vicinity of Yucca Mountain (Norris, 1989).

Figure 1. Location Map showing Yucca Mountain at the Nevada Test Site

Samples for the present analyses were collected from the UZN-43 borehole, in the interval from 6 to 41 feet below land surface, and from the UZ-1 borehole, from a depth of 97 feet to 1270 feet. Samples were also collected from various locations within the U12G tunnel. The tunnel is a horizontal shaft in the side of Rainier Mesa, with approximately 1300 feet of overburden. Additional samples were collected from Trench 14 to help identify the origin of the carbonate veins. Figure 2, a map of the Nevada Test Site, shows the location of the sampling sites.

All of the analytical work presented in this report was done prior to October, 1989. Some repeat chloride and bromide analyses were performed for UZ-1 and UZN-43 samples during August. However, these results are not included in this report because they did not meet our QA guidelines.

BACKGROUND

Chloride as an Environmental Tracer

The complexities and uncertainties inherent in measuring water and solute transport in unsaturated, fractured rock are significant (Montazer and Wilson, 1984). Some of these problems can be side-stepped by using the distribution of environmental tracers to reconstruct patterns of infiltration and solute movement.

Chlorine is the ideal environmental tracer for this purpose because of its simple geochemistry. In nature, it occurs solely as the chloride ion, due to its high oxidation potential. Solubility controls are absent in fresh waters and vapor phase transport does not occur within normal temperature ranges. In addition, chloride is one of the least likely of all ions to sorb on natural surfaces, because of its negative charge and small ionic radius. Consequently, infiltrating water effectively leaches chloride from soils and other subsurface materials. Shallow groundwater systems therefore tend to be relatively dilute with respect to chloride. In deeper groundwater systems, chloride can be enriched due to such subsurface sources as fluid inclusions, microfractures, minerals containing chloride, and the admixture of water from saline aquifers.

Atmospheric chloride originates predominantly as sea salt. Circulating weather patterns redistribute atmospheric chloride so that its concentration decreases exponentially with distance from the ocean. Atmospheric chloride enters the lithosphere through precipitation and dry fallout after a mean

residence time in the atmosphere of about one week (Junge and Gustafson, 1957).

The movement of chloride in the unsaturated zone is a function of antecedent moisture conditions, matrix characteristics, average water velocity, and the existence of preferential flow paths. Numerous studies, both at the laboratory and field scale, have shown that chloride moves either at the same rate as the average water velocity, or faster, due to the phenomenon of ion exclusion. A list of these studies is included in the 1985 Hydro Geo Chem annual report to Los Alamos. None of these studies examined the fate of chloride during vapor phase transport of water.

The distribution of chloride in the subsurface can provide insight into the movement of moisture and solutes in the unsaturated zone. For example, the distribution of chloride both in and in the vicinity of a fracture can be used to determine whether chloride or water have moved into the bulk rock matrix and whether chloride is being deposited as water movement changes from liquid to vapor phase.

Chlorine-36 as an Environmental Tracer

Chlorine-36 (^{36}Cl) is a radioactive isotope with a half-life of approximately 301,000 years. It is produced in the atmosphere by both spallation and neutron activation, and it enters the lithosphere in much the same way as stable chloride. Distribution is a function of both latitude and distance from coastal areas (Bentley et al., 1985).

Subsurface production of ^{36}Cl is by neutron activation of ^{35}Cl . The major source of neutrons at depth is the α -n reactions due to the decay of uranium and thorium series elements. With time, a secular equilibrium will be established between the decay of ^{36}Cl and its subsurface production. The secular equilibrium $^{36}\text{Cl}/\text{Cl}$ ratio for most rocks is on the order of 10^{-14} , varying with altitude and the chemical composition of the rock (Bentley et al., 1985).

Various authors (Bentley et al., 1982, 1985; Phillips et al., 1984; Hydro Geo Chem, 1985) have studied the feasibility of using ^{36}Cl to study infiltration and fluid transport in the unsaturated zone. These studies have suggested that the movement of ^{36}Cl in the subsurface corresponds to that of stable chloride.

Atmospheric testing of nuclear weapons in the late 1950's and early 1960's released a pulse of ^{36}Cl that is approximately three orders of magnitude higher than pre-bomb meteoric concentrations. Measurements taken from two trench samples earlier in this project suggested a pre-bomb, meteoric-origin $^{36}\text{Cl}/\text{Cl}$ ratio of 500×10^{-15} to 600×10^{-15} (Gifford, 1987; Norris et al., 1987). This ratio is consistent with ratios predicted by Bentley et al. (1985). The bomb pulse, measured in terms of ^{36}Cl concentrations and $^{36}\text{Cl}/\text{Cl}$ ratios, shows a clearly defined downward movement of ^{36}Cl such that the depth to which meteoric water has infiltrated within the past thirty years can be identified.

Both the ratio of ^{36}Cl to stable chloride and ^{36}Cl concentration are valuable parameters in terms of hydrologic tracing. ^{36}Cl concentrations will increase due to evaporation, transpiration or losses of water from the system. The effects of rainfall distribution and variable infiltration on the

concentrations of both chloride and ^{36}Cl in the subsurface can also be significant. On the other hand, since there is no measured fractionation of ^{36}Cl in the subsurface, the isotopic ratio should remain constant, despite these influences. Both dilution of meteoric chloride by rock-origin chloride and ^{36}Cl decay will reduce the isotopic ratio (Bentley et al., 1985).

In order to accurately infer infiltration and percolation history from chloride and ^{36}Cl distributions, it is necessary to identify both the meteoric and the rock component of chloride in the subsurface.

Use of Bromide to Determine Meteoric and Rock End Members

Bromine is another excellent candidate for an environmental tracer. A minor element in both the lithosphere and hydrosphere, its geochemical behavior is very similar to that of chlorine. Bromine is biophilic in nature so that it tends to be enriched in organic-rich soils and sediments. However, bioconcentration of bromine is not expected to be significant in arid areas such as Yucca Mountain. Bromine exists almost exclusively as the bromide ion in natural waters, although traces can be found in dissolved organic compounds. The chloride to bromide ratio (Cl/Br) in salts of meteoric origin is reputed to be near constant at 120 to 130, as determined for the arid regions of southern Arizona (Davis, 1989). Various other authors have suggested a range of Cl/Br ratios from 14 to 330 (Whittemore, 1988).

Under the assumption of a constant Cl/Br ratio in rainfall, this ratio can be used as a chemical signature with which to identify different waters and their

sources. In the subsurface, the Cl/Br ratio of fresh, meteoric water reflects the relationship of these halides in local precipitation. The Cl/Br ratio of the rock matrix may be distinctly different. Thus, bromide concentrations can theoretically be used in conjunction with chloride to identify water that is solely meteoric in origin versus pure rock leachate. With identification of end members, mass balance estimates can be used to determine the meteoric and rock component of leachable halides in samples with an intermediate Cl/Br ratio.

Ultimately, analysis of bromide concentrations in the unsaturated zone should aid in the quantification of subsurface chloride that is originating from the rock. This information will then allow corrections to be made in $^{36}\text{Cl}/\text{Cl}$ data so that the percolation history can be more accurately reconstructed.

EXPERIMENTAL PROCEDURES

If the meteoric input of chloride is significant, leaching a sample of undisturbed rock should yield primarily meteoric halides. However, the drilling and sampling process crushes the rock to some degree, breaking open crystals, inclusions, and micro fractures within the rock. When this sample is subsequently leached, its halide composition reflects the input of meteoric salts superimposed on some partial component of rock halide that is released due to the degree of pulverization. Each sample could therefore potentially represent a unique mixture of meteoric and rock component halides. Since the meteoric component is of primary interest, it would be very useful to identify the halide composition of the rock component. With this information, leachable chloride values can be corrected to reflect only the meteoric component. Once these two end members are defined, mass balance calculations can be used to determine the $^{36}\text{Cl}/\text{Cl}$ ratio in the meteoric component. Depth profiles can then be constructed for both meteoric halides and meteoric $^{36}\text{Cl}/\text{Cl}$.

Step-wise Leaching Experiment

Repeated crushing and leaching of a given sample should progressively increase the rock component of the leachable halides. The step-wise leaching experiment was designed by Cheng and Norris, specifically to characterize the halide ratio of the rock end member. The procedure involves leaching, washing, drying, and shatterbox crushing of rock samples. This sequence is repeated several times, with chloride, bromide, and ^{36}Cl concentrations being measured at each interval.

Effects of Sieving

Previous data indicate that the halide concentration of rock samples may vary significantly from one sample to another, possibly due in part to the effect of variable size fractions (Cheng, 1989). To reduce this effect on halide distribution, the samples were sieved into several size fractions prior to leaching. The sieve sizes used were 35, 60, and 230 mesh. A depth profile of halide distribution constructed from sieved samples should provide insight into the chloride depositional processes and, possibly, a more accurate interpretation of percolation in the past.

Shatterbox Experiment

The shatterbox experiment was designed to further explore the relationship between grain size and leachable halides. A given sample is crushed in the shatterbox for a period of time. An aliquot is removed and the remaining sample is crushed for an additional period of time. This process is repeated a number of times, resulting in several sub-samples, each of which is crushed for a progressively longer period of time than the previous sub-sample.

This experiment can also be used to estimate the halide rock component. Longer crushing time equates with increased leachable halides so that the slope of the best-fit line through the data points which represent sample Cl/Br ratios, approaches the Cl/Br ratio of the rock end member. The advantage of this approach over the step-wise leaching experiment is that it is far more time efficient.

Analytical Procedures

Chloride and bromide concentrations are determined by Hydro Geo Chem using ion chromatography. Chloride and bromide are detected by conductivity; bromide is also detected optically to enhance detection. Analysis for ^{36}Cl is by tandem accelerator mass spectrometry (TAMS), according to procedures described by Elmore et al. (1979). All TAMS ^{36}Cl measurements are performed at the Nuclear Structure Research Laboratory at the University of Rochester. Analytical procedures are outlined in Appendix A.

ANALYTICAL RESULTS

Halide Analyses of Sieved Samples

Samples from UZ-1 and UZN-43 were sieved and analyzed for chloride and bromide. The analytical results are presented in Table 1. Vertical profiles for chloride and bromide in UZ-1 are shown in Figures 3 and 4, respectively. Repeat analyses of the two shallowest UZN-43 samples indicated that the initial Cl/Br ratios may have been low due to error in procedure or analyses. Because the UZN-43 samples were collected relatively close to the ground surface, they probably contain a greater meteoric component than do the UZ-1 samples.

As discussed earlier, leachable halides were expected to increase with decreasing grain size, and sieving the samples would make this phenomenon apparent. However, the data do not support this theory. In some cases, leachable halides increase in the finer grained fraction; in other cases they decrease, or show variable behavior.

Estimation of Meteoric Halide Ratio

Data from the UZ-1 and UZN-43 sieve analyses were used to estimate the halide ratio of the meteoric end member. Cl/Br ratios were plotted versus chloride concentrations for UZ-1, producing a "semi-normal" distribution. This plot is shown in Figure 5. The mode, falling at approximately 130, is viewed as the most probable value for the meteoric end member. A Cl/Br ratio of 130 is in

TABLE 1. CHLORIDE, BROMIDE, AND CHLORIDE/BROMIDE RATIOS, UZ-1 AND UZ-43 SAMPLES

SAMPLE	DEPTH (FT)		CHLORIDE, PPM IN ROCK				BROMIDE, PPM IN ROCK				CHLORIDE/BROMIDE RATIO			
	FROM	TO	A	B	C	D	A	B	C	D	A	B	C	D
UZ-43	6	7	69.7	155.	134.	143.	2.78	13.5	13.7	17.5	25	11	10	8
	10	11	58.6	157.	15.8	163.	5.58	2.79	14.8	7.29	11	56	1	22
	15	16	45.9	87.8	89.1	76.5	0.145	0.337	0.329	0.312	316	260	270	245
	20	21	3.18	6.91	5.87	11.7	0.022	0.034	0.037	0.060	143	205	158	196
	25	26	1.45	3.81	5.08	10.7	0.012	0.025	0.027	0.052	122	150	189	205
	30	31	7.23	5.35	5.98	7.10	0.049	0.015	0.013	0.012	148	349	474	582
	35	36	---	7.89	6.66	7.68	---	0.011	0.016	0.006	---	745	411	1400
	40	41	1.01	1.36	1.47	2.72	0.005	0.003	0.007	0.005	190	430	223	525
UZ-43 REPEATS														
	6	7	99.4	289.	299.	377.	0.460	1.43	1.48	1.94	216	203	201	194
	10	11	11.8	---	34.8	---	0.051	---	0.153	---	231	---	228	---
	35	36	9.03	---	---	---	0.025	---	---	---	369	---	---	---
SAMPLE	DEPTH (FT)		CHLORIDE, PPM IN ROCK				BROMIDE, PPM IN ROCK				CHLORIDE/BROMIDE RATIO			
	FROM	TO	A	B	C	D	A	B	C	D	A	B	C	D
UZ-1	97	101	28.3	37.7	44.7	55.4	0.167	0.163	0.159	0.274	170	232	282	202
	250	255	22.58	25.0	25.7	30.5	0.182	0.168	0.184	0.241	124	149	139	126
	295	300	1.82	2.01	3.23	3.74	0.012	0.015	0.014	0.023	147	138	224	161
	347	350	7.64	4.22	3.53	3.57	0.052	0.034	0.023	0.025	148	124	152	143
	495	500	5.23	5.37	5.11	6.91	0.025	0.019	0.025	0.021	210	279	208	337
	500	502	8.67	10.0	10.3	7.30	0.017	0.024	0.023	0.037	499	412	453	196
	595	599	7.97	7.40	7.94	9.15	0.054	0.048	0.057	0.058	147	153	138	157
	715	720	0.96	0.95	1.20	1.28	0.010	0.014	0.011	0.009	94	71	112	142
	830	835	1.96	2.18	1.86	2.40	0.007	0.009	0.006	0.010	271	244	327	238
	845	850	4.96	2.32	1.82	2.01	0.014	0.013	0.014	0.012	352	181	132	162
	920	925	2.12	2.22	2.39	2.68	0.025	0.022	0.020	0.021	84	100	121	129
	945	950	2.85	2.52	2.04	3.18	0.020	0.010	0.011	0.035	146	249	181	90
	970	975	3.45	2.61	3.33	3.86	0.019	0.019	0.012	0.012	178	141	269	321
	1020	1025	1.15	2.28	2.29	2.94	0.009	0.015	0.015	0.023	126	157	155	129
	1047	1050	2.32	4.33	2.52	3.34	0.010	0.014	0.011	0.014	236	320	231	238
	1070	1075	2.31	1.72	2.31	3.39	0.010	0.013	0.012	0.011	228	131	191	311
	1147	1150	4.46	5.15	4.17	6.66	0.022	0.015	0.012	0.021	198	344	340	322
	1170	1175	3.76	3.68	2.88	6.66	0.014	0.015	0.016	0.015	259	245	186	450
	1195	1200	2.59	2.87	2.27	2.97	0.017	0.017	0.016	0.024	150	171	138	126
	1220	1225	3.18	3.88	3.70	5.43	0.017	0.013	0.016	0.046	191	304	237	118
	1245	1250	4.17	3.90	3.76	3.65	0.018	0.021	0.015	0.012	228	190	248	302
	1258	1260	6.88	10.5	11.7	13.8	0.043	0.076	0.067	0.087	160	138	174	158
	1267	1268	1.95	3.81	4.43	13.3	0.008	0.015	0.014	0.027	248	258	310	483
	1268	1270	2.94	3.82	3.99	4.93	0.015	0.015	0.024	0.031	197	252	168	160

A = >35 B = 35-60 C = 60-230 D = <230

FIGURE 3. VERTICAL CHLORIDE PROFILE FOR UZ-1 SAMPLES

SIEVE SIZE

..... > 35

----- 35-60

***** 60-230

◇◇◇◇◇ < 230

FIGURE 4. VERTICAL BROMIDE PROFILE FOR UZ-1 SAMPLES

SIEVE SIZE

XXXXX > 35

XXXXX 35-60

***** 60-230

◇◇◇◇◇ < 230

FIGURE 5. CHLORIDE/BROMIDE RATIO VERSUS CHLORIDE CONCENTRATION
FOR UZ-1 SAMPLES

SIEVE SIZE

▲▲▲▲▲ > 35

△△△△△ 35-60

***** 60-230

◇◇◇◇◇ < 230

excellent agreement with Cl/Br ratios reported in the literature, particularly determined for precipitation in the Tucson Basin (Davis, 1989). Figures 6(a) and 6(b) plot the chloride versus bromide concentrations for the UZ-1 samples. Also included on the plots are lines of Cl/Br ratio equal to 130:1 for the meteoric end member, and 500:1 for the rock end member, which will be discussed next.

Step-wise Leaching Experiment

Several samples from UZ-1 have been analyzed according to the procedures for the step-wise leaching experiment. Results are presented in Table 2.

Sample 250-255 has been analyzed most extensively. The results from two separate experiments are very similar. In both cases, the final value for Cl/Br ratio is approximately 490. This ratio may represent the rock end member. If so, then the pattern of leaching suggests a mixing of halide sources up to and including step D of the experiment.

Shatterbox Experiment

Three UZ-1 samples have been analyzed in the shatterbox experiment (97-101, 250-255, 295-300). These results are shown in Table 3.

Figure 7 is a plot of chloride versus bromide for the three UZ-1 samples. The slopes of the three lines represent the ratio of leachable halides in the samples. The Cl/Br ratio for sample UZ-1 295-300 is approximately 470, which means it may represent the rock end member. The other two samples have halide

FIGURE 6a. BROMIDE VERSUS CHLORIDE FOR UZ-1 SAMPLES
(LOW CONCENTRATIONS ONLY)

SIEVE SIZE

***** > 35

◇◇◇◇◇ 35-60

***** 60-230

◇◇◇◇◇ < 230

FIGURE 6b. BROMIDE VERSUS CHLORIDE FOR UZ-1 SAMPLES
(ALL DATA POINTS)

SIEVE SIZE

○ ○ ○ ○ ○ > 35

□ □ □ □ □ 35-60

***** 60-230

◇ ◇ ◇ ◇ ◇ < 230

FIGURE 7. SHATTERBOX EXPERIMENTS FOR UZ-1 SAMPLES

○ ○ ○ ○ UZ-1 97-101

☆ ☆ ☆ ☆ UZ-1 250-255

◇ ◇ ◇ ◇ UZ-1 295-300

TABLE 2. RESULTS OF STEP-WISE LEACHING EXPERIMENT, UZ-1 SAMPLES

SAMPLE DEPTH (FT)			PPM IN SOLUTION		WATER	SAMPLE	PPM IN ROCK			CL-36/CL
FROM	TO	EXT.**	CL	BR	VOLUME	WEIGHT	CL	BR	CL/BR	1E-15
					(ML)	(GM)				
500	502	(3)	---	---	--	---	---	---	---	1885
1220	1225	(3)	0.997	0.003	50	25.42	1.96	0.006	322	---
1195	1200	(3)	1.04	0.003	50	25.37	2.06	0.005	418	340
1020	1025	(3)	0.935	0.001	50	25.09	1.86	0.002	850	159
250	255	2 MIN.	8.56	0.032	20	10.21	16.8	0.062	270	---
		B(STEP)	2.46	*	20	10.28	4.79	---	---	91
		C(STEP)	9.08	*	20	10.11	18.0	---	---	---
		D(STEP)	2.44	0.009	20	10.02	4.86	0.018	271	36
		E(STEP)	4.65	0.010	20	10.02	9.28	0.019	489	25
250	255	A(STEP)	10.9	0.041	50	10.01	54.4	0.202	269	193
		B(STEP)	0.740	*	50	10.03	3.69	---	---	91
		C(STEP)	2.38	*	50	10.03	11.9	---	---	---
		D(STEP)	1.20	0.005	50	10.04	5.98	0.027	222	36
		E(STEP)	1.90	0.004	50	10.12	9.39	0.019	487	25

* : CONTAMINATED BY HIGH NO₃⁻ , COULD NOT BE ANALYZED

** : SAMPLE LEACHING/CRUSHING STEPS ARE DESCRIBED BELOW:

- (3) : SAMPLE WAS LEACHED, DRIED, CRUSHED FOR 3 MINUTES, AND LEACHED FOR HALIDE ANALYSIS
- 2 MIN. : ORIGINAL SAMPLE WAS CRUSHED FOR 2 MINUTES BEFORE LEACHING.
- A(STEP): ORIGINAL SAMPLE WAS LEACHED, WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.
- B(STEP): SAMPLE A(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.
- C(STEP): SAMPLE A(STEP) WAS WASHED, DRIED, CRUSHED FOR 10 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.
- D(STEP): SAMPLE B(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.
- E(STEP): SAMPLE D(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.

ratios between 245 and 270, indicating a composite source of halides for these samples, assuming the rock Cl/Br ratio is about 500.

Sample 97-101 is anomalous because its concentration of leachable halides decreased with increased crushing. This may be due to its extremely high moisture content and the fact that the sample formed a dense, clayey cake upon crushing in the shatterbox, presumably rendering it less leachable.

TABLE 3. RESULTS OF SHATTERBOX EXPERIMENT, UZ-1 SAMPLES

SAMPLE DEPTH (FT)		TIME (MINUTES)	PPM IN SOLUTION		WATER VOLUME (ML)	SAMPLE WEIGHT (GM)	PPM IN ROCK		
FROM	TO		CL	BR			CL	BR	CL/BR
250	255	0	8.16	0.045	50	10.89	37.5	0.204	184
		2	9.85	0.053	50	10.01	49.2	0.263	187
		5	13.7	0.064	50	10.02	68.4	0.321	213
		8	14.6	0.067	50	10.03	72.8	0.332	220
		11	17.0	0.080	50	10.05	84.8	0.397	214
		15	23.4	---	50	10.05	117.	---	---
		20	34.2	---	50	10.04	170.	---	---
		25	---	---	50	10.01	---	---	---
		25	---	---	50	10.01	---	---	---
97	101	0	12.6	0.046	50	10.07	62.7	0.227	277
		2	10.8	0.043	50	10.08	53.7	0.211	255
		5	11.0	0.040	50	10.11	54.5	0.197	277
		8	10.7	0.038	50	10.01	53.5	0.189	283
		11	10.7	0.038	50	10.01	53.5	0.188	285
		15	10.1	0.033	50	10.02	50.6	0.162	312
		20	10.1	---	50	10.10	49.9	---	---
		25	9.16	0.032	50	10.08	45.4	0.160	284
		25	9.16	0.032	50	10.08	45.4	0.160	284
295	300	0	0.274	0.003	50	10.40	1.32	0.015	88
		2	0.451	0.004	50	10.01	2.25	0.018	125
		5	0.741	0.004	50	10.13	3.66	0.021	176
		8	1.24	0.005	50	10.01	6.18	0.027	234
		11	1.77	0.007	50	10.01	8.82	0.034	263
		15	2.53	0.009	50	10.04	12.6	0.045	281
		20	---	---	50	10.04	---	---	---
		25	3.95	0.011	50	10.01	19.7	0.054	362
		25	3.95	0.011	50	10.01	19.7	0.054	362

Correction for Meteoric Chloride

Meteoric and rock end member Cl/Br ratios can be used to determine the component of meteoric chloride in each sample. The derivation of this calculation is given in Appendix B. Table 4 lists the weighted averages of chloride and bromide concentrations for the sieved samples presented in Table 1, in addition to the calculated concentration of meteoric chloride.

Chlorine-36 Analyses

Seven UZ-1 and three UZN-43 samples were analyzed for ^{36}Cl , in addition to a blank. The analytical results, shown in Table 5, indicate that UZ-1 samples 97-101 and 500-502 may contain bomb-pulse ^{36}Cl .

TABLE 4. METEORIC CHLORIDE COMPONENT, UZ-1 AND UZN-43 SAMPLES

SAMPLE	DEPTH (FT)		CHLORIDE	BROMIDE	CL/BR RATIO	METEORIC CHLORIDE	PERCENT METEORIC
	FROM	TO	PPM IN ROCK	PPM IN ROCK		COMPONENT	COMPONENT
UZN-43	6	7	111.	9.55	12	---	---
	10	11	81.1	8.10	10	---	---
	15	16	74.9	0.281	266	23.1	30.8
	20	21	4.38	0.025	175	2.86	65.3
	25	26	5.25	0.029	181	3.25	61.9
	30	31	6.42	0.022	290	1.64	25.5
	35	36	7.34	0.011	668	---	---
	40	41	1.47	0.005	295	0.36	24.4
SAMPLE	DEPTH (FT)		CHLORIDE	BROMIDE	CL/BR RATIO	METEORIC CHLORIDE	PERCENT METEORIC
	FROM	TO	PPM IN ROCK	PPM IN ROCK		COMPONENT	COMPONENT
UZ-1	97	101	38.8	0.071	223	16.9	43.5
	250	255	24.9	0.184	136	23.5	94.4
	295	300	2.70	0.016	169	2.32	85.9
	347	350	4.76	0.034	142	4.24	89.1
	495	500	5.59	0.023	248	2.00	35.8
	500	502	9.43	0.021	440	0.46	4.9
	595	599	8.12	0.054	150	6.68	82.3
	715	720	1.07	0.011	94	---	---
	830	835	2.10	0.008	262	0.67	31.8
	845	850	2.82	0.013	212	1.35	47.8
	920	925	2.34	0.022	106	---	---
	945	950	2.65	0.019	140	2.41	90.8
	970	975	3.31	0.016	213	1.16	47.2
	1020	1025	1.97	0.014	145	1.70	86.1
	1047	1050	3.23	0.012	260	1.05	32.4
	1070	1075	2.45	0.011	213	1.16	47.2
	1147	1150	4.84	0.017	287	1.27	26.2
	1170	1175	4.24	0.015	283	1.14	26.9
	1195	1200	2.70	0.017	157	2.08	77.0
	1220	1225	3.97	0.022	183	2.43	61.1
	1245	1250	3.87	0.016	235	1.54	39.8
	1258	1260	9.95	0.064	157	7.67	77.0
	1267	1268	5.05	0.015	341	0.90	17.8
	1268	1270	3.80	0.020	189	2.20	57.8

TABLE 5. CHLORINE-36 RESULTS, UZ-1 AND UZN-43 SAMPLES
(REPORTED AS ATOMIC RATIO OF CL-36/TOTAL CHLORIDE)

SAMPLE	DEPTH (FT)		CL-36/TOTAL CL	
	FROM	TO	1E-15	+/-
UZ-1	97	101	11400	360
	170	175 *	2498	198
	250	255	436	25
	395	400 *	390	48
	495	500	403	42
	500	502	2046	103
	1020	1025	245	38
	1195	1200	454	61
	1220	1225	102	11
UZN-43	6	7	759	59
	10	11	526	38
	15	16	470	25

* these values were determined in 1985

Step-wise Leaching for Chlorine-36

The UZ-1 250-255 sample has been processed according to the step-wise leaching procedures and analyzed for ^{36}Cl . Results are given in Table 6.

The sample representing the E step in Table 6 contains a very low $^{36}\text{Cl}/\text{Cl}$ ratio (25×10^{-15}) and a high Cl/Br ratio (487). These data are consistent with representing a rock end member. As mentioned earlier, the local pre-bomb $^{36}\text{Cl}/\text{Cl}$ ratio is about 500×10^{-15} . Based on this estimate and the results from sample 250-255 E step, the $^{36}\text{Cl}/\text{Cl}$ ratio of meteoric and rock end members is tentatively set at 500×10^{-15} and 25×10^{-15} , respectively. The estimated Cl/Br ratios of the end members is 130 for the meteoric component and 500 for the rock component.

The analytical results for sample 250-255 are plotted in Figure 8, along with the theoretical mixing curve. The Cl/Br ratio for both the D and E step are average values, since the entire step-wise leaching experiment was run twice.

TABLE 6. CHLORINE-36 RESULTS OF STEP-WISE LEACHING EXPERIMENT, UZ-1 SAMPLES
(REPORTED AS ATOMIC RATIO OF CL-36/TOTAL CHLORIDE)

SAMPLE	DEPTH (FT)		EXT**	CL-36/TOTAL CL	
	FROM	TO		1E-12	+/-
UZ-1	500	502	(3)	1885	150
	1020	1025	(3)	159	12
	1195	1200	(3)	340	15
UZ-1	250	255	A(STEP)	193	7
	250	255	B(STEP)	91	4
	250	255	D(STEP)	36	11
	250	255	E(STEP)	25	5

** : SAMPLE LEACHING/CRUSHING STEPS ARE DESCRIBED BELOW:

- (3) : ORIGINAL SAMPLE WAS LEACHED, DRIED, CRUSHED FOR 3 MINUTES, AND LEACHED FOR HALIDE ANALYSIS
- A(STEP) : ORIGINAL SAMPLE WAS LEACHED, WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.
- B(STEP) : SAMPLE A(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.
- D(STEP) : SAMPLE B(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.
- E(STEP) : SAMPLE D(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.

With the exception of the D step sample, the data follow the theoretical curve.

Three additional UZ-1 samples (500-502, 1020-1025, 1195-1200) were analyzed for ^{36}Cl after a 3 minute crushing and leaching step. The $^{36}\text{Cl}/\text{Cl}$ ratios of these samples decreased in value as a result of the additional crushing and leaching. The Cl/Br ratio of these same samples increased, although this trend should be viewed qualitatively rather than quantitatively, in light of the potential analytical error associated with measuring low bromide concentrations.

Chlorine-36 in the U12G Tunnel

Two samples from the U12G tunnel that were analyzed early in the year, showed both high and low $^{36}\text{Cl}/\text{Cl}$ ratios. In order to increase the data base and

FIGURE 8. STEP-WISE LEACHING EXPERIMENTS FOR UZ-1 SAMPLE 250-255

address the uncertainty as to the source of the elevated ^{36}Cl content, six additional samples were collected from various locations in the tunnel (Norris, 1989) and analyzed for ^{36}Cl . The analytical results are included in Table 7. Figure 9 shows the location of these samples.

TABLE 7. CHLORINE-36 RESULTS, U12G TUNNEL
(REPORTED AS ATOMIC RATIO OF $\text{Cl-36/TOTAL CHLORIDE}$)

SAMPLE	CL-36/TOTAL CL	
	1E-15	+/-
AC-1 RUNS 16 & 17	412	18
AC-2 RUNS 19 & 20	306	22
AC-2 RUNS 38-40	845	76
DH-1 RUNS 5 & 6	1539	101
DH-1 RUNS 7-9	1964	75
DH-2 RUNS 7 & 8	1709	70
NBS SRM#919 NaCl	1	5
AC-1 RUN 1	1243	87*
AC-1 RUNS 2-3	3044	360*
U12G-TN-D1 (DUST SAMPLE)	2045	148
NBS SRM#919 NaCl	46	6

*THESE SAMPLES CONTAINED HIGH SULFUR CONTENT AND
COULD REPRESENT LOWER LIMITS ON THE $^{36}\text{Cl}/\text{Cl}$ RATIOS

One possible source of the elevated $^{36}\text{Cl}/\text{Cl}$ ratios is contamination of the rock samples by dust occurring in the tunnel (Fabryka-Martin, 1989). To address this possibility, three dust samples were collected from the tunnel for analysis of ^{36}Cl .

Results on the ^{36}Cl analyses for the dust samples indicated that there was insufficient sample volume for analysis in all but one of the samples, despite addition of a carrier. The one sample that was analyzed was collected from the Experiment Drift and showed an elevated $^{36}\text{Cl}/\text{Cl}$ ratio of 2045×10^{-15} . Results are given in Table 7.

Figure 9. ³⁶Cl/Cl Ratios in the U12G Tunnel, Plan View

Trench 14 Samples

Ten samples were collected from Trench 14 and analyzed for chloride and bromide. The purpose behind this investigation is to determine the origin of carbonate veins in the trench. A high Cl/Br ratio should indicate a hydrothermal origin, whereas a low Cl/Br ratio would suggest an eolian origin. Table 8 shows the results of these analyses.

TABLE 8. CHLORIDE, BROMIDE, AND CHLORIDE/BROMIDE RATIO, TRENCH 14 SAMPLE

SAMPLE	PPM IN SOLUTION		WATER VOLUME (ML)	SAMPLE WEIGHT (GM)	PPM IN ROCK		
	CL	BR			CL	BR	CL/BR
4-1-1	0.330	0.005	50	20.32	0.812	0.013	61
4-1-2	2.80	0.016	50	20.12	6.96	0.040	176
4-1-3	16.6	0.030	50	20.12	41.2	0.076	545
4-1-4B	4.38	0.043	50	20.16	10.9	0.107	102
4-1-4K	0.626	0.044	50	20.33	1.54	0.108	14
4-1-5	4.44	0.080	50	20.01	11.1	0.199	56
4-1-6	1.20	0.074	50	20.01	2.99	0.185	16
4-1-7	442.	3.52	50	20.35	1090.	8.66	125
4-1-8	69.0	0.741	50	20.12	171.	1.84	93
4-1-9	7.00	0.092	50	20.40	17.2	0.225	76

The Cl/Br ratios of the Trench 14 samples vary greatly, from approximately 15 to greater than 500. This variation makes it difficult to pinpoint an origin for the carbonate based on high or low Cl/Br ratios. The potential chloride and bromide contributors to this carbonate deposit include meteoric origin salts, shallow subsurface origin seeps, and deep subsurface origin waters. Additional work is suggested to evaluate or infer the Cl/Br and $^{36}\text{Cl}/\text{Cl}$ ratios of the possible sources.

DISCUSSION OF RESULTS

Estimation of End Members

The Cl/Br ratio appears to be a valuable tool in the identification of meteoric and rock components of leachable halides in the unsaturated zone. This end member identification is important because it allows for corrections to be made in the ^{36}Cl data in order to derive only meteoric input.

The procedure for end member identification is different for each end member. The rock end member is effectively isolated through progressive crushing and leaching, both in the step-wise leaching and shatterbox experiments. With sufficient processing, this approach should produce a fairly consistent and accurate estimate. The meteoric end member, however, is estimated by a frequency distribution and is consequently less precise.

In addition to the limitations associated with estimating end member values, there are several other issues which need to be addressed before this tool can be used successfully.

The basic assumption behind end member identification is that the end members are constant. However, it is as likely that end members are represented by a range of values and that scatter in the data reflects different degrees of mixing between variable end members.

Furthermore, different aliquots of the same sample have produced somewhat

different results, probably due to the effects of sample heterogeneities, analytical error, or both.

Bromide concentrations tend to be very low in many of the samples. Analytical error can therefore be significant, and the Cl/Br ratios calculated with these data are probably not very precise.

All three of these factors, variable end members, sample heterogeneities, and current analytical limitations, can contribute to the scatter observed in the data. Methods can be developed to deal with the problems of sample heterogeneities and analytical error. However, the variability in end member values, should it be substantial, cannot be easily compensated for.

Use of ^{36}Cl to Determine Extent of Percolation

Elevated $^{36}\text{Cl}/\text{Cl}$ ratios were reported in the UZ-1 500-502 sample and in portions of the U12G tunnel. These elevated ratios provide some insight into the role that the fault and fracture system plays in movement of water in the unsaturated zone at Yucca Mountain.

The UZ-1 500-502 sample contains a higher chloride concentration than adjacent samples (9.43 ppm) and has a relatively high Cl/Br ratio of 440 (from Table 4). One possible explanation for the observed halide characteristics of this sample is mixing of rain water containing bomb-pulse ^{36}Cl and halides originating from the rock. If the meteoric and rock halide end members are assumed to be 130 and 500, respectively, then the Cl/Br ratio of the 500-502

sample is consistent with this hypothesis. Addition of rock halides to meteoric halides would tend to increase the chloride concentration and the Cl/Br ratio, but would decrease the $^{36}\text{Cl}/\text{Cl}$ ratio.

Using a mass balance estimation, the leachable halides in the 500-502 sample represent a 5% meteoric component and an 95% rock component. The meteoric component of chloride and bromide concentrations would be 0.45 ppm and 0.0035 ppm, respectively. The $^{36}\text{Cl}/\text{Cl}$ ratio of the meteoric component would be $40,445 \times 10^{-15}$. The derivation of this calculation is given in Appendix B.

The two samples immediately above the 500-502 sample (495-500 and 395-400) do not appear to contain bomb-pulse ^{36}Cl . Therefore, the high $^{36}\text{Cl}/\text{Cl}$ ratio in this sample may indicate that the borehole has intercepted a fracture at this depth, possibly originating along the west face of Yucca Mountain (Norris, 1989). It is also possible that the sample from this interval was contaminated from drilling fluid used in the upper 50 feet of the borehole; this contamination would have increased the Cl/Br ratio but would not have been likely to contribute measurable ^{36}Cl .

There are several speculations as to the cause of the high $^{36}\text{Cl}/\text{Cl}$ value in the U12G tunnel. One possibility is that a fault intersecting the tunnel may act as a conduit for rapidly percolating meteoric water. Another possibility is contamination from underground testing that has been performed in the tunnel complex. The water used in the drilling process is also a potential source of elevated ^{36}Cl concentrations.

The three DH samples, taken from the Experiment Drift, had $^{36}\text{Cl}/\text{Cl}$ ratios between 1500×10^{-15} and 2000×10^{-15} . The AC-1 samples are from the Demonstration Drift whereas the AC-2 samples are from the Laser Drift. Both sets of samples represent several different intervals along their respective boreholes.

Given the fact that the samples taken from the portion of the Experiment Drift intersected by a fault show some of the highest $^{36}\text{Cl}/\text{Cl}$ ratios, the most likely origin for the high chlorine-36 content in the U12G tunnel is bomb-pulse meteoric water percolating along the fault and into the rock matrix. This hypothesis is supported by the fact that both sets of AC samples show what appears to be a breakthrough of bomb-pulse ^{36}Cl . In the AC-1 borehole, run 16-17 has a $^{36}\text{Cl}/\text{Cl}$ ratio of 412×10^{-15} which is consistent with estimated meteoric levels. Runs 1 and 2-3, which are closer to the fault, have elevated $^{36}\text{Cl}/\text{Cl}$ ratios (1243×10^{-15} and 3044×10^{-15}), indicative of meteoric bomb-pulse contamination. Similarly, in the AC-2 borehole, the sample closer to the fault, run 38-40, shows some component of bomb-pulse $^{36}\text{Cl}/\text{Cl}$ (845×10^{-15}). Run 19-20, which is further from the fault, has a $^{36}\text{Cl}/\text{Cl}$ ratio intermediate between the estimated meteoric and rock component end members (306×10^{-15}). Interestingly, run 19-20 which shows the lowest $^{36}\text{Cl}/\text{Cl}$ ratio, is closer to the location of historic subsurface nuclear weapons testing than any of the other samples.

Unless some unknown local subsurface source of uranium or thorium is present which could account for the elevated $^{36}\text{Cl}/\text{Cl}$ ratios, the most plausible explanation for these high ratios is percolation of bomb-pulse meteoric water along the fault in the Experiment Drift. The radius of influence of the meteoric bomb-pulse appears to be between 150 and 225 feet, for the two directions

measured (Norris, 1989).

Although results were only obtained for one of the dust samples collected, if viewed quantitatively, the potential for significant dust contamination of cuttings from the tunnel is minimal. Consider the DH-1 sample, run 5-6. If the true $^{36}\text{Cl}/\text{Cl}$ ratio of this sample is comparable to the meteoric estimate of 500×10^{-15} , then 67% by weight of the sample collected would need to be dust, assuming a $^{36}\text{Cl}/\text{Cl}$ ratio of 2045×10^{-15} in the dust, in order to obtain the actual $^{36}\text{Cl}/\text{Cl}$ ratio observed. If the "uncontaminated" sample had a $^{36}\text{Cl}/\text{Cl}$ ratio closer to the estimated background level of 25×10^{-15} , then 95% of the sample weight would need to be dust in order to obtain the observed ratio. This is highly unlikely.

SUGGESTIONS FOR FUTURE WORK

Estimation of End Members

The most obvious place to start in addressing the issues affecting end member estimation is improvement and standardization of the analytical procedures. A critical part of this task will be to improve the bromide detection limit.

Improving the analytical procedures should not only produce more accurate and reliable data, but also should have the added benefit of shedding light on the extent to which variable end members and sample heterogeneity affect chemical composition.

After modifications are made in the analytical procedures, repeat analyses should be done in all sample categories (sieved samples, step-wise leaching experiment, and shatterbox experiment) in order to verify current findings.

A time-series experiment, in which a given sample is analyzed after successive increases in leaching time, could determine the optimal leaching time for future work. The UZ-1 250-255 sample would be ideal for this experiment, because it has been more extensively analyzed than any other sample. It also has relatively high chloride and bromide concentrations, which would reduce the analytical time.

Additional work is needed to provide a more accurate estimate of the halide

ratio in the rock component. The step-wise leaching experiment should be expanded to include a few additional steps. These would provide data to indicate whether or not the Cl/Br ratio has reached a plateau and therefore actually does represent the rock end member. Likewise, the shatterbox experiment can be modified to include additional crushing time to determine at what point the slope remains constant and therefore represents the halide ratio of the rock end member.

Current estimates of the rock end member are based on analysis of the 250-255 sample according to procedures for the step-wise leaching experiment, and analysis of the 295-300 sample, using procedures for the shatterbox experiment. Although these data show good agreement, more than two points are needed to arrive at a valid estimate. Additional experiments should include determination of the rock end member at different depths, using both the step-wise leaching and the shatterbox experiments. This work is particularly important given the likelihood of variable end members.

Improved analytical procedures should yield a better estimate of the meteoric halide ratio. However, an independent estimate of the meteoric Cl/Br ratio can be obtained by determining the ratio in local precipitation.

Problems arising due to sample heterogeneities can be minimized by performing chloride, bromide, and ^{36}Cl analyses on the same subsample. This procedure is cumbersome in the case of chloride and bromide analyses, since the sample volume is so large.

Use of ^{36}Cl to Determine Extent of Percolation

The ^{36}Cl analyses conducted on UZ-1 samples indicate that bomb-pulse ^{36}Cl is present at a depth of 500 feet. The low $^{36}\text{Cl}/\text{Cl}$ ratio in the sample immediately overlying the 500-502 interval implies that the extent of this "bomb-pulse contamination" may be very limited. Additional work should focus on identifying the profile of the $^{36}\text{Cl}/\text{Cl}$ ratio in an extended interval centered on 500 feet with particular attention paid to examination of the lithology of this depth interval.

Results from the U12G tunnel support the theory that the fault and fracture system at Yucca Mountain may be providing pathways for the relatively rapid percolation of meteoric water. Additional work should focus on identifying the $^{36}\text{Cl}/\text{Cl}$ profile at distance from the fault. This could be accomplished by drilling six orthogonal boreholes, radiating outward from the fault in the Experiment Drift, and collecting samples for ^{36}Cl analyses (Norris, 1989).

Additional dust samples should be collected and analyzed for ^{36}Cl , in order to address the possibility of contamination. However, to avoid controversy, a procedure should be developed to ensure that samples removed from the sample bag contain only cuttings from inside the bag and no dust from the outside of the bag (Norris, 1989).

Work in the U12G tunnel is particularly significant because these samples provide insight into a three dimensional distribution of ^{36}Cl .

CONCLUSION

Samples from the UZ-1 and UZN-43 boreholes at Yucca Mountain have been analyzed for chloride, bromide, and ^{36}Cl in order to understand the history of local infiltration and flow in the unsaturated zone.

The step-wise leaching experiment and the shatterbox experiment are used to isolate and identify the Cl/Br ratio of the rock matrix. The halide ratio of the meteoric component is estimated using a frequency distribution. These values are 130:1 for the meteoric end member, and 500:1 for the rock end member.

Various factors, such as variable end members, sample heterogeneities, and current analytical limitations, may affect the validity of these estimates. Additional work is planned which will standardize the analytical procedures and expand the current experimental work. The result should be more accurate and verifiable data, in addition to a better estimate of end member values.

Chlorine-36 data indicate that meteoric water containing bomb-pulse ^{36}Cl is present at a depth of 500 feet in the UZ-1 borehole. Similar high $^{36}\text{Cl}/\text{Cl}$ ratios are found in the U12G tunnel. It appears that the fault system at Yucca Mountain is behaving as a conduit for percolating water, and that ^{36}Cl measurement may be a valuable tool in the identification of such faults.

The current value of ^{36}Cl data will be enhanced once the meteoric and rock end members are accurately defined. Corrections can then be made in the ^{36}Cl data so that a more accurate picture of local percolation can be obtained.

Preliminary estimates for the $^{36}\text{Cl}/\text{Cl}$ ratio of the meteoric and rock end members are 500×10^{-13} and 25×10^{-13} , respectively.

REFERENCES

- Bentley, H. W., Phillips, F. M., Davis, S. N., Gifford, S. K., Elmore, D., Tubbs, L. E., and Gove, H. E. 1982. Thermonuclear ^{36}Cl pulse in natural water. *Nature* 300(5894), pp.737-740.
- Bentley, H. W., Phillips, F. M., and Davis, S. N. 1985. Chapter 10: Chlorine-36 in the terrestrial environment. *Handbook of Environmental Isotope Geochemistry*, P. Fritz and J. C. Fontes (editors). Elsevier.
- Cheng, S. L. 1989, personal communication.
- Davis, S. N. 1989, personal communication.
- Elmore, D., Fulton, B. R., Clover, M. R., Marsden, J. R., Gove, H. E., Naylor, H., Purser, K. H., Killius, L. R., Beukins, R. P., and Litherland, A. E. 1979. Analysis of Cl-36 in environmental water samples using an electrostatic accelerator. *Nature* 277, pp.22-25.
- Fabryka-Martin, J. T. 1989, personal communication.
- Gifford, S. K. 1987. Use of chloride and chlorine isotopes in the unsaturated zone to characterize recharge at the Nevada Test Site, unpublished Masters thesis, University of Arizona, Department of Hydrology.
- Hydro Geo Chem. 1985. Use of Cl and Cl Isotopes in the Unsaturated Zone to Characterize Recharge at the Nevada Test Site. Annual Report prepared for Los Alamos National Laboratory.
- Junge, C. E. and Gustafson, P. E. 1957. On the distribution of sea salt over the United States and its removal by precipitation. *Tellus*. 9, p.164.
- Montazer, P. and Wilson, W. E. 1984. Conceptual Hydrologic Model Flow in the Unsaturated Zone, Yucca Mountain, Nevada. U. S. Geological Survey Report, USGS-WRI-84-4345.
- Norris, A. E., Wolfsberg, K., Gifford, S. K., Bentley, H. W. and Elmore, D. 1987. Infiltration at Yucca Mountain, Nevada, Traced by ^{36}Cl . *Nuclear Inst. Meth. in Phys. Res.* B29:276.
- Norris, A. E. 1989 personal communication.
- Phillips, F. M., Trotman, K. N., Bentley, H. W., Davis, S. N. 1984. Bomb ^{36}Cl pulse as a tracer for soil-water movement near Socorro, NM. In: *Selected papers on water quality and pollution in New Mexico*. W. B. Stone, ed. New Mexico Bureau of Mines and Mineral Resources, Hydrologic Report #7, pp. 271-280.

REFERENCES -- Continued

- Waters, A. C. and Carroll, P. R. (editors), 1981. Preliminary Stratigraphic and Petrologic Characterization of Core Samples from USW-G1, Yucca Mountain, Nevada. Los Alamos National Laboratory Report #LA-8840-MS.
- Whittemore, D. O. 1988. Bromide as a tracer in groundwater studies: geochemistry and analytical determination. Proceedings of the Ground Water Geochemistry Conference. National Water Well Association, Denver, Colorado.

**APPENDIX A:
PROCESSING OF ROCK SAMPLES FOR
CHLORIDE, BROMIDE, AND CHLORINE-36 ANALYSES**

SAMPLE SIEVING AND CRUSHING

The sieving process is designed to evaluate the effect of variable grain size on leachable halides. Each sample is sieved into four fractions using the 35, 60, and 230 mesh screens. Between each sample the sieves are brushed to dislodge grains that may be stuck in the screens, and both the sieves and pans are washed with distilled water. The sieves are dried in a convection oven (Toastermaster Model #7071) before use.

Rock samples are crushed in a shatter box (Donaldson Model #90-140, 7.5 HP, with 14 cm ID by 5 cm depth steel mill). Crushing time is used as a relative indicator of grain size.

Processing of a sample begins by loading approximately 300 ml of dry sample powder into the shatterbox. At least 50 ml of the original sample is reserved. The 300 ml sample is pulverized for 2 minutes, at which point an aliquot of sample (about 30 ml) is taken. The remaining sample is reloaded into the shatterbox and crushing is resumed. This cycle is repeated several times, with crushing intervals ranging from 2 to 5 minutes. The total crushing time is 25 minutes, unless otherwise specified. The shatterbox is cleaned with distilled, deionized water and dried with compressed air between samples.

Sample Leaching

Leachable halides are extracted from the rock samples by mixing known amounts of rock powder (10 to 50 gm) with measured amounts of distilled,

Determination of Concentrations

The concentration of leachable chloride in the rock sample is calculated as follows:

$$Cl \text{ (rock)} = \frac{Cl \text{ (water)} \times V \text{ (water)}}{Wt \text{ (rock sample)}}$$

where: $Cl \text{ (rock)}$ = leachable chloride concentration
in rock sample in ppm
 $Cl \text{ (water)}$ = leachable chloride concentration
in water in mg/l
 $V \text{ (water)}$ = volume of water used for leaching
in liters
 $Wt \text{ (rock)}$ = weight of rock sample in kg

The same leachate is used for bromide analysis. Leachable bromide is calculated using the same equation.

Preparation of Chloride for Chlorine-36 Analyses

Leaching

Depending on the leachable chloride concentration of the rock sample, 1 to 20 kg of rock are mixed with distilled, deionized water in a polyethylene container. The sample is allowed to leach at least overnight, although preferably for several days. The water is then decanted from the container and saved in a pyrex flask or beaker. More water is added to the rock sample for additional leaching. This cycle is repeated for a total of at least three times in order to achieve a final chloride content of 10 mg. This is the minimum

required for analysis on the tandem accelerator.

Chloride Extraction

Fine particles in suspension make it difficult to separate leachate from the bulk sample. These particles can be coagulated by adding Ultrex nitric acid at a ratio of 2 ml per liter of leachate. Further clarification is achieved by filtering the leachate through a 0.45 μm filter. Nucleopore filter membranes are not used because they may contain excess ^{36}Cl .

Enough silver nitrate (AgNO_3) is added to the clear leachate to precipitate the total estimated chloride present as silver chloride (AgCl). If the total volume of leachate contains less than 10 mg of chloride, then it becomes necessary to add a known amount of ^{36}Cl -free chloride as a carrier, to bring the total amount of chloride up to at least 10 mg. This introduction of carrier should be done prior to addition of AgNO_3 .

The AgCl precipitate is collected by filtering the leachate through a 0.45 μm filter. The rate and efficiency of filtration can be increased by gently heating the leachate to just below boiling prior to filtering in order to increase the size of the AgCl particles. However, the leachate should be cooled before filtration in order to avoid loss of AgCl dissolved in the hot water.

Purification of Silver Chloride

The AgCl precipitate is washed with distilled, deionized water on a filter

**APPENDIX B:
ANALYTICAL DERIVATIONS**

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX. (602) 623-6984

October 9, 1990

WBS: 1.2.3.3.2.1
QA
TWS-INC-7-10-90-11

Dr. June Fabryka-Martin
Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Subject: Review of work performed during FY90

Dear June:

This letter summarizes the work performed by Hydro Geo Chem in fiscal year 1990, and the work that is currently in progress.

The fiscal year began with a change of project manager at Hydro Geo Chem; Songlin Cheng left the company and Susan Maida took over the position. After a preliminary review of the scope of the project by the new project manager, we prepared a formal report documenting work done in fiscal year 1989. Hydro Geo Chem spent several months renovating and upgrading its in-house laboratory in order to expand capabilities. Laboratory work was not possible during this period of time.

Beginning in February, time was spent modifying the chlorine-36 sample preparation procedure in order to improve yields. Previous work had produced the necessary AgCl using a procedure that relied on filtration to separate the AgCl precipitate from the aqueous solution. Reprecipitation was accomplished by heating the NH_4OH -AgCl solution to evaporate the NH_4OH . This approach had been satisfactory for samples containing abundant chloride. However, there were many steps involving physical transfer from one container to another, with chloride potentially lost at each one of those steps. This loss was a significant problem with samples containing low chloride. The AgCl yield was improved substantially by modifying the sample preparation procedure so that a centrifuge is used in place of filtration and reprecipitation is accomplished by acidification. Preliminary results using this modified approach have produced yields of close to 95% on samples containing no more than 10 mg of chloride.

Dust samples from the U12G Tunnel were prepared for chlorine-36 analyses, using the new procedure. However, these samples were never analyzed because of potential contamination in the Tunnel and because the focus of the project shifted.

After project leadership was transferred from Ted Norris to you, we shifted our focus to a potential new site to characterize at Apache Leap, Arizona. We anticipated quality samples since we felt we would have some control over the drilling technique. Rock cuttings were collected from a deep borehole using a prototype, dry-drilling technique. Unfortunately, these samples were sufficiently dissimilar to the samples from Yucca Mountain that their use was questionable.

We are currently caretakers to a relatively large volume of soil and rock samples. These samples have been organized and inventoried, with results included for the UZ-1 borehole.

Significant time was spent updating our quality assurance matters, preparing for an audit by Los Alamos National Laboratory, and being trained under the Yucca Mountain Project Quality Assurance Program.

Work has also been done preparing quality assurance documents for a collaborative effort with Sandia National Laboratory, characterizing a site in the vicinity of Frenchman Flats, Nevada. We are currently participating in the site selection and project design phase. We anticipate using chloride, bromide, and chlorine-36 as environmental tracers, while Sandia National Laboratory looks at tritium, oxygen-18, and deuterium.

As we move into the last months of this year we will be devoting our time to developing a new column leaching technique that we believe will be more effective and efficient than the method we have been using to date. We will also be preparing several samples that represent the meteoric and rock end members for stable chlorine isotope analyses.

Sincerely,

Susan Maida

cc: 1 copy TWS
2 copies RPC

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

FAXED THIS DATE

31 July 1989

Dr. Ted Norris
Los Alamos National Laboratory
TWS-INC7, MS J-514
Los Alamos, New Mexico 87545

Dear Ted:

This letter summarizes the work performed by Hydro Geo Chem during the first 10 months of the fiscal year 1988-1989, and the work that is in progress. Enclosed are tables and figures of analytical results.

A. Halide Analyses

We have sieved UZ-1 (24 samples) and UZN-43 (8 samples) and analyzed these samples for chloride and bromide. Each sample was sieved into four fractions. The sieve sizes used were 35, 60, and 230 mesh. Table 1 lists the analytical results. Repeat analyses of some UZN-43 samples indicated that some usually low Cl/Br ratios during the first analysis may have been caused by contamination or analytical error. Figures 1(a) and (b) plot the chloride vs. bromide concentrations including lines of Cl/Br equal to 130:1 and 500:1. Based on the observed ratios, we hypothesize that these two ratios represent rain water and rock halide components in the system, respectively.

B. Step-wise Crushing and Leaching Experiments

Because the rock samples from Yucca Mountain have been pulverized to various degrees during drilling, their leachable halides contain both meteoric and rock components. The step-wise crushing-leaching experiment was designed to characterize the halide ratio of the rock end member. The experimental procedure includes a sequence of leaching, washing, drying and shatter-box crushing of rock samples followed by another sequence. After each sequence, the halide was analyzed for chloride, bromide, and $^{36}\text{Cl}/\text{Cl}$ ratios.

July 31, 1989
Dr. Ted Norris
Los Alamos National Laboratory
Page 2

Table 2 lists the analytical results of the experiment. Samples 250-250 B (step) and 250-255 C (step) contain high NO_3^- , which interferes with the bromide analysis. A possible source of NO_3^- is nitric acid used in processing sampling for ^{36}Cl analysis.

We have also continued and expanded the shatter-box experiments. Table 3 lists the chloride and bromide concentrations of UZ-1 samples 97-101, 250-255, and 295-300 after crushing in a shatter-box. The sequence of digits following the sample designation is the sequence of crushing time in minutes. For example, the sequence "2333" indicates that the sample was crushed for 2 + 3 + 3 + 3 minutes. Figure 2 plots the bromide vs. chloride concentrations of these three samples. The slopes of these three lines indicate the chloride/bromide ratios of halides released from the rock matrix upon crushing. Results from sample UZ-1 (295-300) indicate that the rock halides' Cl/Br ratio is ~500. The halides of samples UZ-1 (250-255) and UZ-1 (97-101) that were produced by crushing may be a mixture of meteoric halides (Cl/Br = 130) concentrated in small pore spaces and rock halides (Cl/Br = 500); therefore, the samples would have a Cl/Br ratio between the mixing end members. Unlike the other samples, the leachable halide concentration of UZ-1 (97-101) decreases upon crushing. The moisture content of this sample was so high that the sample was molded into a dense clayey cake upon crushing in the shatter-box, which is likely the cause of lower halide concentration. Additional study is in progress to continue the shatter-box experiment on sample UZ-1 (250-255) so that the rock end member (assumed to be Cl/Br = 500/1) shows on the plot.

C. ^{36}Cl Analyses

Nine UZ-1 and UZN-43 samples were analyzed for chlorine-36 including a blank. Table 4 lists the chlorine-36 results. High chlorine-36 was observed in the UZ-1 drill hole at depth intervals of 97-101 feet and 500-502 feet. These intervals may contain bomb-pulse chlorine-36. The sample at 500-502 feet also contains a higher chloride concentration than the adjacent samples and has a higher Cl/Br ratio of ~400 (Table 1). A plausible explanation for the halide characteristics of UZ-1 500-502 is mixing between rainwater containing chlorine-36 of bomb-pulse origin and halides originating from the rock. Because the rainwater and rock halides may have Cl/Br ratios of 130 and 500, respectively, the observed Cl/Br ratio

July 31, 1989
Dr. Ted Norris
Los Alamos National Laboratory
Page 3

of the 500-502 foot sample is consistent with this mixing hypothesis. Addition of rock halides to rain halides would have increased chloride concentration, but decreased the Cl/Br and $^{36}\text{Cl}/\text{Cl}$ ratio. Based on mass balance estimation, the leachable halides of UZ-1 500-502 contain a 14% rainwater halide component and an 80 % rock component. If so, the rainwater component of this sample would be a chloride, bromide, and $^{36}\text{Cl}/\text{Cl}$ ratio of 1.23 ppm, 0.095 ppm, and 14600×10^{-15} , respectively.

D. Step-wise Leaching ^{36}Cl Results

The step-wise leaching experiments of UZ-1 250-255 (Table 5), indicate that the rock halide may contain a $^{36}\text{Cl}/\text{Cl}$ ratio less than 25×10^{-15} . Sample 250-255 E (step) contains very low chlorine-36 (25×10^{-15}) and has a Cl/Br ratio of 487. This sample may represent a rock end member. We will calculate the subsurface production of chlorine-36 if U and Th content of this rock unit are available. Seth Gifford (unpublished M.S. thesis, University of Arizona, 1987) measured two $^{36}\text{Cl}/\text{Cl}$ ratios of trench samples and suggested that the pre-bomb $^{36}\text{Cl}/\text{Cl}$ ratio is 500×10^{-15} to 600×10^{-15} . Based on sample 250-255 E (step) as the rock end member, we assume the $^{36}\text{Cl}/\text{Cl}$ and Cl/Br ratios of rain and rock end members to be (500×10^{-15} , 130) and (25×10^{-15} , 500), respectively. Figure 3 plots the analytical results of sample 250-255 and the theoretical mixing curve. Except sample 250-255 D (step), the data follow the theoretical curve closely. We will re-analyze chloride and bromide concentrations of step-wise leached samples. Three UZ-1 samples (500-502, 1020-1025, and 1195-1200) were processed for chlorine-36 analysis after leaching, washing, drying, and 3-minute crushing. $^{36}\text{Cl}/\text{Cl}$ ratios of these three samples decreased as leaching progressed by about 100×10^{-15} and Cl/Br ratios increased (See Table 2). Because of low bromide concentrations, the analytical error could be high; therefore, the reported Cl/Br ratios are not very precise. More chloride and bromide analyses will be done on these samples, with procedures modified to increase analytical sensitivity.

E. Additional Halide and ^{36}Cl Results

Six samples obtained from U12G tunnel were analyzed for halides and ^{36}Cl . The results are shown in Table 6. Three samples from U12G tunnel contains over 1500×10^{-15} of $^{36}\text{Cl}/\text{Cl}$. We feel that the most likely source of this high ^{36}Cl content is meteoric water that percolated along the fault that cuts through the Experiment Drift.

July 31, 1989
Dr. Ted Norris
Los Alamos National Laboratory
Page 4

Other possible sources include the water used in the drilling process and high ^{36}Cl originating from underground bomb tests. Studies are in progress to identify the source(s) of high chlorine-36 content in DH-1 and DH-2 samples.

F. Work in Progress

We are in the process of repeating chloride and bromide analyses to ensure the analytical quality. We are also currently processing nine samples for chlorine-36 analysis; these samples are mostly from U12G tunnel.

We will apply a mass-balance approach to calculate $^{36}\text{Cl}/\text{Cl}$ ratios of the rainwater component once chloride and bromide analyses are finalized. We will use a weighted average with correction for the rock component to construct a depth profile of meteoric halide and $^{36}\text{Cl}/\text{Cl}$ ratio at UZ-1.

G. Discussion of Results and Suggested Future Experiments

Based on chlorine-36 analysis at UZ-1 site, bomb-pulse chlorine-36 may have penetrated as deep as 500 feet. The presence of high chlorine-36 at a depth of 500 - 502 foot level and absence of high chlorine-36 in the adjacent sample intervals suggests that fracture system in the Yucca Mountains could behave as a conduit for water movement. Both these results and those of the U12G tunnel samples suggest that chlorine-36 could be a very effective tool to identify the presence of water-conducting fractures.

Regarding the halide composition of rock, one important question is that of uniformity. To address the question of uniformity, we need to process more rock samples and isolate the rock component by step-wise leaching.

We believe the above analytical results and proposed experiments are of fundamental importance in understanding the halide geochemistry of Yucca Mountain. When sufficient data are available, we will prepare a manuscript for publication.

July 31, 1989
Dr. Ted Norris
Los Alamos National Laboratory
Page 5

More work will be done on this project in the remaining two months of this fiscal year. We therefore consider this Progress Report to be interim. We will prepare a final Annual Report in late September. Please call me if you have any questions or comments regarding either this Summary or the work in general.

Sincerely,

Songlin Cheng

SC/cah

Enclosures

TABLE 1. CHLORIDE, BROMIDE, AND CHLORIDE/BROMIDE RATIOS OF UZN-43 AND UZ-1 SAMPLES.

SAMPLE	FROM	TO	-----CHLORIDE, PPM IN ROCK-----				-----BROMIDE, PPM IN ROCK-----				----- CHLORIDE/BROMIDE RATIO -----			
			A	B	C	D	A	B	C	D	A	B	C	D
UZN-43	6	7	69.74	154.70	134.30	142.80	2.782	13.540	13.720	17.530	25.1	11.4	9.8	8.1
	10	11	58.61	156.50	15.75	162.60	5.581	2.790	14.820	7.286	10.5	56.1	1.1	22.3
	15	16	45.85	87.81	89.07	76.47	0.145	0.337	0.329	0.312	316.2	260.6	270.7	245.4
	20	21	3.18	6.91	5.87	11.72	0.022	0.034	0.037	0.060	142.7	205.1	157.7	196.0
	25	26	1.45	3.81	5.08	10.66	0.012	0.025	0.027	0.052	122.2	150.1	189.4	205.4
	30	31	7.23	5.35	5.98	7.10	0.049	0.015	0.013	0.012	148.1	349.4	474.2	582.1
	35	36		7.89	6.66	7.68		0.011	0.016	0.006		744.7	410.9	1396.9
	40	41	1.01	1.36	1.47	2.72	0.005	0.003	0.007	0.005	190.4	430.5	223.2	524.9
UZN-43 REPEATS														
	6	7	99.40	289.31	298.59	377.29	0.460	1.427	1.483	1.942	215.9	202.7	201.3	194.3
	10	11	11.79		34.84		0.051		0.153		231.2		228.5	
	35	36	9.03				0.025				368.6			
UZ-1			-----CHLORIDE, PPM IN ROCK-----				-----BROMIDE, PPM IN ROCK-----				-----CL/BR RATIO IN ROCK-----			
			A	B	C	D	A	B	C	D	A	B	C	D
UZ-1	97	101	28.28	37.66	44.74	55.36	0.167	0.163	0.159	0.274	169.7	231.8	282.3	202.1
	250	255	22.58	25.03	25.66	30.51	0.182	0.168	0.184	0.241	123.8	148.9	139.2	126.5
	295	300	1.82	2.01	3.23	3.74	0.012	0.015	0.014	0.023	147.2	138.3	223.5	160.5
	347	350	7.64	4.22	3.53	3.57	0.052	0.034	0.023	0.025	148.1	124.4	152.2	142.6
	495	500	5.23	5.37	5.11	6.91	0.025	0.019	0.025	0.021	210.0	279.0	208.4	336.8
	500	502	8.67	10.01	10.31	7.30	0.017	0.024	0.023	0.037	499.1	411.6	452.6	195.7
	595	599	7.97	7.40	7.94	9.15	0.054	0.048	0.057	0.058	146.7	153.1	138.3	156.7
	715	720	0.96	0.95	1.20	1.28	0.010	0.014	0.011	0.009	94.1	70.6	111.7	141.8
	830	835	1.96	2.18	1.86	2.40	0.007	0.009	0.006	0.010	270.5	244.1	327.2	238.1
	845	850	4.96	2.32	1.82	2.01	0.014	0.013	0.014	0.012	352.5	181.1	131.6	162.3
	920	925	2.12	2.22	2.39	2.68	0.025	0.022	0.020	0.021	84.1	99.6	120.9	129.3
	945	950	2.85	2.52	2.04	3.18	0.020	0.010	0.011	0.035	145.6	249.4	181.3	89.9
	970	975	3.45	2.61	3.33	3.86	0.019	0.019	0.012	0.012	177.7	140.5	268.5	321.4
	1020	1025	1.15	2.28	2.29	2.94	0.009	0.015	0.015	0.023	125.7	156.7	154.5	129.4
	1047	1050	2.32	4.33	2.52	3.34	0.010	0.014	0.011	0.014	236.5	320.2	231.3	237.7
	1070	1075	2.31	1.72	2.31	3.39	0.010	0.013	0.012	0.011	228.2	130.6	191.1	311.0
	1147	1150	4.46	5.15	4.17	6.66	0.022	0.015	0.012	0.021	198.2	344.3	340.0	321.9
	1170	1175	3.76	3.68	2.88	6.66	0.014	0.015	0.016	0.015	259.3	244.7	185.7	449.7
	1195	1200	2.59	2.87	2.27	2.97	0.017	0.017	0.016	0.024	149.9	171.3	138.1	125.9
	1220	1225	3.18	3.88	3.70	5.43	0.017	0.013	0.016	0.046	190.7	303.9	236.8	118.2
	1245	1250	4.17	3.90	3.76	3.65	0.018	0.021	0.015	0.012	227.8	189.7	248.3	301.7
	1258	1260	6.88	10.48	11.74	13.77	0.043	0.076	0.067	0.087	160.2	137.6	174.4	158.1
	1267	1268	1.95	3.81	4.43	13.26	0.008	0.015	0.014	0.027	248.3	258.1	309.8	483.2
	1268	1270	2.94	3.82	3.99	4.93	0.015	0.015	0.024	0.031	196.8	252.4	168.4	159.8

TABLE 2. RESULTS OF STEP-WISE LEACHING EXPERIMENTS OF UZ-1 SAMPLES.

-----SAMPLE-----			--PPM IN SOLUTION-WATER			SAMPLE WEIGHT	----PPM IN ROCK----			CL-36/CL 1E-15
FROM	TO	EXT.**	CL	BR	VOLUME		CL	BR	CL/BR	
500	502	(3)								1885
1020	1025	(3)	0.935	0.0011	50	25.09	1.863	0.0022	850.0	159
1195	1200	(3)	1.044	0.0025	50	25.37	2.058	0.0049	417.6	340
1220	1225	(3)	0.997	0.0031	50	25.42	1.961	0.0061	321.6	
250	255	2 MIN.	8.563	0.0317	20	10.21	16.774	0.0621	270.1	
		B(STEP)	2.460	*	20	10.28	4.786			91
		C(STEP)	9.080	*	20	10.11	17.962			
		D(STEP)	2.437	0.0090	20	10.02	4.864	0.0180	270.8	36
		E(STEP)	4.648	0.0095	20	10.02	9.277	0.0190	489.3	25
250	255	A(STEP)	10.900	0.0405	50	10.01	54.421	0.2022	269.1	193
		B(STEP)	0.740	*	50	10.03	3.689			91
		C(STEP)	2.380	*	50	10.03	11.860			
		D(STEP)	1.200	0.0054	50	10.04	5.975	0.0269	222.2	36
		E(STEP)	1.900	0.0039	50	10.12	9.387	0.0193	487.2	25

* : CONTAMINATED BY HIGH NO3-

** : SAMPLE LEACHING/CRUSHING STEPS ARE DESCRIBED BELOW:

(3) : SAMPLE WAS LEACHED, DRIED, CRUSHED FOR 3 MINUTES, AND
LEACHED FOR HALIDE ANALYSIS

2 MIN. : ORIGINAL SAMPLE WAS CRUSHED FOR 2 MINUTES BEFORE LEACHING.

A(STEP): ORIGINAL SAMPLE WAS LEACHED, WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND
LEACHED FOR HALIDE ANALYSIS.

B(STEP): SAMPLE A(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND
LEACHED FOR HALIDE ANALYSIS.

C(STEP): SAMPLE A(STEP) WAS WASHED, DRIED, CRUSHED FOR 10 MINUTES, AND
LEACHED FOR HALIDE ANALYSIS.

D(STEP): SAMPLE B(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND
LEACHED FOR HALIDE ANALYSIS.

E(STEP): SAMPLE D(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND
LEACHED FOR HALIDE ANALYSIS.

TABLE 3. CHLORIDE AND BROMIDE RESULTS OF SHATTER BOX EXPERIMENTS

DRILL HOLE	-----SAMPLE-----			--PPM IN SOLUTION--		WATER VOLUME C.C.	SAMPLE WEIGHT GM	----PPM IN ROCK----		
	FROM	TO	TIME SEQ. (MINUTES)	CL	BR			CL	BR	CL/BR
UZ-1	250	255		8.164	0.0445	50	10.89	37.484	0.2043	183.5
			2	9.846	0.0527	50	10.01	49.181	0.2632	186.8
			23	13.702	0.0643	50	10.02	68.373	0.3209	213.1
			233	14.608	0.0665	50	10.03	72.822	0.3315	219.7
			2333	17.036	0.0798	50	10.05	84.756	0.3970	213.5
			23334	23.439	---	50	10.05	116.612		
			233345	34.195	---	50	10.04	170.294		
			2333455	---	---	50	10.01			
	97	101		12.634	0.0457	50	10.07	62.731	0.2269	276.5
			2	10.830	0.0425	50	10.08	53.720	0.2108	254.8
			23	11.024	0.0398	50	10.11	54.520	0.1968	277.0
			233	10.714	0.0379	50	10.01	53.516	0.1893	282.7
			2333	10.714	0.0376	50	10.01	53.516	0.1878	284.9
			23334	10.145	0.0325	50	10.02	50.624	0.1622	312.2
			233345	10.070	---	50	10.10	49.851		
			2333455	9.157	0.0322	50	10.08	45.422	0.1597	284.4
	295	300		0.274	0.0031	50	10.40	1.317	0.0149	88.4
			2	0.451	0.0036	50	10.01	2.253	0.0180	125.3
			23	0.741	0.0042	50	10.13	3.657	0.0207	176.4
			233	1.238	0.0053	50	10.01	6.184	0.0265	233.6
			2333	1.765	0.0067	50	10.01	8.816	0.0335	263.4
			23334	2.525	0.0090	50	10.04	12.575	0.0448	280.6
			233345	---	---	50	10.04			
			2333455	3.947	0.0109	50	10.01	19.715	0.0544	362.1

TABLE 4. CHLORINE-36 CONTENTS OF ROCK SAMPLES FROM YUCCA MOUNTAIN.
(REPORTED AS ATOMIC RATIO OF CL-36/TOTAL CHLORIDE.)

SAMPLE NAME	CL-36/CL	
	1E-15	+/-
UZ-1 97-101	11400	360
UZ-1 250-255	436	25
UZ-1 500-502	2046	103
UZ-1 1020-1025	245	38
UZ-1 1195-1200	454	61
UZ-1 1220-1225	102	11
UZN-43 6-7	759	59
UZN-43 10-11	526	38
UZN-43 15-16	470	25

TABLE 5. STEP-WISE LEACHING CL-36 RESULTS FOR UZ-1 SAMPLES.
(REPORTED AS ATOMIC RATIO OF CL-36/TOTAL CHLORIDE.)

SAMPLE NAME		CL-36/CL	
		1E-15	+/-
UZ-1 500-502 (3)	**	1885	150
UZ-1 1020-1025 (3)	**	159	12
UZ-1 1195-1200 (3)	**	340	15
UZ-1 250-255 A(STEP)	**	193	7
UZ-1 250-255 B(STEP)	**	91	4
UZ-1 250-255 D(STEP)	**	36	11
UZ-1 250-255 E(STEP)	**	25	5

** : SAMPLE LEACHING/CRUSHING STEPS ARE DESCRIBED BELOW:

(3) : ORIGINAL SAMPLE WAS LEACHED, DRIED, CRUSHED FOR 3 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.

A(STEP): ORIGINAL SAMPLE WAS LEACHED, WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.

B(STEP): SAMPLE A(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.

D(STEP): SAMPLE B(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.

E(STEP): SAMPLE D(STEP) WAS WASHED, DRIED, CRUSHED FOR 5 MINUTES, AND LEACHED FOR HALIDE ANALYSIS.

TABLE 6. CHLORINE-36 CONTENTS OF BLANK AND SAMPLES FROM U12-G TUNNEL.

(REPORTED AS ATOMIC RATIO OF CL-36/TOTAL CHLORIDE.)

SAMPLE NAME	CL-36/CL	
	1E-15	+/-
AC-1 RUNS 16 & 17	412	18
AC-2 RUNS 19 & 20	306	22
AC-2 RUNS 38-40	845	76
DH-1 RUNS 5 & 6	1539	101
DH-1 RUNS 7-9	1964	75
DH-2 RUNS 7 & 8	1709	70
NBS SRM#919 NaCl	0	5

Figure 1(a). UZ-1 BROMIDE VS. CHLORIDE

Figure 1(b) UZ-1 BROMIDE VS. CHLORIDE

SHATTER BOX EXPERIMENTS

Figure 2.

UZ-1 250-255 STEP-WISE LEACHING

Figure 3.

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

29 September 1988

Dr. Ted Norris
Los Alamos National Laboratory
TWS-INC7, MS J-514
Los Alamos, New Mexico 87545
FAX: (505)667-2964

Dear Ted,

The letter summarizes the work performed by Hydro Geo Chem during the fiscal year 1988. The work can be divided into three categories:

- I. Chloride analysis. We purchased a Dionex Ion Chromatograph subsystem in May. The system has been successfully set up for routine chloride analysis. With this analytical capability, we now can expedite our experimental progress in a timely and accurate manner.
- II. Induction furnace. We have designed and order sample crucibles and chloride traps. The cooling system is currently under construction and the 3 phase/480 volt power line has been installed. We expect to test the performance of this chloride extraction system soon.
- III. Rock sample processing. We have leached and analyzed 15 UZ-1 samples to establish depth profile of leachable chloride. Eight samples were selected for processing to analyze chloride that is leachable after crushing in a shatter box. We have also finished shatter box experiments on sample 250-255 to study the effects of grain size distribution on leachable chloride. The source of chloride released upon crushing will be investigated. Depth profile and Cl-36 of this chloride could be the keys to the answer.

We are currently processing UZ-1 samples 97-101 and 250-255 for Cl-36 analysis and expect to finish this work by the end of this week. The leached samples will be crushed and processed for Cl-36 for two more repeating cycles. When all the experiments and analyses are done, we should be able to establish the chloride, and hence meteoric water, infiltration history of the UZ-1 site.

We have leached pebbles and fine particles (<60 mesh) of eight UZN-43 samples. The leached pebbles were subsequently washed, dried, crushed in the shatter box and then leached for chloride analysis. Both pebble and fine particle fractions of UZN-43 have relatively high chloride concentrations on the top 16 feet ^{to} and the lower 20 feet. However, the washed and crushed samples have a more or less constant profile.

It was nice to know that the current fiscal year contract will be extended to the end of October. Although it is shorter than we would like to have. Nevertheless, this extension should give us enough time for processing UZ-1 samples. I hope to see you soon so that we can discuss the results of our experiments and the work for the next fiscal year. Please let me know if you have any questions regarding this progress summary.

Sincerely yours,

Songlin Cheng
Project Manager

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

January 15, 1991

Dr. June Fabryka-Martin
Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear June:

This letter summarizes the work covered by the enclosed invoice for the months of October through December, 1990. Work covered by this invoice includes sample preparation and analyses of stable chlorine isotopes for five samples from the UZ-1 borehole. Results were satisfactory and showed a correlation with the Cl/Br ratio. The attachment to this letter gives the analytical results. Work was also done developing a column leaching set-up to replace the large volume leaching presently done in five gallon plastic jugs. Initial results were favorable; future work will focus on refining the details of the procedure, such as, decreasing the flow rate, reducing the amount of colloidal material suspended in the leachate, and determining the number of pore volumes moving through the sample.

Time was spent preparing outlines and draft versions of detailed procedures for sample preparation and analyses. These will be submitted during the next quarter for incorporation into Los Alamos National Laboratory's Quality Assurance Program Plan.

We have recently completed the arduous task of taking inventory of all of the Yucca Mountain samples in our possession. This information will be forthcoming. We were also audited by Los Alamos National Laboratory in October and spent some time in preparing for this event.

Supplies needed for the laboratory were purchased, including a mechanical beam balance for weighing samples, a large hot plate for evaporating sample leachate down to manageable volumes, and miscellaneous glassware and chemical reagents.

Please let me know if you have any questions concerning the work involved or the charges covered by this invoice.

Sincerely,

A handwritten signature in cursive script, appearing to read "Susan".

Susan Maida

Enclosure: Invoice #9047-29

Analytical Results

Sample	$\delta^{37}\text{Cl}$, ‰	Cl/Br
UZ-1, 250-255	0.3	136
UZ-1, 845-850	1.0	212
UZ-1, 1147-1150	0.9	287
duplicate	0.8	287
sea water blind	0.0	
	0.0	

Values of $\delta^{37}\text{Cl}$ are expressed relative to San Diego sea water (SDSW), for which $\delta^{37}\text{Cl} = 0\text{‰}$. Analytical precision = $\pm 0.18\text{‰}$ (2σ), based on many analyses of SDSW.

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

August 16, 1990

Dr. June Fabryka-Martin
Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear June:

This letter summarizes the work covered by the enclosed invoice for the months of May through July, 1990. Work covered by this invoice includes researching relevant procedures and writing a document to address quality assurance objectives for the upcoming Sandia site characterization at Frenchman Flats. Travel time to the initial project meeting in Albuquerque and related costs are included in this invoice.

Other work covered by this invoice includes preparation for the first meeting between you and your colleagues and Hydro Geo Chem. Stan Davis was a consultant both prior to and during this meeting. Quality assurance work was performed in preparation for the audit, including a meeting with Terry Morgan. Work continues in the area of a detailed sample inventory. And lastly, work has begun on refining the analytical procedures to improve bromide sensitivity.

Please let me know if you have any questions concerning the work involved or the charges covered by this invoice.

Sincerely,

Susan Maida

Enclosure: Invoice #47-27

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

March 13, 1990

Dr. Ted Norris, TWS-INC7
Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear Ted:

This letter summarizes the work covered by the enclosed invoice for the months of January and February, 1990. Work covered by this invoice includes modifying and refining the chlorine-36 sample preparation process for samples that contain low chloride content. Two samples were processed using the new technique but were not analyzed due to the cancellation of the run.

We have rented an individual storage facility and moved all of the Los Alamos cuttings to that location. The cuttings will be inventoried within the next month or two. We also had a voltage adjustor made for the purposes of decreasing the output on the Dionex so it is compatible with the SpectraPhysics integrator. The water purification system, which was originally to appear on this invoice, will instead be purchased by us directly, and will not be billed to Los Alamos.

Please let me know if you have any questions concerning the work involved or charges covered by this invoice.

Sincerely,

Susan Maida

SM/jb

Enclosure: Invoice #47-24

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

January 15, 1990

Dr. Ted Norris, TWS-INC7
Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear Ted:

This letter summarizes the work covered by the enclosed invoice for the months of October through December, 1989. Work covered by this invoice includes preparation of a draft copy of the annual report for the 1989 fiscal year, modification of our own QA procedures to better comply with LANL QA, and documentation of previous work in accordance with QA guidelines.

We have also purchased an integrator and a water purification system (which will appear on the next invoice) for this project in order to improve our analytical capabilities. Full-scale laboratory operations will begin by the end of this week as laboratory modification will be finished within the next few days.

Please let me know if you have any questions concerning the work involved or charges covered by this invoice.

Sincerely,

Susan Maida

SM/jb
Enclosure: Invoice #47-24

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

20 September 1989

Dr. Ted Norris, TWS-INC7
Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear Ted:

The enclosed invoice covers charges for the month of August, 1989. Labor charges were for preparation of samples that were sent to the University of Rochester for Cl-36 analyses and for Cl analysis on the Dionex. We had to rent an integrator several months ago as our integrator was sent to the field, therefore the rental charges.

As you know, Songlin Cheng has accepted a teaching position at Wright State University. We have hired Susan Maida, a hydrologist/geochemist, to act as project hydrologist for the Los Alamos project. Harold will continue to oversee the project. Susan holds a masters degree in hydrology from the University of Arizona. Her master's thesis involved the distribution of iodide and halides in the Milk River aquifer, a regional confined system of great interest to hydrogeochemists. Her experience in chlorine-36 preparation and analysis will be of significant value to our project.

If you have any questions concerning the work covered by this invoice, please contact Alex Yiannakakis or Harold.

Sincerely,

Sherril K. Bentley

Enclosure: Invoice # 47-22

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

3 July 1989

Dr. Ted Norris
TWS-INC7, Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear Ted,

This letter summarizes the work covered by the enclosed invoices for the months of May and June, 1989. We prepared and analyzed nine samples for chlorine-36 content in May. We have analyzed 24 UZ-1 samples, 8 UZN-43 samples, and 10 trench #14 samples for chloride and bromide analyses. Each UZ-1 and UZN-43 analysis consisted of four separated size fractions. We also conducted shatter box experiments on UZ-1 samples 97-101, 250-255, and 295-300 and processed these samples for chloride and bromide analyses.

Please let me know if you have questions regarding either the invoice or the work performed.

Sincerely,

Songlin Cheng

Enclosures: Invoices 47-19 and 47-20 (copies)

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

2 May 1989

Dr. Ted Norris, TWS-INC7
Mail Stop J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear Ted:

This letter summarizes the work covered by the enclosed invoice for the month of April. We completed step-wise crushing-leaching for Cl-36 the following samples:

UZ-1 250-255 (D)
UZ-1 250-255 (E)
UZ-1 500-502 (3)
UZ-1 1020-1025 (3)
UZ-1 1195-1200 (3)

We also leached the following samples for Cl-36 analysis:

DH-1 17.3-27.3
DH-1 27.3-37.0
AC-2 44.1-54.2
AC-2 92-106

The above nine samples will be ready for Cl-36 counting this month. Please let me know if you have any questions regarding either the invoice or the work performed.

Sincerely,

Songlin Cheng

SC/mel

Enclosure: Invoice #47-18

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

17 April 1989

Dr. Ted Norris, TWS-INC7
Los Alamos National Laboratory
Mail Stop J-514
Los Alamos, N.M. 87545

Dear Ted,

This letter summarizes the work covered by the enclosed invoice for the month of March. We have sieved and leached all 25 UZ-1 samples for chloride and bromide analysis. As you know, UZ-1 samples 500-502 and 1020-1025 have been leached once before for Cl-36. These two leached samples have been dried and pulverized and are ready to be leached again for Cl-36.

Enclosed is a copy of the work plan we prepared for the current fiscal year. We will prepare a new work plan when the current work plan approaches the end. Your comments will be greatly appreciated.

Please let me know if you have questions regarding either the invoice or the work performed.

Sincerely,

Songlin Cheng

Enclosures: Invoice #47-17
Work Plan 3/24/89

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

30 December 1988

Dr. Ted Norris
Los Alamos National Laboratory
Mail Stop J-514
Los Alamos, N.M. 87545

Dear Ted,

This letter summarizes the work covered by the enclosed invoice for the month of November, 1988. We prepared three UZN-43 samples and three U12G Tunnel samples for chlorine-36 analysis. We also prepared a summary of analytical results and a sample processing procedure for chlorine-36 analysis. Please let me know if you have any questions regarding either the invoice or the work performed.

Sincerely,

Songlin Cheng

Enclosures: Invoice #47-14 (copy)

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

16 November 1988

Dr. Ted Norris
Los Alamos National Laboratory
TWS-INC7, MS J-514
Los Alamos, New Mexico 87545

Dear Ted,

This letter briefly summarizes the work completed by Hydro Geo Chem in the month of October. I will send you 1) experimental procedures and 2) summary of analytical results for the fiscal year 1988 (through October 1988) in the next few days.

During the month of October, we processed six USW UZ-1 samples for Cl-36 analysis, one of them (250-255') was processed for step-wise leaching-crushing for a total of three chlorine extractions. We also prepared two blank NaCl samples for Cl-36 analysis. Eight UZ-1 samples were washed-crushed-leached for studying the effect of pulverization on leachable chloride. We have improved our detection limit of bromide to about 10 ppb. We have leached UZ-1 samples (eight) and U12G tunnel samples (three) for Cl/Br ratios. U12G tunnel samples will be processed for Cl-36 in the month of November.

Please let me know if you have any questions regarding either this summary of work or the enclosed invoice.

Sincerely yours,

Songlin Cheng

Enclosure: Invoice (copy)

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

12 October 1988

Dr. Ted Norris, TWS-INC7
Los Alamos National Laboratory
Mail Stop J-514
Los Alamos, New Mexico 87545

Dear Ted,

The enclosed invoice covers the work performed by Hydro Geo Chem during the month of September. We analyzed the chloride profile of UZ-1 samples that is leachable only after crushing the rock powder in a shatter box. We also prepared UZ-1 97-101 and 250-255 samples for chlorine-36 analysis.

The technician labor is for designing the cooling system for the induction furnace. Please call me if you have any questions regarding this invoice or the work.

Sincerely yours,

Songlin Cheng
Project Manager

Enclosure: Invoice (copy)

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

12 August 1988

Dr. Ted Norris
Los Alamos National Laboratory
Mail Stop J-514
Los Alamos, N.M. 87545

Dear Ted,

This letter summarizes the work covered by the enclosed invoice for the month of July, 1988. We have concentrated on the UZ-1 samples and have constructed the depth profile of leachable chloride for the original UZ-1 samples listed in your letter dated April 15, 1988. We have also found that grain size distribution has significant affect on leachable chloride. In order to establish the grain size-leachable chloride relationship, we also leached aliquots of UZ-1 250-255' that have been crushed in a shatter box for time periods between 0.5 and 10 minutes. We are presently in the process of analyzing these samples.

Sincerely yours,

Songlin Cheng

Enclosure: Invoice (copy)

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

13 July 1988

Dr. Ted Norris
Los Alamos National Laboratory
Mail Stop J-514, TWS-INC7
Los Alamos, NM 87545

Dear Ted,

Enclosed is our invoice for June, 1988. Work covered by this invoice includes arranging for the installation of the transformer and phase converter for the furnace, installation of the Dionex chromatography unit and standardizing the analytical procedure, and performance of leaching experiments to standardize the leaching procedure for rock samples. The power to the furnace should be completed by the middle of this month. Please give me a call if you have any questions.

Sincerely yours,

Songlin Cheng

Enclosure: Invoice \$47-9 (copy)

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

15 June 1988

Dr. Ted Norris
TWS-INC7, MS J-514
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Dear Ted,

As you can see from the enclosed invoice, the majority of these charges are associated with equipment purchases (Dionex ion exchange chromatograph) and installation of the power line for the furnace. As I discussed recently with Tom Christopherson, the power for the furnace has gone far over what we were originally quoted. However, due to changes in design, we expect that the crucible costs will be less, therefore material charges will be closer to our quote.

Labor charges are mainly associated with meetings, ordering equipment, and investigation of experimental procedures. Work has started on water-leaching of the rock samples. Songlin appears to have developed an interesting procedure to clarify the leach waters (which are very turbid and difficult to filter). In small scale experiments, the insertion of a low voltage pair of electrodes clarifies the water remarkably quickly. I have not seen a similar procedure described in the literature. If this technique is original, it might produce a small technical paper.

We have also gotten the Dionex up and running this month and to date have run a small number of chromatograms. If you have any questions or suggestions, please don't hesitate to call me.

Yours truly,

Harold W. Bentley

Enclosure: Invoice (copy)

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

29 September 1986

Dr. A.E. Norris
Los Alamos National Laboratory
Mail Stop J-4
Los Alamos, New Mexico 87545

Dear Ted,

The enclosed invoice copy, covering charges for September, 1986, represents Harold Bentley's trip to Ohio to visit Ajax Magnethermic Corporation for the purpose of planning the details of the induction furnace. Three hours of project management time connected with planning of the induction furnace is also charged in this invoice as well as two hours for quality assurance administration.

Harold or Dan will be happy to answer any questions concerning the furnace.

Sincerely yours,

Sherril K. Bentley
Sherril K. Bentley

Enclosure: Invoice copy

HYDRO GEO CHEM, INC.

Environmental Technology & Engineering

1430 N. Sixth Avenue
Tucson, Arizona 85705
(602) 623-6981
FAX: (602) 623-6984

11 September 1986

Dr. A.E. Norris
Los Alamos National Laboratory
Mail Stop J-4
Los Alamos, New Mexico 87545

Dear Ted,

This letter summarizes the work completed by Hydro Geo Chem during the month of August. All labor charges were for quality assurance and planning. Specifically, we revised our Quality Assurance Program Plan in accordance with requests from LANL and prepared for and were involved with a quality assurance audit conducted by Ms. Suzann Dye.

A planning meeting was conducted with Gary Walter, Dan Ramey, Harold Bentley, and you concerning the status of the contract, stop-work order, and work elements of the proposed work plan. In addition, we have been planning some aspects of the induction furnace and have received an invoice from Ajax Magnethermic Corporation. We are submitting the invoice for the induction furnace separately.

If you have any questions, please call.

Sincerely yours,

Dan S. Ramey

Enclosures: Invoices (2)

APPENDIX B:

Publications

³⁶Cl studies of water movements deep within unsaturated tuffs

A.E. Norris

Los Alamos National Laboratory, Los Alamos, NM 87545, USA

H.W. Bentley and S. Cheng *

Hydro Geo Chem, Incorporated, 1430 North Sixth Avenue, Tucson, AZ 85705, USA

P.W. Kubik, P. Sharma and H.E. Gove

Nuclear Structure Research Laboratory, University of Rochester, Rochester, NY 14627, USA

Measurements of ³⁶Cl in cuttings from a borehole that was drilled 387 m into unsaturated tuffs indicate the possible detection of significant radioactive decay of cosmogenic ³⁶Cl in two of the samples. However, the ³⁶Cl/Cl ratio was found to vary with the amount of pulverization of the cuttings. Work is in progress to separate the ³⁶Cl/Cl data into cosmogenic and in situ components. The cosmogenic component will be used to trace very slow water movements through the unsaturated zone. Bomb pulse ³⁶Cl was observed as deep as 153 m, and this identification is not constrained by the problem with pulverization. This work shows the efficacy of ³⁶Cl measurements for detecting modern water movements deep in the unsaturated zone.

1. Introduction

Characterization of water movements in unsaturated zone tuffs at Yucca Mountain, Nevada, is important for predicting the water-borne transport of radioactive nuclides to the accessible environment from a potential nuclear waste repository. The information available prior to the start of this study was compiled by Montazer and Wilson [1]. Data came from laboratory measurements of effective permeabilities utilizing cores from Yucca Mountain boreholes and from measurements in boreholes of geothermal heat flux, hydraulic gradients and effective permeabilities. The data indicated that the downward flux of water through the potential repository host rock, the moderately to densely welded portion of the Topopah Spring Member of the Paintbrush Tuff formation, may range from 1×10^{-7} to 0.2 mm/yr. The geothermal heat flux data indicated that the net hydrologic flux may be upward because of vapor phase transport. Measurements of ³⁶Cl have been undertaken to supplement the information about water movements in the unsaturated zone derived from conventional techniques.

The principle of this study derives from the conservative properties of chlorides to trace hydrologic

water movements in the liquid phase [2]. Chlorides have been deposited on the arid land at Yucca Mountain by winds that have lofted sea spray into the troposphere. Then rainfall and melted snow have dissolved some of this environmental chloride and transported it into the unsaturated zone. Some of the chlorine in these chlorides is cosmogenic ³⁶Cl. The radioactive decay of ³⁶Cl, with its 3×10^5 yr half-life, serves as the timing signal for the rate of water percolation through the unsaturated tuffs. The water need not move downward uniformly for this measurement to be meaningful. For example, a pulse of water could dissolve some chloride, carry the solute some distance underground, then evaporate. The nonvolatile chloride will remain where the water deposited it until moved to a different location by another pulse of water. The cosmogenic ³⁶Cl measured in a particular sample may result from multiple pulses of water over a period of time, in which case the data would be interpreted in terms of water movement somewhat faster than that of the chloride deposited by the original pulse, but somewhat slower than that of the solute in the last pulse. The rate of solute transport derived from the ³⁶Cl data, even though it may be an average value, will be useful for calculating transport rates for water-borne nuclides such as ⁹⁹TcO₄⁻ and ¹²⁹I⁻, which are likely to be transported no faster than the average rate derived from the ³⁶Cl data.

The goal of the work reported below is to observe a decrease in the ³⁶Cl/Cl ratio as a function of sample

* Current address: Department of Geological Sciences, Wright State University, Dayton, OH 45435, USA.

AIM

Association for Information and Image Management

1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910

301/587-8202

Centimeter

Inches

MANUFACTURED TO AIM STANDARDS
BY APPLIED IMAGE, INC.

2

of

2

Fig. 1. Map of the Nevada Test Site with the locations of borehole USW UZ-1 and G-Tunnel indicated.

depth, where the decrease in the ratio results from the radioactive decay of cosmogenic ^{36}Cl . The cosmogenic $^{36}\text{Cl}/\text{Cl}$ ratio at the surface of Yucca Mountain was determined to be $(519 \pm 53) \times 10^{-15}$ in a previously published study of infiltration at this site [3]. Obtaining appropriate samples for these analyses has proved to be a problem. First, the solubility of meteoric chloride in water limited the available samples to those from dry-drilled boreholes. Second, the procedure developed for leaching chloride from tuffs at Yucca Mountain that

typically contain 2 ppm Cl^- required the use of ~ 40 kg of material to obtain a sample containing ~ 20 mg Cl^- for the accelerator mass spectrometry analysis. The only dry-drilled borehole from which cuttings in sufficient quantity had been saved to permit the start of this study was USW UZ-1. Its geographical coordinates are $36^\circ 51' \text{N}$ and $116^\circ 27' \text{W}$. Fig. 1 is a map showing the location of USW UZ-1 relative to the proposed repository at Yucca Mountain.

The remainder of this paper presents the results of ^{36}Cl analyses of samples from USW UZ-1, a discussion of the dependence of the measured $^{36}\text{Cl}/\text{Cl}$ ratio on the pulverization of the cuttings, corroborating evidence for the detection of bomb pulse ^{36}Cl deep underground, and an indication of potential applications of the results of this study.

2. Results

Samples for ^{36}Cl analyses were prepared from USW UZ-1 cuttings that were collected at nine depths as the borehole was drilled. The cuttings were leached with chloride-free water, using the same procedure as that for the ^{36}Cl sample preparation in the Yucca Mountain infiltration study [3]. The cuttings from seven depth intervals were sieved into four fractions before leaching. The sieves had mesh sizes of 35, 60 and 230. The chloride contents of the tuffs were measured in each fraction. The results are shown in table 1. The chlorides from all four fractions for each depth interval were combined to yield one AgCl sample for ^{36}Cl analysis.

The $^{36}\text{Cl}/\text{Cl}$ ratios in the nine samples were measured at the University of Rochester's accelerator mass spectrometry facility, together with a reagent blank, as in the previous study [3]. The measured ratios were normalized to standards prepared by diluting standard reference material #4943 from the National Institute of Standards and Technology [4]. The background correc-

Table 1
Results of ^{36}Cl Analyses of USW UZ-1 cuttings

Depth interval below surface [m]	mg Cl/kg tuff in sieve fractions				$^{36}\text{Cl}/\text{Cl}$ ($\times 10^{15}$)
	> 35	35–60	60–230	< 230	
30–31	28.3	37.7	44.7	55.4	11400 ± 360
52–54					2498 ± 198
76–78	22.6	25.0	25.7	30.5	436 ± 25
120–122					390 ± 48
150–152	5.2	5.4	5.1	6.9	403 ± 42
152–153	8.7	10.0	10.3	7.3	2046 ± 103
311–312	1.2	2.3	2.3	2.9	245 ± 38
364–366	2.6	2.9	2.3	3.0	454 ± 61
372–373	3.2	3.9	3.7	5.4	102 ± 11

tion was made by using samples prepared from Weeks Island halite with typical ³⁶Cl/Cl ratios of (0.5 to 2) × 10⁻¹⁵ [5]. The results of the ³⁶Cl analyses are shown in table 1. The standard deviations listed in the table are the ±1σ values based only on the ³⁶Cl counting statistics.

A second set of ³⁶Cl measurements was undertaken to determine whether the measured ³⁶Cl/Cl ratios depend on the size of the particles that are leached. The cuttings from the 76 to 78 m interval that had been used for the ³⁶Cl/Cl measurement shown in table 1 were washed, dried, pulverized for five minutes in a shatter box, then leached again for another ³⁶Cl analysis. The cuttings from this leaching were washed, dried, pulverized for an additional five minutes and leached again, then this sequence was repeated to produce a total of four more AgCl samples from the original cuttings. The results of the ³⁶Cl analyses are shown in table 2, together with the ³⁶Cl result from table 1. Fifty milligrams of chloride from ³⁶Cl-free NaCl was added to each of the first two leachates in this sequence to ensure adequate chloride recovery. The pulverization was observed to result in the leaching of additional chloride from the previously leached cuttings, so carrier was not added to the last two leachates in this sequence.

The data for the 76 to 78 m depth interval in table 2 display a strong dependence of the measured ³⁶Cl/Cl ratio on the amount of pulverization. The last two values in this sequence are statistically the same, which indicates that an asymptotic value may have been observed.

The cuttings from three other depth intervals were pulverized to see how general the dependence of the ³⁶Cl/Cl ratio on pulverization might be. The data in table 2 show a decrease following pulverization in each case, but the decrease is not a constant fraction of the original value, even though the pulverization time was the same in each of these three cases. The data, how-

ever, may be useful for determining the meteoric ³⁶Cl component in an end-member analysis.

3. Discussion

The purpose of this work was to obtain evidence from ³⁶Cl data concerning the percolation flux of water through the unsaturated tuffs at Yucca Mountain. The data in table 1 at the depths of 311 to 312 m and 372 to 373 m indicate the possibility of significant ³⁶Cl radioactive decay at these depths. Had the ³⁶Cl/Cl ratios been similar to the 519 × 10⁻¹⁵ ratio at the surface, further work to develop this technique most likely would not have been justified.

The ratio observed at 364 to 366 m statistically is the same as the 519 × 10⁻¹⁵ ³⁶Cl/Cl ratio observed at the surface. A brief description of the manner in which USW UZ-1 was drilled and why the drilling was terminated should show that the ³⁶Cl ratio measured at this interval can represent a valid indication of the presence of contemporary water (i.e., less than 10⁵ years for these ³⁶Cl measurements) without invalidating the results exemplified by the ³⁶Cl data from the 311 to 312 m and the 372 to 373 m intervals. Whitfield [6] has summarized the stratigraphy and the results of the US Geological Survey's initial hydrologic measurements at USW UZ-1. The information in the rest of this paragraph comes from that reference. Borehole USW UZ-1 was drilled through 17.7 m of alluvium and 369 m of densely welded to nonwelded and bedded tuffs of the Paintbrush Tuff formation. The lithology of the Topopah Spring Member, which extends from 78 m to the bottom of the borehole, is relatively simple below 100 m, with moderately welded tuff to a depth of 221 m and densely welded tuff below 221 m. The first 17.7 m of USW UZ-1 was drilled using 4573 l of water. Sufficient water accumulated at the drill bit so that it interfered with the drilling operations. A diaphragm pump delivered 1514 l of water to the surface, but 3059 l were lost to the formation. The remainder of the hole was drilled using air to cool the drilling bit. Data from drill cuttings and from borehole instrumentation indicated that the water not recovered from the first 17.7 m did not seep below 76 m. The drill hole was planned to terminate in the saturated zone, which starts at a depth of 470 m at the USW UZ-1 location. Drilling actually was discontinued at 387 m, where a large volume of water was encountered. The water was found to contain a chemical polymer with a composition identical to that used in drilling hole USW G-1, which is located 305 m southeast (downdip) of USW UZ-1. Approximately 8.7 × 10⁶ l of polymer drilling fluid was not recovered during the drilling of USW G-1. Whitfield interprets the data in terms of a major fracture zone that provided a hydrological connection between the two holes or a

Table 2
Decrease of ³⁶Cl/Cl ratio with pulverization time

Depth interval below surface [m]	Shatterbox time [min]	Added chloride [mg]	³⁶ Cl/Cl (× 10 ¹⁵)	% of original ratio
76-78	0	0	436 ± 25	
	5	50	193 ± 7	44
	10	50	91 ± 4	21
	15	0	36 ± 11	8
	20	0	25 ± 5	6
152-153	0	50	2046 ± 103	
	3	0	1885 ± 150	92
311-312	0	50	245 ± 38	
	3	0	159 ± 12	65
364-366	0	50	454 ± 61	
	3	0	340 ± 15	75

naturally occurring perched-water zone that is contaminated with the USW G-1 drilling polymer. If a major fracture zone does connect the two holes, then the ³⁶Cl data observed at the 364 to 366 m interval may indicate that water used to drill USW G-1 reached this interval, as well as the 387 m depth where USW UZ-1 was terminated.

The strong dependence of the measured ³⁶Cl/Cl ratio on the amount of pulverization, as shown by the data in table 2, resulted in additional analyses to separate the ³⁶Cl/Cl ratios shown in table 1 into two components. One component would represent meteoric chloride containing cosmogenic ³⁶Cl, while the other would represent chloride associated with the tuff that included ³⁶Cl formed in situ by neutron activation of ³⁵Cl. The neutrons necessary for the second component are generated in the decay of uranium and thorium series elements in the tuffs, both from spontaneous fission and from (α , n) reactions on light elements that are present in trace quantities [2].

The last two ³⁶Cl/Cl ratios shown in table 2 for the 76 to 78 m depth interval may represent the end member for the in situ component, if they prove to be asymptotic values. Bromide concentrations in many samples of cuttings were measured to determine whether the Cl/Br ratio could be used with the ³⁶Cl/Cl ratio to identify the meteoric and the in situ members in each sample. The Cl/Br ratio for the meteoric component appeared to be ~130, and the ratio for the in situ component appeared to be ~500. However, no consistent pattern for the meteoric component has been obtained with the data currently available.

The unanticipated finding in this work is the observation of bomb pulse ³⁶Cl at a depth as great as 153 m. The origin of the bomb pulse is global fallout from high-yield nuclear device testing between 1952 and 1962 that caused ³⁵Cl in seawater to be activated with neutrons and subsequently injected into the stratosphere. The characteristic that permits a ³⁶Cl/Cl ratio to be identified as originating from the bomb pulse is a value significantly greater than the 519×10^{-15} cosmogenic ratio. Ratios of ³⁶Cl/Cl higher than the cosmogenic ratio were observed in saline waters from the Stripa mine in Sweden [7], but these were explained on the basis of in situ neutron irradiation of ³⁵Cl. The Yucca Mountain tuffs cannot reasonably be expected to generate high ³⁶Cl/Cl ratios, as at Stripa, because concentrations of chlorine, thorium and uranium are not high and these trace elements appear to be distributed relatively homogeneously. The geochemical gradients reported by two laboratories [8,9], based on measurements of samples from 166 locations, show chlorine generally below neutron activation analysis detection limits of ~100 ppm (except in the USW G-1 interval between 402 and 424 m), thorium contents of 20.8 ± 5.3 ppm and uranium contents of 3.7 ± 1.1 ppm. The thorium and uranium

data contain a few points approximately a factor of two or less either above or below the mean elemental contents. There is no evidence for a combination of high values all at one location that might contribute to a high ³⁶Cl/Cl ratio from in situ production processes. Consequently, the only reasonable source of the high ³⁶Cl/Cl ratios observed in this work is bomb pulse ³⁶Cl.

The data in table 1 show that bomb pulse ³⁶Cl is present in cuttings from the intervals at 30 to 31 m, 52 to 54 m, and 152 to 153 m. The ³⁶Cl/Cl ratios for these three intervals may represent lower limits, because of the dependence of the ³⁶Cl/Cl ratio on pulverization, which was noted above. Also, bomb pulse ³⁶Cl may be present in cuttings from other intervals, but unrecognizable in samples with ³⁶Cl/Cl ratios of 519×10^{-15} or lower. The water that transported the bomb pulse ³⁶Cl from the surface to the depths where the bomb pulse is observed in cuttings collected in 1983 must have been younger than 30 years.

The pathways by which the ³⁶Cl bomb pulse was transported to the locations where it is observed should be determined to help understand water flow in unsaturated tuffs. Our previous work near this location [3] showed a ³⁶Cl bomb pulse profile that reached a maximum about 0.5 m below the surface, with no evidence for the bomb pulse below 2 m. Observation of the bomb pulse at significantly greater depths at the USW UZ-1 location could have resulted from the use of water to drill the first 17.7 m. The water used for this drilling came from well J-13. A sample of water from well J-13 was collected around the time of the drilling operations and analyzed for ³⁶Cl. The ³⁶Cl/Cl ratio was measured to be $(531 \pm 41) \times 10^{-15}$. Thus, the drilling water could have washed the bomb pulse down from the alluvium near the surface. However, the large volume of water that was used (4573 l) had practically no contact with the top two meters of alluvium. It is more likely that this volume of water would tend to dilute any entrained bomb pulse to undetectable levels. The absence of a distinct ³⁶Cl bomb pulse at three intervals above that at 152 to 153 m, including the interval at 150 to 152 m, indicates that lateral water flow is involved in the transport of the bomb pulse, but some downward flow is possible. The profile of the bomb pulse below 153 m could not be determined, because the next interval where cuttings were available was at 183 m. If lateral water flow did transport the ³⁶Cl bomb pulse to the locations where it has been detected, then cuttings from a borehole drilled without water at a location updip from USW UZ-1 also should show the presence of the bomb pulse deep in the unsaturated zone.

Recently reported analyses of tritium performed by the US Geological Survey [10] support the interpretation of the ³⁶Cl data in terms of lateral water flow. The tritium was measured in moisture from cores collected at two Yucca Mountain locations, UE-25 UZ#4 and

UE-25 UZ #5. High tritium values, indicating the presence of modern water, were found to decrease rapidly to a low value at 10 to 12 m in UE-25 UZ #4. Then spikes of high tritium were observed at 25 m and at 46 to 50 m in this same borehole and at 28 m and at 34 m in UE-25 UZ #5. The tritium spikes were taken to indicate recent water movement in fractures or bedded units in the unsaturated tuffs at Yucca Mountain. Another observation from this work is that pulses of water penetrate only to a depth of ~ 10 m. Additional work in which tritium and bomb pulse ³⁶Cl were measured at the same depths in the same hole would provide incontrovertible evidence for the presence of modern water.

Additional evidence has been obtained for the presence of bomb pulse ³⁶Cl deep in unsaturated tuffs. Cuttings from dry drilling operations conducted in the Nevada Test Site's G-Tunnel, located ~ 45 km northeast of Yucca Mountain (see fig. 1) were analyzed for ³⁶Cl. The results are shown in table 3. The horizontal distances shown in the table are relative to a vertical fault visible in the welded, unsaturated Grouse Canyon tuff in a drift near some of the drilling. The fault is not thought to go to the surface, which is some 400 m overhead. The first two ³⁶Cl measurements with cuttings from G-Tunnel were undertaken with the expectation that the ³⁶Cl/Cl ratios would be significantly less than 519×10^{-15} . Observation of the ³⁶Cl bomb pulse in one of the two samples was a complete surprise. Additional measurements have confirmed the presence of the bomb pulse, as is evident in table 3. The high ³⁶Cl values appear to be nearly symmetric in a northeast-southwest plane that is centered around the fault near borehole DH-1. Additional drilling was planned to define the ³⁶Cl bomb pulse in three dimensions, but the US Department of Energy closed G-Tunnel to all further work in December, 1989, before this additional drilling could be started. These G-Tunnel data confirm the observations from the USW UZ-1 cuttings that the ³⁶Cl bomb pulse can be detected deep within unsaturated zone

tuffs. The G-Tunnel data are insufficient to define the transport path or paths to the location where the bomb pulse is observed. The two lowest ³⁶Cl/Cl ratios are significantly lower than the cosmogenic ratio. They provide additional evidence that the radioactive decay of cosmogenic ³⁶Cl may serve to trace the percolation of water through unsaturated zones over long times.

4. Applications

The determination of the meteoric component of the ³⁶Cl/Cl ratio in unsaturated zone water flow, if accomplished successfully, combined with the observation of the ³⁶Cl bomb pulse deep within the unsaturated zone, offers the potential for discriminating between matrix flow and fracture flow in a complex hydrologic regime. This information is important for calculating the future performance of a nuclear waste repository located in the unsaturated zone.

The unambiguous identification of bomb pulse ³⁶Cl deep underground establishes a new technique for detecting where modern water has flowed in hydrologic studies of unsaturated zones. The most likely pathways for such flow is through faults and fractures. Evidence for the presence of modern water wherever it occurs in the unsaturated zone is important not only for studies pertaining to high-level nuclear waste disposal, such as this one, but also for low-level nuclear waste and hazardous waste sites, which frequently are underlain by unsaturated zones. Measurements of tritium in conjunction with bomb pulse ³⁶Cl are desirable for identifying the presence of modern water deep within unsaturated media.

5. Summary

The original purpose of this work was to determine fluxes of water percolating through unsaturated zone tuffs as traced by the radioactive decay of ³⁶Cl. The USW UZ-1 data indicate that the decay of cosmogenic ³⁶Cl may have been detected, but the observed dependence of the measured ³⁶Cl/Cl ratios in cuttings on the amount of pulverization requires additional work to separate the meteoric component of the ³⁶Cl/Cl ratio from the in situ ratio.

The unexpected finding in this work was the observation of bomb pulse ³⁶Cl as deep as 152 to 153 m below the ground surface. The absence of recognizable bomb pulse ³⁶Cl between 250 and 500 m is interpreted as evidence for lateral water flow, in accord with the interpretation provided by the US Geological Survey of tritium data from two nearby boreholes. Confirmation that the ³⁶Cl bomb pulse can be detected deep under-

Table 3
Results of ³⁶Cl analyses of G-Tunnel cuttings

Sample	Distance from fault [m]	Direction from fault	³⁶ Cl/Cl ($\times 10^{15}$)
AC-2	70.9-73.9	northeast	306 $\pm$ 22
AC-2	55.0-57.6	northeast	845 $\pm$ 76
DH-1	5.3	southwest	1539 $\pm$ 101
DH-1	5.3	southwest	1964 $\pm$ 75
DH-2	7.8	southwest	1709 $\pm$ 70
AC-1	43.1-44.8	southwest	1243 $\pm$ 87
AC-1	44.8-47.4	southwest	3044 $\pm$ 360
AC-1	56.0-58.3	southwest	412 $\pm$ 18

ground is provided by data from G-Tunnel, about 45 km away, where the bomb pulse was observed in samples from borehole cuttings that were collected ~ 400 m underground. Applications of this work include the potential for discriminating between matrix and fracture flow in the unsaturated zone and for detecting recent water movements as traced by the ³⁶Cl bomb pulse at hazardous materials disposal sites.

Acknowledgements

This work was funded by the Yucca Mountain Project of the US Department of Energy. The ³⁶Cl measurements at the University of Rochester were funded in part by the National Science Foundation. The USW UZ-1 cuttings were kindly provided by the US Geological Survey, through the assistance of M.P. Chornack and M.S. Whitfield. Measurements of ³⁶Cl in the first two samples of USW UZ-1 cuttings and of ³⁶Cl in well J-13 water were made by S.K. Gifford, while an employee of Hydro Geo Chem, and the final ³⁶Cl samples were prepared by another Hydro Geo Chem employee, A. Yiannakakis. R.J.D. Teng and S. Tullai-Fitzpatrick helped in the accelerator mass spectrometric measurements of ³⁶Cl.

References

- [1] P. Montazer and W.E. Wilson, US Geological Survey Report USGS-WRI-84-4345.
- [2] H.W. Bentley, F.M. Phillips and S.N. Davis, in: *Handbook of Environmental Isotope Geochemistry*, vol. 2B, eds. P. Fritz and J.-Ch. Fontes, (Elsevier, Amsterdam, 1986) p. 427.
- [3] A.E. Norris, K. Wolfsberg, S.K. Gifford, H.W. Bentley and D. Elmore, *Nucl. Instr. and Meth.* B29 (1987) 376.
- [4] P. Sharma, P.W. Kubik, U. Fehn, H.E. Gove, K. Nishizumi and D. Elmore, these Proceedings (AMS 5) *Nucl. Instr. and Meth.* B52 (1990) 410.
- [5] P.W. Kubik, P. Sharma, R.T.D. Teng, S. Tullai-Fitzpatrick, S. Datar, U. Fehn, H.E. Gove and D. Elmore, *ibid.*, p. 238.
- [6] M.S. Whitfield, *Proc. NWWA Conf. on Characterization and Monitoring of the Vadose (Unsaturated) Zone* (Nat. Water Well Assoc., Worthington, Ohio, 1985) p. 413.
- [7] J.N. Andrews, J.-Ch. Fontes, J.-L. Michelot and D. Elmore, *Earth Planet. Sci. Lett.* 77 (1986) 49.
- [8] D.E. Broxton, R.G. Warren, R.C. Hagan and G. Luedemann, Los Alamos National Laboratory Report LA-10802-MS (1986).
- [9] B.C. Schuraytz, T.A. Vogel and L.W. Younker, Lawrence Livermore National Laboratory Report UCRL-53698 (1986).
- [10] US Department of Energy/Yucca Mountain Project Study Plan 8.3.1.2.2.7, Rev. 0 (1989).

APPENDIX C:

Sample Inventory

SAMPLE	DEPTH (FT)	VOLUME (IN ³)	WEIGHT (LBS)	DESCRIPTION
UZ-1	72-79	727.1	29	cuttings, fist sized pieces (8"x3"), originally were waxed
UZ-1	72-74	727.1	39	40% cuttings, 60% sand
UZ-1	72-79	1661.9	80+	50% cuttings, 50% gritty silt
UZ-1	97-101	311.6	17	90% cuttings (1"-3"), 10% sand
UZ-1	97-101	207.7	27	5 waxed samples (4"-6")
UZ-1	97-101	1038.7	55	60% cuttings, 40% sand
UZ-1	250-255	727.1	37	gritty silt, lid cracked during opening
UZ-1	250-252	779.0	41	gritty silt, "debris from junk basket"
UZ-1	295-300	1921.5	80+	90% dark sand, 10% gravel
UZ-1	347-350	1921.5	80+	95% very gritty silt, 5% gravel
UZ-1	350-352	467.4	37	30% cuttings, 70% silt, top of container shattered
UZ-1	350-352	675.1	54	30% cuttings, 70% gritty silt, "top portion of core in junk basket"
UZ-1	395-406	363.5	40	sandy silt
UZ-1	445-450	1973.5	80+	95% very gritty silt, 5% gravel
UZ-1	495-500	1765.7	80+	gritty silt, top of container split but waxed shut
UZ-1	500-502	-----	18	several fist sized rocks
UZ-1	545-550	1973.5	80+	95% very gritty silt, 5% gravel
UZ-1	595-599	1765.7	80+	gritty silt
UZ-1	695-700	1973.5	80+	gritty silt with some gravel, top of container split
UZ-1	715-720	830.9	53	gritty sand
UZ-1	795-800	1973.5	80+	gritty silt
UZ-1	830-835	727.1	57	gritty silt, bottom of container broken
UZ-1	845-850	1661.9	80+	gritty silt
UZ-1	895-900	1973.5	80+	gritty silt
UZ-1	920-925	779.0	55	sandy silt
UZ-1	945-950	1817.7	80+	gritty silt
UZ-1	995-1000	1038.7	65	gritty silt, 80% in bags of about 10 lbs. each
UZ-1	995-1000	2077.3	80+	gritty silt, 80% in bags of about 10 lbs. each
UZ-1	1020-1025	51.9	10	gritty silt
UZ-1	1020-1025	830.9	67	gritty silt
UZ-1	1045-1050	1713.8	80+	gritty silt
UZ-1	1095-1100	882.9	73	gritty silt
UZ-1	1095-1100	1090.6	75	gritty silt
UZ-1	1147-1156	1765.7	80+	gritty silt, top of container split but waxed shut
UZ-1	1170-1175	830.9	52	gritty silt
UZ-1	1195-1200	415.5	41	gritty silt
UZ-1	1220-1225	207.7	26	gritty silt
UZ-1	1245-1250	1973.5	80+	gritty silt, "bulk cuttings"
UZ-1	1258-1260	623.2	49	70% cuttings, 30% sand
UZ-1	unknown	311.6	22	80% gritty silt with one large cutting
UZ-1	unknown	1246.4	66	gritty silt
UZ-1	unknown	1765.7	80+	very gritty silt
UZ-1	unknown	1973.5	80+	gritty silt

SAMPLE	DEPTH (FT)	VOLUME (IN ³)	WEIGHT (LBS)	DESCRIPTION
UZ-6	50	381.7	28	95% sand and gravel
UZ-6	100	339.3	53	60% sand and gravel, 40% very gritty silt
UZ-6	150	339.3	27	95% sand and gravel
UZ-6	200	424.1	27	rock cuttings, 30% gravel, 70% sand
UZ-6	250	367.6	29	sandy silt
UZ-6	300	367.6	28	25% sand, 75% gritty silt
UZ-6	350	311.0	24	very gritty silt, 1% gravel
UZ-6	400	197.9	11	40% sand and gravel, 60% very gritty silt
UZ-6	455	339.3	24	very gritty silt, 1% gravel
UZ-6	500	395.8	28	10% gravel, 40% sand, 50% very gritty silt
UZ-6	550	325.1	24	very gritty silt, 20 % sand
UZ-6	600	353.4	22	sandy silt
UZ-6	650	381.7	26	very gritty silt, 10% sand and pebbles
UZ-6	700	480.6	27	gritty sandy silt, 10% sand
UZ-6	750	367.6	25	gritty silt
UZ-6	800	311.0	21	gritty silt
UZ-6	850	339.3	25	40% sand, 60% gritty silt
UZ-6	900	353.4	26	80% gritty silt, 20% sand and gravel
UZ-6	950	353.4	26	80% gritty silt, 20% sand and gravel
UZ-6	1000	367.6	25	80% gritty silt, 20% sand and gravel
UZ-6	1050	367.6	28	80% gritty silt, 20% sand and gravel
UZ-6	1100	339.3	23	gritty silt
UZ-6	1150	381.7	27	gritty silt, 10% sand
UZ-6	1200	395.8	25	gritty silt
UZ-6	1250	325.1	24	gritty sandy silt, 10 % sand
UZ-6	1300	311.0	22	very gritty silt
UZ-6	1350	339.3	22	very gritty silt
UZ-6	1400	367.6	25	very gritty silt, 1% gravel
UZ-6	1450	311.0	21	95% very gritty silt, 5% sand and gravel
UZ-6	1500	367.6	29	gritty silt, 1% sand
UZ-6	1550	339.3	21	gritty silt
UZ-6	1600	367.6	22	gritty silt
UZ-6	1650	353.4	23	gritty silt
UZ-6	1700	339.3	21	gritty silt
UZ-6	1750	339.3	22	gritty silt
UZ-6	1800	339.3	24	very gritty silt
UZ-6	1850	325.1	20	gritty silt, 1% sand

SAMPLE	DATE COLLECTED	DEPTH (FT)	WEIGHT (LBS)	DESCRIPTION
SB1	10-30-89, 11:27	50	2.5	core
SB1	10-30-89, 13:25	65	3	core
SB1	10-30-89, 13:18	70	3	core
SB1	10-31-89, 09:01	80	2	sand
SB1	10-31-89, 09:29	85	2.5	sand
SB1	10-31-89, 10:48	95	2	core
SB2	10-30-89	15	1	
SB2	10-30-89	20	1	
SB2	10-30-89	25	1	
SB2	10-30-89	27.5	1	
SB3	10-31-89, 15:01	10	2	small clods
SB4	11-01-89, 08:30	10	2.5	core
SB4	11-01-89, 08:52	20	2	sand
SB4	11-01-89, 09:06	25	1.5	sand and clods
SB4	11-01-89, 09:20	30	2	core
SB5	11-01-89, 11:01	10	2	core
SB5	11-01-89, 11:26	15	2	sand
SB5	11-01-89, 11:38	20	1	clods
SB6	11-01-89, 13:26	20	1.5	clods

SAMPLE	DEPTH (FT)	WEIGHT (LBS)	DESCRIPTION
SR2	55-60	3.5	pebbles and sand
SR2	60-65	3.5	pebbles and sand
SR2	60-65	3	pebbles and sand
SR2	65-70	3.5	pebbles and sand
SR2	70-75	1	pebbles and sand
SR2	75-80	1	clods
SR2	80-85	3	pebbles and sand
SR2	85-90	3.5	pebbles and sand
SR2	90-95	3	pebbles and sand
SR2	95-105	1	pebbles and sand
SR2	100-105	1	sandy clods
SR2	105-110	1	clods
SR2	110-115	1	clods
SR2	115-120	1	clods
SR5	55-60	4	
SR5	60-65	4	
SR5	50-55	4	
SR5	65-70	4	
SR5	70-75	4	
SR5	75-80	4	
SR5	80-85	4	
SR5	85-90	4.5	
SR5	90-95	4	
SR5	95-99	3.5	
SR5	99-103	4	
KBRG	100-105	2	sandy clods
KBRG	105-110	2	sandy clods
KBRG	110'3"-110'6"	1	brown clods
KBRG	110'9"-111'	1	white and tan sand
KBRG	110-115	1.5	sandy clods
KBRG	123-130	1.5	sand and pebbles
KBMW2	30'9"-31'	1	orange tan small pebbles

SAMPLE	DEPTH (FT)	WEIGHT (LBS)	DESCRIPTION
NW1	0-5	3	brown sand
NW1	5-10	3	sand, gravel and small stones
NW1	25-30	3	brown sand
NW1	30-35	3	brown gravel
NW1	35-40	3	brown sand
NW1	40-45	2.5	clay clods
NW1	45-50	3.5	brown sand and pebbles
NW1	50-55	3	sand and gravel (bag broken)
NW1	55-60	4	one brown clod
NW1	60-65	3	tan sand and pebbles
NW1	70-75	3	tan sand and pebbles
NW1	75-80	3	tan sand and pebbles
NW1	120-125	3	sand and clods
NW3	0-5	3	
NW3	5-10	3	
NW3	10-15	3	
NW3	75-80	6	
NW3	80-85	3	
NW3	85-90	3	
NW3	90-95	3	
NW3	100-105	3	
NW3	105-110	3	
NW3	110-115	3	
NW3	115-120	3.5	
NW3	120-125	3	
NW3	120-131.5	3	
NW3	125-129	3	

SAMPLE	DEPTH (FT)	WEIGHT (LBS)	SAMPLE	DEPTH (FT)	WEIGHT (LBS)
NW4	0-5	1	W1	0-5	3
NW4	0-5	2	W1	5-10	3
NW4	5-10	3	W1	10-15	3
NW4	5-10	3	W1	15-20	3
NW4	10-15	3	W1	15-20	3.5
NW4	10-15	3	W1	20-25	3
NW4	15-20	3.5	W1	25-30	4
NW4	15-20	4	W1	30-35	4
NW4	20-25	4.5	W1	35-40	3
NW4	20-25	3	W1	40-45	3
NW4	25-30	3	W1	45-50	4
NW4	25-30	3.5	W1	50-55	4.5
NW4	30-35	3	W1	55-60	4
NW4	30-35	3	W1	60-65	4
NW4	35-40	4	W1	65-70	4
NW4	35-40	3.5	W1	70-75	4
NW4	40-45	4	W1	75-80	4
NW4	45-50	5	W1	80-85	3
NW4	50-55	3	W1	85-90	4
NW4	55-60	4.5	W1	90-95	4
NW4	60-65	5	W1	95-10	4
NW4	60-65	5	W1	100-105	4
NW4	65-70	3	W1	105-110	4
NW4	65-70	4			
NW4	70-75	3	W2	0-5	4
NW4	70-75	4	W2	5-10	3.5
NW4	75-80	3	W2	10-15	4
NW4	80-85	3	W2	15-20	3.5
NW4	80-85	4.5	W2	20-25	4
NW4	85-90	3.5	W2	25-30	3.5
NW4	85-90	6	W2	30-35	4
NW4	90-95	3	W2	35-40	3.5
NW4	90-95	4	W2	40-45	4
NW4	95-100	4.5	W2	45-50	3.5
NW4	100-105	4	W2	50-55	3
NW4	100-105	3.5	W2	55-60	4
NW4	105-110	4	W2	80-85	4
NW4	110-115	4.5	W2	85-90	4
NW4	115-120	4.5	W2	90-95	4.5
NW4	120-125	4.5			

SAMPLE	DEPTH (FT)	WEIGHT (LBS)	DESCRIPTION
NW5	60-65	5	sand and pebbles
NW5	65-70	6	sand, gravel, and stones
NW5	75-80	5	sand, gravel, and stones
NW5	75-80	6	sand, gravel, and stones
NW5	80-85	5	sand, gravel, and stones
NW5	85-90	4	sand, gravel, and pebbles
NW5	90-95	5	sand, gravel, and stonesclods
NW5	95-100	6	sand and pebbles
NW5	100-105	5	tan clods
FD2	0-5	1	very fine sand
FD2	5-10	1.5	fine sand with pebbles
FD2	10-15	1.5	fine sandy soil
FD2	15-20	1.5	fine sandy soil
FD2	20-25	1	brown sandy soil
FD2	25-30	1.5	damp, sandy soil
FD2	30-35	1.5	medium sand
FD2	35-40	2	coarse sand
FD2	40-45	2	coarse sand with pebbles
FD2	45-50	1.5	medium coarse sand with stones
FD2	50-55	1.5	medium sand
FD2	55-60	1	coarse sand
FD2	60-65	1.5	damp, coarse sand
FD2	65-70	1.5	damp, coarse sand
FD2	70-75	3	
FD2	75-80	4	
FD2	80-85	4	
FD2	85-90	3.5	
FD2	90-95	3	
FD2	95-100	4	
FD2	100-105	4	
FD2	105-110	4	
FD2	110-115	4	
FD2	115-120	4	

SAMPLE	DEPTH (FT)	WEIGHT (LBS)	SAMPLE	DEPTH (FT)	WEIGHT (LBS)
LE2	0-5	3.5	SR4	0-5	4.5
LE2	5-10	3.5	SR4	5-10	4.5
LE2	10-15	4	SR4	10-15	6
LE2	15-20	4	SR4	15-20	4
LE2	20-25	3.5	SR4	20-25	6
LE2	25-30	4	SR4	25-30	6
LE2	30-35	4.5	SR4	30-35	5.5
LE2	35-40	4	SR4	35-40	4
LE2	40-45	4	SR4	40-45	5
LE2	45-50	3	SR4	45-50	4.5
LE2	50-55	3	SR4	50-55	4.5
LE2	55-60	3.5	SR4	55-60	4
LE2	60-65	3.5	SR4	60-65	5
LE2	65-70	4	SR4	70-75	4
LE2	70-75	2.5	SR4	75-80	4
LE2	75-80	2	SR4	80-85	4
LE2	85-90	4	SR4	85-90	4
LE2	90-95	1	SR4	90-95	5
LE2	95-100	2	SR4	95-100	4.5
LE2	100-105	4	SR4	100-105	4
			SR4	105-110	3
LE3	50-90	10			
LE3	50-90	2	SR5	0-5	3.5
LE3	90-95	4.5	SR5	5-10	4
LE3	95-100	4	SR5	10-15	4
LE3	100-105	4	SR5	15-20	4
LE3	105-110	4	SR5	20-25	4
LE3	110-115	4	SR5	25-30	4.5
LE3	115-120	4	SR5	30-35	4
LE3	120-125	4	SR5	35-40	4
LE3	125-130	3.5	SR5	40-45	4.5
LE3	130-135	3.5	SR5	45-50	3
			SR5	50-55	4

SAMPLE	DEPTH (FT)	WEIGHT (LBS)	DESCRIPTION
AC1, run #1		7	U12G Tunnel
AC1, run #4-6		35	U12G Tunnel
AC1, run #7		35	U12G Tunnel
AC1, run #14-15		55	U12G Tunnel
AC1, run #16-17		27	U12G Tunnel
AC2, run # 5-7		65	U12G Tunnel
AC2, run # 8-12		50	U12G Tunnel
AC2, run #13-16		70	U12G Tunnel
AC2, run #17-18		50	U12G Tunnel
AC2, run #19-20	44.2-54.2	25	U12G Tunnel
AC2, run #19-20		27	U12G Tunnel
AC2, run #25-26		65	U12G Tunnel
AC2, run #27-31		65	U12G Tunnel
AC2, run #32-37		65	U12G Tunnel
AC2, run #38-40		22	U12G Tunnel
AC2, run #38-40	92.0-106.0	27	U12G Tunnel
AC2, run #41-44		65	U12G Tunnel
AC2, run #45-50		70	U12G Tunnel
AC2, run #55-457		55	U12G Tunnel
DH1, run # 5-6	17.3-27.3	45	U12G Tunnel
DH1, run # 7-9	27.3-37.0	53	U12G Tunnel
DH2, run # 7-8	20-25	31	U12G Tunnel
DH3, run #2-3	0-10.8	65	U12G Tunnel
DH3, run #4	10.8-15.8	10	U12G Tunnel
DH3, run #5		28	U12G Tunnel

DATE

FILMED

11 / 01 / 94

END