Yucca Mountain Site Characterization Project

Peer Review Report

on

CHLORINE-36 STUDIES AT YUCCA MOUNTAIN

Peer Review Team Approval

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Table of Contents

List of Figures

Executive Summary

Introduction

This report presents the conclusions and recommendations of a formal peer review of the ^{36}Cl and related hydro-geochemical investigations for the Yucca Mountain Project (YMP). The YMP charged the peer review team (PRT) with the following tasks:

- Review the most recent synthesis reports on ^{36}Cl and other environmental isotopes in the context of the three-dimensional geologic framework model and updates of the unsaturated flow and transport models.
- Evaluate the adequacy of the sampling approach and locations utilized.
- Evaluate the adequacy of the fluid-extraction and isotopic analytical approach, the precision of the data, and the accuracy of the data.
- Evaluate the adequacy of interpretation of the ^{36}Cl and other environmental isotope data in the context of the current conceptual flow models and modeling approach that incorporate the site-hydrogeologic features, rock heterogeneity, alteration and fracture mineralogy, and geochronologic data on the fracture-lining minerals.

Issues

The Peer Review Team identified five major issues. These were:

1. Is the bomb signature real?
2. Can the anomalies in $^{36}Cl/Cl$ ratio be explained by variations in the source strength with time or mixing of waters with different $^{36}Cl/Cl$ ratios? Could variability be explained in terms of mixing with fluid inclusions from the rock?
3. Are ^{36}Cl anomalies an artifact of sampling and analysis (Section 3.5) -- This issue addresses whether the selection of sites, sampling methods, or analysis procedures bias the ^{36}Cl measurements.
4. Is there adequate integration of isotope and environmental tracer programs. Each component of the hydrochemical sampling program reflects a somewhat different feature or process in the flow system. Are the various isotope and environmental tracer programs properly integrated to achieve a consistent conceptual model of the flow system?
5. Integration with conceptual and numerical flow models -- are ^{36}Cl and other environmental tracers being used effectively in the context of conceptual and

numerical flow models? This issue considers (1) the use of models to test the plausibility of fast-path hypotheses, (2) the use of ^{36}Cl to constrain model estimates of water flux and travel time through the repository horizon, and (3) the value of bomb-pulse observations in the ESF for predicting flow below the repository through the CHn.

Bomb Pulse Source of ^{36}Cl

The opinion of this report is that the elevated $^{36}Cl/Cl$ ratios found in the ESF and other samples originate from the infiltration of ^{36}Cl from weapons tests fallout during the past 50 years. Furthermore, a 1250×10^{-15} $^{36}Cl/Cl$ threshold is conservative in identifying bomb waters. It is very important to note, however, that the corollary is not necessarily true -- waters below that threshold have not been claimed to be free of bomb ^{36}Cl and could well be a mixture of non-bomb and bomb-pulse waters. Flow simulations further support the bomb-pulse origins of ^{36}Cl at the ESF. Other explanations of the elevated $^{36}Cl/Cl$ ratios such as in-situ formation of ^{36}Cl in surface exposures containing calcite do not explain the elevated $^{36}Cl/Cl$ ratios.

This report concurs with YMP Principal Investigators that the elevated $^{36}Cl/Cl$ ratios are consistent with unsaturated zone models that contain fast paths and these pathways can only be attributed to water movement through fractures that occurs with minimal matrix interaction in the Paintbrush Tuff (PTn).

Sampling Challenges and Need for Better Understanding of Fast-path Geology and Geometry

Mixing and dilution of chloride have large effects on masking bomb-pulse ^{36}Cl. Many chloride-bearing pulses of water are likely to have passed through the fractures at the ESF level. Current ^{36}Cl levels should represent a mix of sources from both before and after the nuclear testing era. Further mixing and dilution may occur during sampling from the inclusion of significant volumes of rock in addition to the fracture surfaces.

This report concurs with conclusions of YMP Principal Investigators that dilution and mixing seriously affect the ability to detect bomb-pulse chlorine. Bomb-pulse chlorine is an indicator of a fast path, however, *the absence of bomb-pulse chlorine does not preclude the presence of a fast path.*

Effective sampling will require a better understanding of the fracture flow and transport processes. The geologic nature of the fast paths is not yet well enough understood to guide sampling efforts to likely locations with elevated chlorine isotope ratios. Bomb-bearing paths are not mineralogically distinctive. Although there may be a relationship of pathways to faults, fracture pathways in the welded tuffs may track fast paths well away from fault locations. In summary, sampling approaches require improvement to better define fast-path locations. Sampling needs to be feature-specific as systematic samples are unlikely to yield useful results.

Importance of Isotope and Environmental Tracer Studies to Modeling and Site Assessment

Hydrochemical data, including $^{36}Cl/Cl$ ratios, are an essential complement to the modeling of water flow in the Yucca Mountain site. Without hydrochemical data, the ranges of modeling parameters are highly under-constrained. The matrix saturation, which is the main calibration criterion, can be matched by a wide range of models and data sets, including the now-superseded single permeability models. Although the dual permeability formulations represent a major conceptual improvement, major uncertainties remain in such parameters as the fracture-matrix interaction terms and the in situ fracture saturations. Furthermore, fundamental processes of unsaturated flow in the fractures remain uncertain. The introduction of thin-film flow concepts (Tokunaga and Wan 1997) may indicate flux capacities in excess of those theorized by more conventional two-phase, porous-media approaches.

In addition to the flux calculations, using hydrochemistry to study pathways through the PTn to the ESF provides a useful analog for the downstream flow from the repository through the Calico Hills tuff to the saturated zone.

Importance of $^{36}Cl/Cl$ Studies to the YMP

The $^{36}Cl/Cl$ data are critical to understanding flow and transport at Yucca Mountain. $^{36}Cl/Cl$ studies provide field observations of flow and transport over large space and time scales and provide valuable information on travel times. Studies at the ESF unequivocally demonstrate fast flow to this level that has been attributed to bypassing of the PTn by faulting and increased fracturing associated with faults. Lower $^{36}Cl/Cl$ ratios (500 to 1250 $\times 10^{-15}$) have been interpreted to represent older water (>10,000 years). Integration with other isotopic data (^{3}H, ^{99}Tc, and ^{2}H and ^{18}O) will be very important to confirm or refute the hypothesis that $^{36}Cl/Cl$ ratios between 500 and 1250×10^{-15} reflect old water or a component of bomb pulse water. Because of the potential for false negatives with the $^{36}Cl/Cl$ analyses as a result of mixing and dilution, it is possible that there are more fast pathways than suggested by the conceptual model. Although it has been suggested that the volume of water flowing through fast pathways is negligible (~ 1%), this volume has been estimated based on the current conceptual model for ^{36}Cl that has yet to be confirmed. ^{36}Cl ratios between 500 and 1250×10^{-15} could represent a component of bomb pulse that would also increase the volume of water flowing in fast pathways. The volume of flow in fast pathways could be much greater beneath areas of higher infiltration. This hypothesis will be tested when the E-W drift is constructed because infiltration rates based on Flint et al. (1996) are 3 to 10 times greater above the E-W drift. Until information on these issues is obtained, (fast flow under higher infiltration, interpretation of ^{36}Cl/Cl ratios between 500 and 1250×10^{-15}), it is premature to estimate quantities of water involved in fast flow or to evaluate fluxes associated with fast flow.

In addition to providing information of flow processes (i.e. fast pathways) the ^{36}Cl/Cl data have played a key role in determining the type of model to be used at Yucca Mountain and in refining fracture matrix parameter sets for the numerical simulations. Initially equivalent continuum models were used to simulate flow and transport through Yucca Mountain (Wittwer et al., 1995); however, such models were unable to reproduce the distributions of ^{36}Cl/Cl found at the level of the ESF. The occurrence of bomb pulse ^{36}Cl/Cl at the ESF forced the modelers to use a dual permeability model to simulate fast pathways (Robinson et al., 1995). Therefore, the ^{36}Cl/Cl data effected a major change in the modeling process. Several parameter sets have been developed during the course of the Yucca Mountain program which demonstrate that calibration of these parameters to matrix saturation's is highly non-unique (Banduragga and Bodvarsson, 1997). The ^{36}Cl/Cl data provide a critical constraint to these parameters sets for the current flow and transport model (Robinson et al., 1997) and will continue to play a role in refining these parameter sets as new data becomes available. The infiltration map developed by Flint et al. (1996) indicates that the ESF is located under a zone of low infiltration whereas the E-W drift will be located under a zone of ~ 3 to 10 times higher infiltration. ^{36}Cl/Cl and Cl data from the E-W drift will help evaluate whether fracture flow is much more prevalent through the PTn under wetter conditions. For example, if bomb pulse ^{36}Cl/Cl ratios are found in many areas of the E-W drift, this would confirm the higher infiltration rates and indicate that fracture flow can be sustained through the PTn under wetter conditions. Such results would lead to major changes in the hydrologic parameter set to be consistent with such data.

While most studies focus on flow through the PTn, studies are also required to evaluate flow through the CHn unit. Tests should be conducted to determine if there are bomb pulse tracers associated with faults in the CHn in the Busted Butte program. These data will help constrain hydrologic parameters for modeling this unit. In summary, the ^{36}Cl/Cl data provide a valuable tool to (1) evaluate flow processes through various units, (2) constrain hydrologic parameter sets for numerical simulations, and (3) test the surface infiltration model.

Conclusions

The peer review reached five major conclusions which are the following:

1. Bomb-pulse sources are currently the only plausible explanation of the elevated (>1250×10^{-15}) ^{36}Cl/Cl ratios observed in the ESF.

2. ^{36}Cl/Cl has provided the main evidence for fast paths, and continued effort in sampling for ^{36}Cl/Cl ratios, particularly in the E-W drift and in accessible portions of the CHn such as at Busted Butte, are essential for understanding site hydrogeology.

3. Current sampling programs almost certainly underestimate the numbers of occurrences of bomb-pulse $^{36}Cl/Cl$ in the ESF due to (a) the dilution and mixing with stable chloride and (b) the challenges of identifying and sampling the likely fast-flow paths.

4. Measurement programs for $^{36}Cl/Cl$ require strong coordination with sampling efforts for other isotopes and environmental tracers.

5. The numerical modeling programs require geochemical data, including $^{36}Cl/Cl$, to constrain the ranges of input parameters, particularly as the geology and flow processes of fast paths may not be understood well.

Recommendations

Need for $^{36}Cl/Cl$ Ratio Studies and Improvement of Sampling Strategies

Future field work should focus upon feature-based sampling and should include provision for the analysis of the multiple suite of isotopes mentioned in this set of recommendations.

The method to select feature-based samples for ^{36}Cl analysis should be reviewed and clearly documented. Greater effort should be made to geologically distinguish potential fast paths (though mineralogy has not been successful in this regard). Also methods should collect sufficient sample material for analyses of other isotopes.

The total concentration of stable Cl should be measured in each rock type so that corrections can be made for the amount of dilution introduced during the leaching of the samples.

A method should be developed to estimate the amount of Cl (and ^{36}Cl) extracted from the rock and fluid inclusions during the pore water extraction. Possible approaches that could be explored are differences in the pore water and rock in ^{35}Cl, ^{37}Cl, ^{87}Sr, and anion ratios in the extract. Such a correction would increase the power of the ^{36}Cl method so that the influence of dilution is minimized.

The additional analyses referenced above should be used to examine alternative conceptual models of the ^{36}Cl distribution. These models should include geologic features, dilution, proximity to fractures, and the pneumatic characteristics of the rock mass.

Integration with Other Isotopic and Environmental Tracer Methods

The panel recommends that further work in isotope hydrology should use multiple isotopes (^{3}H, ^{99}Tc, $^{13,14}C$, ^{36}Cl, and stable isotopes, ^{18}O and D). Samples should be large enough to provide splits for analyzing multiple isotopes.

^{99}Tc should be measured in a suite of about 10 ESF samples that have already been analyzed for ^{36}Cl (with appropriate blanks and standards). ^{87}Sr/^{86}Sr isotope ratios, which reflect rock water interaction, should be measured to assist in distinguishing waters passing through fast paths from those traveling matrix pathways in the non-welded units.

The presence or absence of tritium concentrations greater than ~ 1 TU in pore waters provides key corroboration of recent or post bomb water recharge. However, the reliability of past low level tritium measurements (<~16 TU) is not clear because procedural blanks have not been performed routinely and lower limits of detection are often not clearly stated. Better detection limits are needed for this work. The panel recommends that the program investigate the use of isotope enrichment and gas flow proportional counting to achieve an acceptably low background detection threshold (~0.5 TU). The panel recommends that the program develop and use a method to determine the tritium concentration in field blanks.

Integration of Environmental Tracer Studies with Testing and Modeling

The panel believes that the surface infiltration studies and related model development provide a key constraint on the flow through the unsaturated zone. The amount of infiltration drives flow and transport in the PA models. Work on the infiltration model should be continued with an emphasis on refining the upper limits on infiltration under wetter climates.

The panel recommends that the Yucca Mountain Project develop and test, through experimentation, alternative conceptual models of the flow processes, such as thin-film process models. The results of percolation tests on natural fractures and observations of re-wetting of water bearing fractures under controlled humidity conditions underground should contribute greatly to understanding conductive fracture frequencies and flux localization. These data should be critical to focused sampling for hydrochemistry.

The assumption that there is fracture flow through the PTn is a key assumption that must be invoked to decrease travel times so that bomb pulse nuclides can rapidly penetrate into underlying units. The panel recommends that the YMP investigate ways to locate and quantify flow through this breach.

Transport rates and water fluxes through major faults are key parameters in the YMP site studies. The panel recommends that:

> Determining the hydraulic properties of fractures (as opposed to matrix flow) should receive proportionately more effort in future work.
>
> All appropriate aspects of the program should test their understanding of the flow system by documenting their predictions of the hydrologic properties of the Solitario Canyon fault before it is crossed in the E-W drift. The value of predictions of bomb-pulse occurrence will be increased greatly if uncertainty in

sampling methods and sampling site selection (i.e. targeted fractures) can be decreased.

Transport rates and water fluxes through major faults in units below the repository are key parameters in the YMP performance assessment work. The panel believes that the test program at Busted Butte provides an excellent way to learn about fast-path potential in the CHn and to test predictions and isotopic techniques developed in the ESF studies. The panel recommends that:

The Busted Butte program plan should be reviewed to integrate more isotopic and environmental tracer work.

The hydraulic properties of faults and fractures through Busted Butte should be investigated.

All appropriate aspects of the program should test their understanding of the flow system by documenting their predictions of the hydrologic properties of the Busted Butte system before it is instrumented.

The Busted Butte flow system should be investigated using multiple isotopes (^{3}H, ^{99}Tc, $^{13,14}C$, ^{36}Cl) and stable isotopes (^{18}O-and D). in order to test the predictive power of the techniques. The panel recommends that multiple isotopes should be used to test for fast pathways and to estimate water fluxes in the unsaturated zone in geologic units below the repository horizon.

1. Introduction

1. 1 Objective of the Peer Review

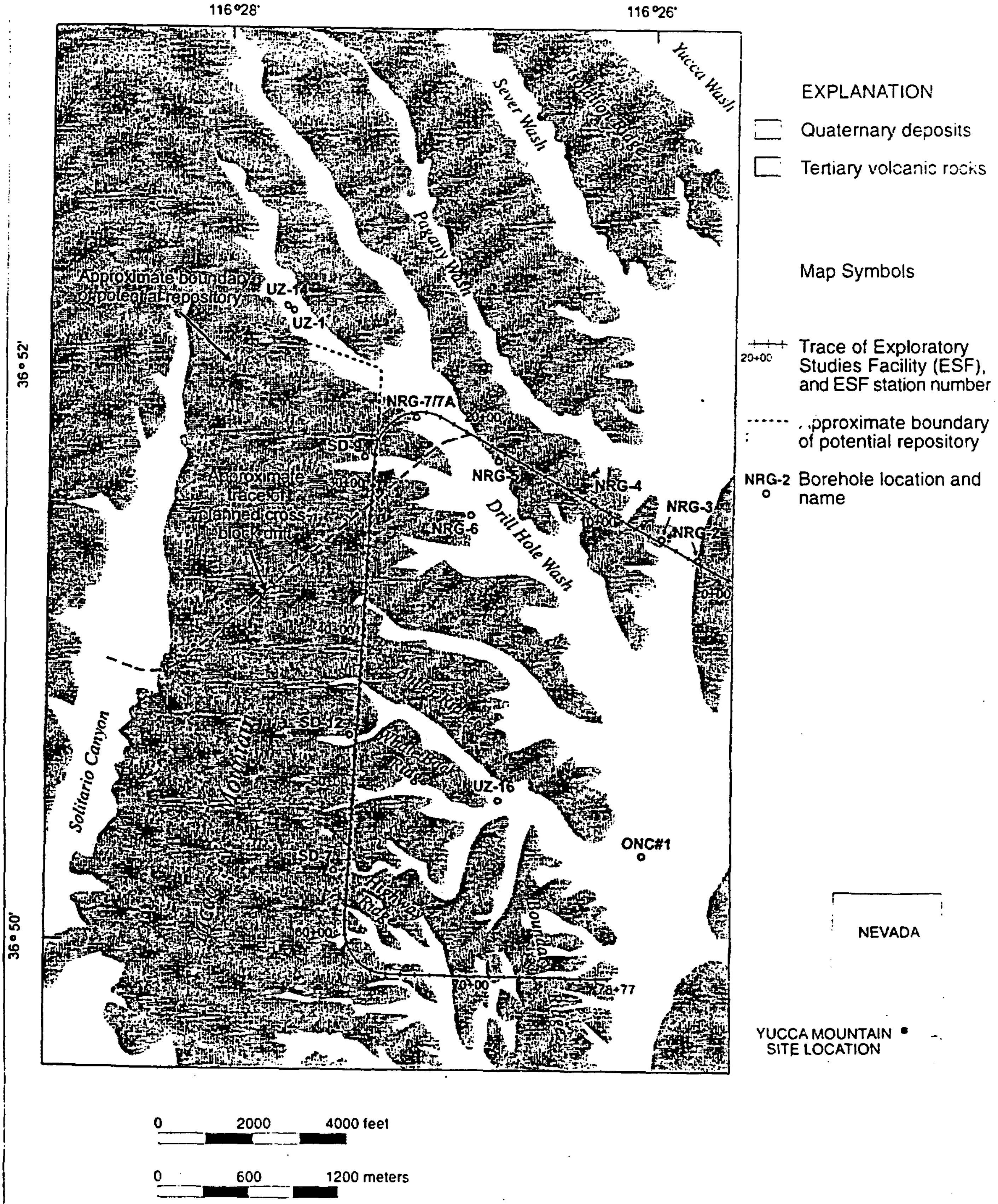
The Yucca Mountain Site Characterization office of the DOE's Office of Civilian Radioactive Waste Management established a Peer Review Team (PRT) to conduct an external review of the sampling, analysis, interpretation, and application of the ^{36}Cl and other environmental isotope data collected by the Yucca Mountain Project. This report discusses the following:

- the background of the ^{36}Cl studies,
- key issues regarding ^{36}Cl and implications for fast-path groundwater flow, and
- the conclusions and recommendations of the Peer Review Team.

1.1.1 Origins of ^{36}Cl and Its Discovery at Yucca Mountain

The naturally occurring radioactive isotope, ^{36}Cl, has its major natural source in cosmic rays bombarding stable chlorine, ^{35}Cl, and argon in the atmosphere. Over the past 50,000 years the ratio of ^{36}Cl to stable chlorine in atmospheric deposition at Yucca Mountain has varied between about 500×10^{-15}(present day) to about 1000×10^{-15}. This range was greatly exceeded for several years during the late 1950's and early 1960's when ratios as high as $217{,}000\times10^{-15}$ appeared in the atmosphere as by-products of atmospheric nuclear testing (Synal, et al., 1990). The major portion of the atmospheric bomb-pulse ^{36}Cl was created by weapons tests in the Pacific Ocean, where the ocean water provided a large chlorine source. Waters which entered subsurface flow systems from precipitation during this time, and which have not picked up significant amounts of chlorine from other sources, carry a distinctive, "bomb-pulse" isotopic signature.

The initial investigations for ^{36}Cl in the Exploratory Test Facility at Yucca Mountain (ESF, Figure 1) expected to find isotopic decay of ^{36}Cl (half -life of ^{36}Cl 301,000 years) which would indicate old pore water. Rather than finding depleted $^{36}Cl/Cl$ ratios, the



Source: Fabryka-Martin et al. 1977. Figure 1-1.

FIGURE 1
MAP OF ESF
YUCCA MT/PEER REVIEW/NV

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Yucca Mountain Project (YMP) investigations discovered elevated isotopic ratios, which suggested that some of the pore waters might contain bomb-pulse ^{36}Cl. A determination of a bomb-pulse origin for this isotopic anomaly would indicate travel times less than 50 years from the surface to the ESF and affirm the existence of fast transport pathways to depths at least as great as the ESF if not greater.

1.1.2 Peer Review Tasks

Given the significance of accurate assessments of ^{36}Cl and other environmental isotopes to the Yucca Mountain Project, the PRT was assigned the following tasks:

- Review the most recent synthesis reports on ^{36}Cl and other environmental isotopes in the context of the 3D geologic framework model and updates of the unsaturated zone flow and transport models.
- Evaluate the adequacy of the sampling approaches and locations utilized.
- Evaluate the adequacy of the fluid-extraction and isotopic analytical approach, the precision of the data, and the accuracy of the data.
- Evaluate the adequacy of interpretation of the ^{36}Cl and other environmental isotope data in the context of the current conceptual flow models and modeling approach that incorporate the site-hydrogeologic features, rock heterogeneity, alteration and fracture mineralogy, and geochronologic data on the fracture-lining minerals.

1.2 Peer Review Process.

The Peer Review was officially initiated by Dr. Steven Brocoum, Assistant Manager for Licensing in the DOE's Yucca Mountain Site Characterization Office, and Mr. Russell Patterson, Geohydrologic Lead, with the issuance of a Peer Review Notice appointing a chairperson (Dr. Thomas W. Doe) and a Technical Contact in Las Vegas (Dr. Ronald Linden of the Management and Technical Support organization of the Yucca Mountain Project Office).

The next step was preparation of a Peer Review Plan (Appendix 1) and selection of the PRT members. The following persons were selected to serve on the panel.

Dr. R. Jack Cornett (Atomic Energy of Canada, Chalk River), Chlorine isotope geochemistry.

Dr. Anthony B. Muller (Booz-Allen Hamilton). Isotope geochemistry, and

Dr. Bridget R. Scanlon (University of Texas/Texas Bureau of Economic Geology), Unsaturated zone transport

Dr. Linden arranged for distribution of the major review materials during the Fall of 1997; a list of major documents which were reviewed by the PRT is included in Appendix 3.

The PRT met in Las Vegas, January 13-16, 1998 to hear presentations, ask questions, and engage in discussion with the major Principal Investigators active in the ^{36}Cl and other environmental isotope studies. Appendix 2 contains the agenda, list of speakers and attendees to the PRT meetings. The second day of the meeting was spent as a field trip to the Yucca Mountain site to inspect sampling localities and to observe the major hydrogeologic features and geologic units both underground and on the surface.

1.3 Organization of the Report

This report is presented in four sections. Section 2 contains background material on the geology and hydrogeology which underlie the YMP's current conceptual model and its numerical implementations. Section 2 also discusses significant alternative conceptual models which were presented to the PRT. Section 3 presents the PRT's assessment of the major issues regarding ^{36}Cl and other environmental isotopes including a discussion of their importance to YMP performance. Section 4 presents the PRT's conclusions and recommendations.

Thomas Doe prepared sections 1 and 2 of the report. The PRT jointly developed the issues listed in section 3, and assigned different PRT members the primary responsibilities for preparing the discussion of each issue. These primary responsibilities and authorships are noted in each of the section 3 headings. The authorships of the conclusions and recommendations in section 4 are similarly noted. Each peer review member reviewed all sections of the report. The conclusions and recommendations are unanimous unless otherwise noted.

2. Background

2.1 Hydrogeologic Setting of the Yucca Mountain Site

This section is a synopsis of the hydrogeologic conceptual model of the Yucca Mountain site as it was presented to the PRT though the review documents and presentations of the Principal Investigators of the program. We will refer to this as the "centrist" model. The synopsis is followed by a discussion of alternatives to the centrist model.

2.1.1 Surficial Units

Descriptions of the surficial materials and their infiltration behavior have been developed by Flint (et al, 1997). The topographic surfaces and surficial materials play an important role in localization of infiltration and storage of chemical constituents such as chloride. The thicknesses of surficial materials vary from over 100 meters in valleys to zero on some side slopes and terraces. Most washes contain fluvial and debris-flow materials, as well as some, but relatively little, bedrock exposure. Calcium carbonate cements and fillings are abundant in surficial materials and in fractures appearing in bedrock exposures.

Surficial and near-surface processes are important to the evaluation of chloride transport and fast-path development. Chloride and ^{36}Cl are stored in the near surface materials, and travel times through the thick surficial materials should be sufficient to preclude these areas as sources for the bomb-pulse nuclides observed in the ESF. Evapotranspiration affects chloride concentrations near the surface, thus becoming the important consideration in using chloride mass balances as a means of estimating groundwater infiltration and fluxes.

Direct measurements of the site infiltration and its local variability are not technically possible, however well-reasoned estimates of material thickness and properties have supported construction of a site wide model. Boreholes for neutron measurements of saturated porosity provide the main body of data for calibration of the infiltration model.

The source of water to the site is precipitation, which averages about 170 mm per year. Flint et. al. (1996) have developed an infiltration model which partitions this source into components of runoff, evapotranspiration, storage in the surficial cover, and ultimate infiltration to the bedrock. The infiltration is estimated to average 4.5 mm/year, however the actual rates are expected to vary considerably both spatially and temporally. Infiltration is thought to be as high as 50-80 mm/year at higher elevations or in areas with thinner soil cover such as on side slopes or benches. Net infiltration may be zero where the surficial materials are thicker than 6 meters. In addition to annual and seasonal variations, the greater portion of bedrock infiltration is thought to occur with major precipitation events, which fill the storage capacity of the thinner surficial materials and overflow into the bedrock fracture system.

2.1.2 Bedrock Hydrostratigraphy

Bedrock at the Yucca Mountain site consists of tilted and faulted volcanic strata. Montazer and Wilson's (1984) definitions of hydrostratigraphic units continue to provide a basis for the current conceptual and numerical models of the site. They proposed five major hydrostratigraphic units, which are the following from shallow to deep (thicknesses from Hinds, et al, 1997).

- Tiva Canyon welded unit (TCw), consisting of welded tuff units in which fracture flow is thought be the dominant mode of air and water movement;
- Paintbrush non-welded unit (PTn), a higher porosity unit in which fractures are few and discontinuous, and in which matrix flow is predicted to be prominent. The PTn retards flow (both physically and chemically) between the surface and the depth of the repository and the ESF. PTn thicknesses vary over the ESF from 70 m in the north to less than 20 m in the south.

- Topopah Springs welded unit (TSw), a fracture-flow dominated unit which hosts the ESF and planned repository. Thicknesses vary from about 240 to 340 m in the area of the ESF.

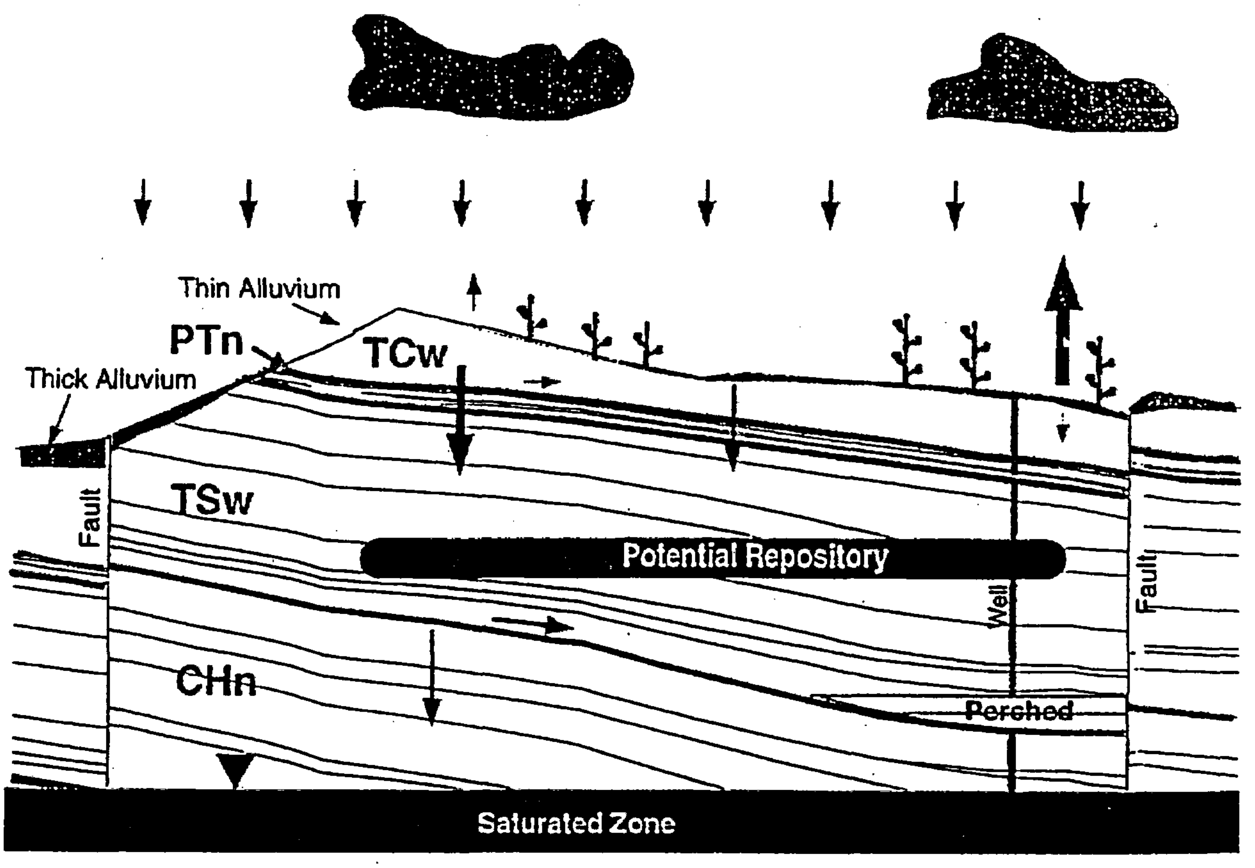
- Calico Hills non-welded unit (CHn), a non-welded tuff which is zeolitized in some sub-units. Its thickness under the ESF varies from 40 to over 100 m. The site conceptual model views the CHn as the primary barrier to radionuclide migration from the repository based on matrix-dominated flow with low relative-permeability to water , and the retardation potential of the zeolitic mineralization. The hydraulic properties of the CHn cause the development of local perched zones.

- Crater Flat undifferentiated unit (CFu), which was not discussed during the peer review.

Figure 2 presents a schematic representation of the site hydrogeology.

2.13 Hydrostructural Units

In addition to the hydrostratigraphic units, hydrostructural units also have important roles in controlling air and water flow at the Yucca Mountain site. Faults create offsets in otherwise continuous units. The hydrologic role of faults may be most pronounced in the non-welded tuff units. Here fractures related to the faults may act as fast pathways which bypass the favorable characteristics of the bedded units for retarding radionuclide transport. Faults cutting the PTn above the ESF and the repository may be the primary pathways for the the bomb pulse ^{36}Cl observed in the ESF. Below the repository, faults across the CHn could affect repository performance. The significant faults in the ESF area are the Sundance Fault, the Ghost Dance fault, and the Drill Wash fault.



Source: Fabryka-Martin et al, 1977, Figure 2-3.

FIGURE 2
YUCCA MOUNTAIN CONCEPTUAL HYDROGEOLOGY
YUCCA MT/PEER REVIEW/NV

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2.1.4 Hydrochemistry

A comprehensive hydrochemical model of the Yucca Mountain site does not yet exist. In part the development of such a model is hampered by difficulties in obtaining samples from fractures or from welded tuff units, which have very low matrix porosities. Nonetheless, a brief discussion of some of the particular hydrochemical constituents -- Cl, ^{3}H, ^{14}C, and Sr -- provides a useful background for reviewing ^{36}Cl distributions. Also, ^{14}C and Cl- contrasts between perched waters and matrix pore waters in surrounding rocks may provide insights into water flow and transport.

The chloride concentration in pore water has particular importance for ^{36}Cl studies. Chloride travels in the water phase and has little interaction with the rock chemistries. Hence chloride distributions should reflect water movements without the ambiguities of air-phase interaction or rock interaction.

Sonnenthal and Bodvarsson (1997, sec. 15.4.1) summarize the chloride distributions. Chloride content in the shallow subsurface is thought to reflect the balance of near surface infiltration and evapotranspiration rates. Near the surface, higher bedrock infiltration rates and lower chloride concentrations appear at higher elevations, while at lower elevations thicker surficial deposits and lower infiltration rates result in higher chloride concentrations. In the absence of lateral movement at non-welded unit contacts, chloride content variations should propagate vertically downward from the surface. The total chloride content is a key issue for detecting bomb-pulse isotopes, as higher concentration of Cl mixing effects may mask the bomb-pulse component of ^{36}Cl.

Perched waters above the CHn have low chloride concentrations which contrast with the higher concentrations in matrix pores. The lack of equilibrium between these waters is a key factor in assessing the origin, age, and degree of mixing of the perched waters.

Strontium isotope, $^{86}Sr/^{87}Sr$, data reflect rock water interactions. Vertical profiles of Sr ratios through the TCw, PTn, and TSw show a the greatest changes over the relatively thin section of PTn, indicating that rock-water interactions occur most strongly in the non-welded tuffs where matrix flow is thought to dominate.

^{14}C is a well-known radionuclide for water dating. Radiocarbon is present at Yucca Mountain in three phases -- mineral phases, vapor phases, and dissolved aqueous phases. The use of carbon isotope ratios is complicated by the possibilities of both vapor and aqueous transport, as well as rock-water interactions which may invalidate inferences of pore water age.

2.2 Conceptual Model of Flow and Transport

This section discusses the centrist conceptual model which is being employed by the two numerical simulation teams, Lawrence Berkeley (LBNL) and Los Alamos (LANL) National Laboratories. Alternate hypotheses within the Yucca Mountain Project are discussed in section 2.3. The conceptual model description starts at the surface and proceeds downwards, and the focus of the discussion is the movement of bomb-pulse ^{36}Cl.

Water is retained in the surficial materials where it may remain in storage, be removed by evapotranspiration, or pass on to the bedrock in periods of heavier rainfall. The capacity for infiltration to the bedrock depends strongly on the storage capacity of the surficial materials, hence bedrock recharge occurs mainly in areas with thin or non-existent surficial materials, particularly at higher elevations where precipitation is greater. Bomb-pulse ^{36}Cl reaches the bedrock at these thin-cover locations. In locations with thicker surficial materials, bomb pulse ^{36}Cl is limited to the surficial materials and does not appear in the underlying bedrock.

Where water does enter the bedrock flow system, it moves rapidly by fracture-flow through the TCw welded tuffs to the surface of the non-welded tuff the the PTn. Matrix-dominated flow in the PTn slows the downward movement of water. Saturations increase at the PTn, and temporal fluctuations in pressure are damped. Significant rock-water interaction occurs as waters pass through the PTn based on changes in the Sr isotope ratios with depth.

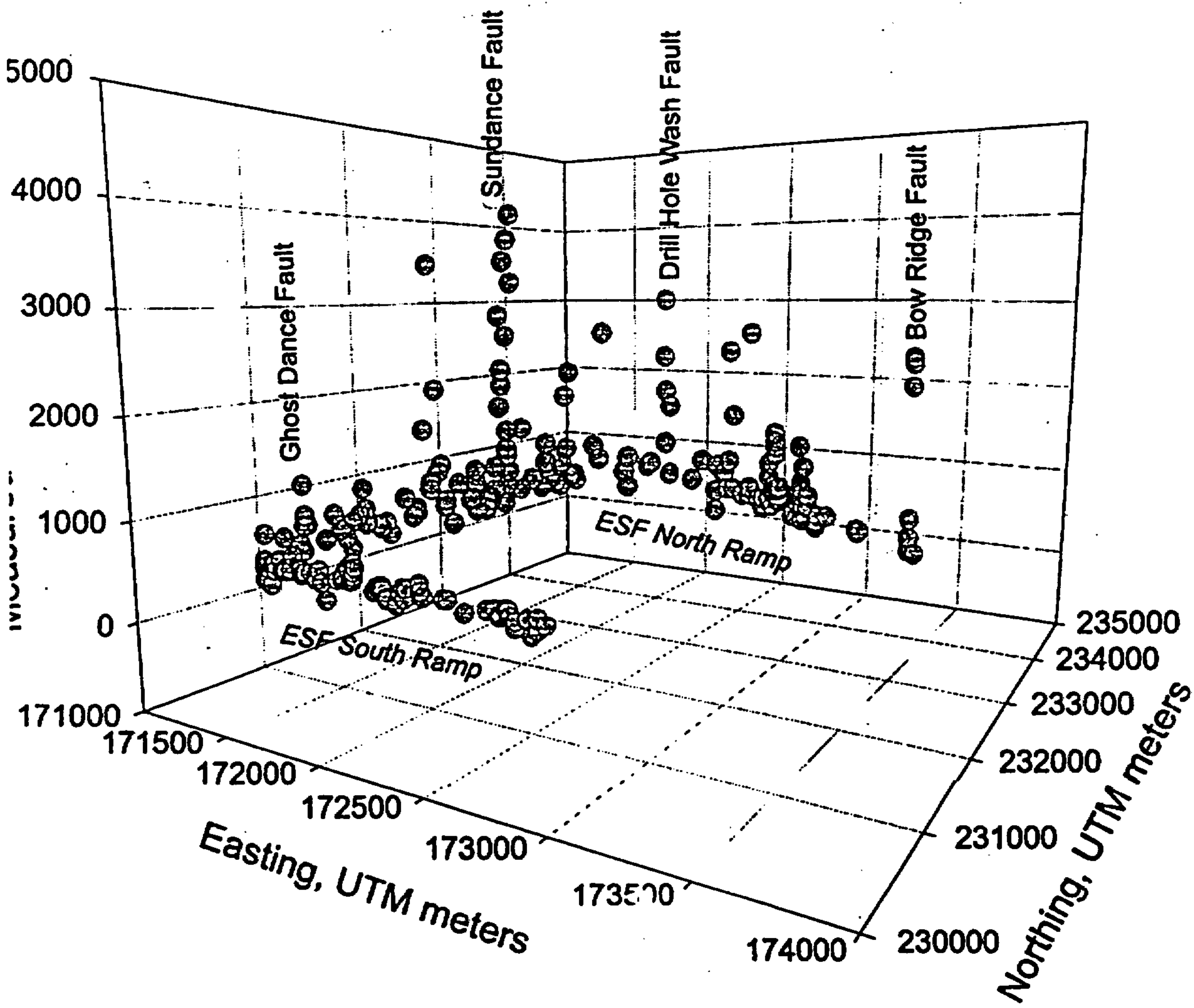
Bomb-pulse ^{36}Cl is relatively widespread in the TCw above the PTn, where it remains except where faults or other fractures penetrate the complete thickness of the PTn. At these locations, bomb-pulse ^{36}Cl and other bomb-pulse tracers may pass relatively unhindered through the PTn and enter fast pathways in the underlying TSw welded tuffs.

Water moves relatively rapidly by fracture flow through the TSw. Pathways which are downstream of fault breaches in the PTn carry the bomb-pulse signature. The materials above the PTn act as a ^{36}Cl reservoir, therefore, the ^{36}Cl supply through the PTn to the TSw follows more of a step function than a pulse. TSw pathways which have matrix-water sources in the PTn may travel with similar velocities as the bomb-pulse pathways, but bear geochemical signatures which reflect the delays of passage through the PTn rather than the bomb-pulse signatures.

Bomb-pulse ^{36}Cl appears in the ESF along those pathways that meet several basic criteria -- (1) a sufficient water source to sustain flow to the ESF depth, (2) thin surficial materials at the origin of the pathway, and (3) a fracture pathway across the PTn that has little or no interaction with the PTn.

Below the ESF and repository horizon, transport continues by fracture flow until it encounters the CHn units. At this point water may move laterally or perch (these two are not mutually exclusive) above the CHn. The CHn is a strong potential flow barrier in both a physical and a chemical sense. Whether or not fracture flow occurs across the CHn in a manner similar to the hypothesized movement across the PTn is an important point, however, the perched water systems provide some constraint on what portion of the flux could be moving rapidly through the CHn in zones where perching occurs.

The observations of ^{36}Cl at the Yucca Mountain site are only partly consistent with the conceptual model described above. There appears to be a relationship between some but not all faults and bomb-pulse nuclide appearances in ESF samples (Figure 3). Also, there



Source: Fabryka-Martin et al, 1977, Figure 2-3.

FIGURE 3

^{36}Cl MEASUREMENTS ALONG ESF

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is an absence of ^{36}Cl in the southern part of the ESF, where a thinner PTn would lead one to expect a greater likelihood of ^{36}Cl. Sonnenthal noted in presentations to the PRT that the absence of ^{36}Cl in the southern area may reflect locally-reduced infiltration. The discussion of the south ramp ^{36}Cl occurrences is preliminary, as not all samples have been analyzed from the south ramp area.

2.3. Numerical Representations of the Conceptual Model

The final discussion point for the centrist conceptual model evaluates how the YMP conceptual model has changed over time. The discovery of bomb-pulse ^{36}Cl forced fundamental rethinking of the transport mechanics of the site. Previously, fractures had not been viewed as the major transport vehicle, as the fractures were though to have low relative permeabilities except when temporarily or locally saturated. These earlier models of flow (Wang and Narasimhan, 1984) surmised that air-gaps in partially saturated fractures would prevent significant fracture flow in the unsaturated zone, and the major portion of flow was assumed to occur by matrix pathways. There had been confidence in the single (matrix) permeability models because they were capable of replicating the matrix saturation profiles, which have been one of the main calibration criteria for the simulations.

Single permeability codes, however, were not able to simulate the fast-path transport that would account for ^{36}Cl occurrences at the ESF. The main conceptual adjustment to allow for fast pathways was adoption of dual permeability models by both simulation groups at Berkeley (LBNL) and Los Alamos (LANL). Dual permeability models view the rock as having two distinct but mutually embedded flow systems, one system representing the matrix and the other the fractures. The flow processes are the same everywhere, but the two systems would have very different properties. The interaction between the two systems would be governed by fracture-matrix interaction terms. These terms are not measured directly, but are inferred from calibration studies of the matrix saturation profiles. As discussed above, there is a large range of fracture properties that may be inferred from the inversion and calibration studies, and a primary function of the isotope studies is to provide further constraints on the ranges of model parameters.

2.3 Alternatives to the Conceptual Model within the YMP

Two major alternatives to the "centrist" conceptual model were presented to the PRT in oral discussions. John Stuckless of the USGS noted that bombardment of surficial calcites, which are abundant at Yucca Mountain, also produces ^{36}Cl. This source, according to this hypothesis, could account for the ^{36}Cl observed at depth. Section 3.3 and Appendix 4 present the PRT's comments on this hypothesis in detail.

The second hypothesis was presented by Mark Tynan, also of the USGS. His main argument was that an absence of bomb-pulse signatures, particularly in the southern part of the ESF, might represent compete flushing of the bomb-pulse nuclides through the system, and that bomb-pulse materials had already traveled below the ESF.

The "flushing" hypothesis may not be the preferred explanation of the absence of ^{36}Cl in parts of the ESF, as there are many other reasons why ^{36}Cl should be difficult to sample or why bomb-pulse could be masked (see section 3.4). Second, flushing should also remove bomb-pulse materials from the surficial materials near the fast-path sources, yet a reservoir of bomb-pulse nuclides appears to exist today in these source areas above the southern ESF. The flushing hypothesis, however, does emphasize the need for a better understanding of the geologic nature of the fast pathways.

3. Issues for Review

3.1 Importance of ^{36}Cl and Other Environmental Tracer Studies to the Yucca Mountain Project

Yucca Mountain is currently the focus of intense scientific and engineering investigations to determine its suitability as a the nation's repository of high-level radioactive wastes. A major part of these suitability studies focuses on the movement of water through the mountain, as most credible release scenarios involve transport of radionuclides by water flow. Before stating the key issues, it is worth reflecting on the major repository performance issues which ^{36}Cl and other environmental tracer studies may impact.

3.1.1 Importance of ^{36}Cl for Assessments for Travel Time and Total Water Flux

The critical hydrogeologic issues for the Yucca Mountain site relate to the flux of water which might pass through the repository horizon and downwards to the Calico Hills non-welded tuffs, and possibly to the water table. The breaching of the engineered containers would require some form of aqueous corrosion, which would depend on the available water flux and its localization. Once breached, water in sufficient quantity and of particular chemical composition would need to be present for significant releases to occur. Water flux thus is a key component in any assessment of Yucca Mountain's viability and ^{36}Cl measurements provide a method to define transit times for flow in the unsaturated zone.

3.1.2 Importance of ^{36}Cl for Understanding Flux Localization

In addition to the magnitude of the groundwater flux, the design of the repository and its safety assessments will require information on the nature of pathways for water movement. Localization of groundwater flow will limit the area of the repository which might be subjected to significant water flow and even raise the possibility of avoiding the high flux areas when designing the disposal facility. Knowledge of the pathway characteristics is also essential for assessments of geochemical retardation. Although a significant retardation potential exists particularly among zeolite minerals, water must move along pathways containing these minerals for this potential to be realized. ^{36}Cl has provided the YMP with a method that identifies locations of transport through fast travel pathways.

3.1.3 Transport to the ESF as an Analog for Transport through CHn

A final consideration of the importance of ^{36}Cl studies is the recognition that flow through the PTn may be an analog for flow through the CHn. Bomb-pulse ^{36}Cl has been the primary indicator of fast flow paths at the depth of the ESF. The site conceptual model asserts that water traveling along these pathways has been retarded little by the non-welded tuffs of the PTn.

Water source and velocity from the surface to ESF are not, by themselves, repository performance issues as surface-to-ESF pathway lies upstream of the repository level. Rather, the value of ESF observation may be that flow through the Paintbrush tuffs could serve as an analog for flow downstream of the repository through the Calico Hills non-welded unit. This downstream flow and transport would be the primary release pathway from the repository. Put in another way, uncertainties in the transport to the ESF could be translated into similar uncertainties in transport below the repository.

3.2 Statement of Peer Review Issues

Within the context of the assigned objectives and an understanding of performance concerns, the Peer Review Team identified five major issues:

1. Is the bomb signature real?(Section 3.3) -- Before considering any other issues of interpretation, the PRT decided to consider arguments that the threshold for discriminating bomb pulse from natural ^{36}Cl was appropriate or that the ^{36}Cl anomalies could have some other origin such as production from spallation of calcium.

2. Interpretation of $^{36}Cl/Cl$ Variability (Section 3.4) -- Can the anomalies in $^{36}Cl/Cl$ ratio be explained by variations in the source strength with time or mixing of waters with different $^{36}Cl/Cl$ ratios? Could variability be explained in terms of mixing with fluid inclusions from the rock?

3. Are ^{36}Cl Anomalies (or absences) an Artifact of Sampling and Analysis (Section 3.5) -- This issue addresses whether the selection of sites, sampling methods, or analysis procedures bias the ^{36}Cl measurements.

4. Integration of Isotope Programs (Section 3.6) -- Each component of the hydrochemical sampling program reflects a somewhat different feature or process in the flow system. Are the various isotope and environmental tracer programs properly integrated to achieve a consistent conceptual model of the flow system?

5. Integration with Conceptual and Numerical Flow Models (Section 3.7) -- Are ^{36}Cl and other environmental tracers being used effectively in the context of conceptual and numerical flow models? This issue considers (1) the use of models to test the plausibility of fast-path hypotheses, (2) the use of ^{36}Cl to constrain model estimates of water flux through the repository horizon, and (3) the value of bomb-pulse observation in the ESF for predicting flow below the repository through the CHn.

3.3 *Is the Bomb-Pulse Signature Real? (Anthony Muller)*

Fundamental to the value of ^{36}Cl, as it has been used by the YMP team, is the ability to distinguish waters with a bomb-pulse contribution of recharge from those which do not have such a contribution based on their $^{36}Cl/Cl$ ratios. This has been done by selecting a ^{36}Cl content, or more precisely, a ratio, above which samples are taken to show clear evidence of bomb water recharge. The question of whether waters with ^{36}Cl above the threshold ratio actually contain bomb-pulse recharge is fundamentally different from the question as to whether waters below the threshold (and therefore deemed non-bomb pulse) may contain such waters. The first question is addressed in this section while the second is addressed primarily in Section 3.2 of this review.

The reliability of a conclusion that a sample represents "bomb water" depends on three factors:

1. The conservatism of the threshold value for distinguishing bomb pulse,
2. The only significant sources of ^{36}Cl are natural atmospheric production and bomb tests, and
3. The likelihood that analytical results above the threshold are incorrect.

Issue 1, conservatism, and Issue 2, alternative ^{36}Cl sources are discussed in sections 3.3.1 and 3.3.2 respectively . Issues related to accuracy of analytical results are discussed in section 3.5.

3.3.1 Conservatism of the Threshold Value

The maximum ^{36}Cl content of non-bomb waters is relevant to evaluating the conservatism of the threshold value in two ways. First, the range of non-bomb values determines the "noise" from which the bomb "signal" must be distinguished. Second, the maximum ^{36}Cl value determines the likelihood that poorly mixed recharge water from the peak (or peaks) is actually contributing near the threshold.

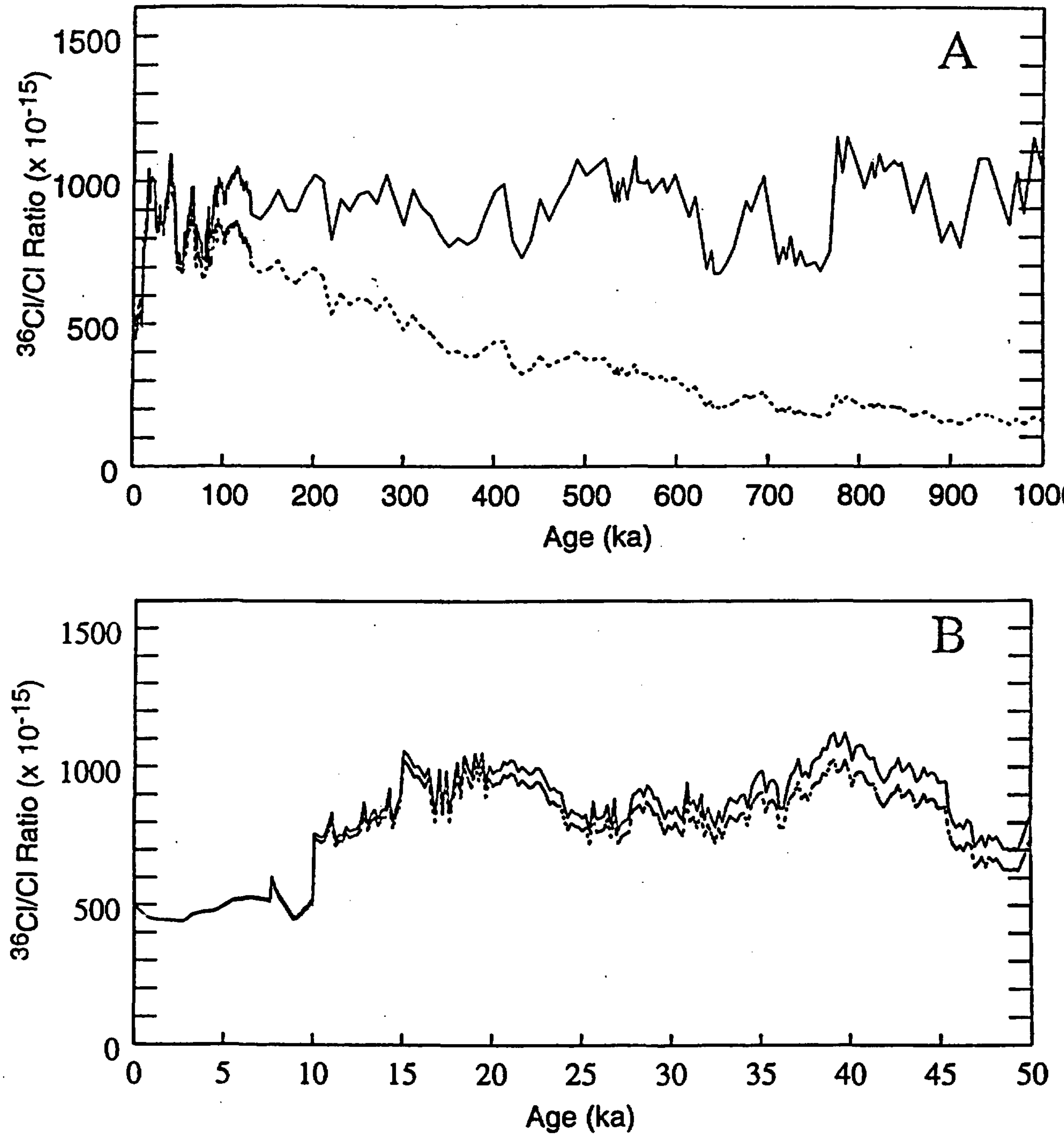
Reconstruction of the $^{36}Cl/Cl$ ratio in atmospheric waters is presented Figure 4 (3-1 of Fabryka-Martin et al., 1997). The reconstruction shows that atmospheric values have averaged about 500 and not exceeded 600×10^{-15} during the Holocene (the last 10,000 years). Decay-corrected values for the Pleistocene (10,000 50,000 years before present) average at about 800×10^{-15} and peak at about 1000×10^{-15}.

Fabryka-Martin et al. (1997) applied Chauvenet's criterion for identifying outliers on the independent data set two hundred forty-seven ^{36}Cl samples from Yucca Mountain. They concluded that at the 95% confidence level, 1250×10^{-15} represents the threshold between background and outlier (bomb) values. Note that this result implies no knowledge of the actual distribution of non-bomb ^{36}Cl ratios in precipitation, and is therefore an indirect method, assuming that the samples are from a single population with some outliers from another distinct population.

Even if the reconstructed peaks in the first analysis are underestimated by 25%, which is unlikely, the 1250×10^{-15} threshold would not be exceeded by the ^{36}Cl ratio of rainfall over the last 50,000 years.

Further, waters from different sources mix in the subsurface. The resulting chemical and isotopic signature is known to represent numerous recharge events over time and reflect the complex interactions of that mixed parcel of water between recharge and sampling. This is confirmed at Yucca Mountain by the complex chemistries and isotopic signatures observed. Even in the unlikely event that all the water from a sample were to have fallen at the atmospheric ^{36}Cl peak, it could not exceed 1250×10^{-15}. If the reconstruction were appreciably (25%) too low, peak ^{36}Cl near the threshold occurred only for short periods at three points over the last 50,000 years (about 15,000 years, 18,000 years, and 39,000 years before present). It is highly unlikely that recharge in the ground-water system could have remained effectively unmixed for that period.

For these reasons, the 1250×10^{-15} threshold is conservative in identifying bomb waters. It is very important to note, however, that the corollary is not



Source: Fabryka-Martin et al, 1977, Figure 3-1.

FIGURE 4
RECONSTRUCTION OF ATMOSPHERIC ^{36}Cl OVER PAST 50,000 YEARS
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necessarily true -- waters below that threshold cannot be claimed to be free of bomb ^{36}Cl and could well be a mixture of non-bomb and bomb waters (with proportions decreasing as the ^{36}Cl ratio goes from 1250 to below 500×10^{-15}). This issue is discussed further in section 3.4.5.

3.3.2 Sources of ^{36}Cl

A fundamental assumption in the interpretation of ^{36}Cl at Yucca Mountain is that all significant amounts of ^{36}Cl in the water are of either natural or weapons test origin in the atmosphere and that they have entered the ground-water system through infiltration. This assumption is examined in depth by Fabryka-Martin et al. (1997), who evaluate in detail local sources, deep subsurface production and surficial production in soils and calcite. While convincing arguments have been made to show these sources to be negligible with respect to natural and weapons test sources in the atmosphere, the possibility that surficial calcite is a significant source has recently been raised and was discussed by Dr. John Stuckless (USGS) in the presentations to the peer review panel.

One may perform a simple bounding calculation to determine if such a source, from spallation of ^{40}Ca by cosmogenic neutrons, is important. The calculation is presented in Appendix 4. The assumptions to the calculation are:

- the ^{40}Ca spallation at the elevation and latitude of Yucca Mountain results in about 152 atoms of ^{36}Cl/ year/ cm^2 of land surface (Fabryka-Martin, et al. 1997),
- cosmogenic neutron attenuation has a half-effect of about 155 g of soil per cm^2 which corresponds to a half-depth of about 100 cm in a soil with bulk density of 1.5 g per cm^3 of soil,
- soils average 10% calcite, which is the only reservoir of calcium available to spallation,

- average precipitation is on the order of 10 cm per year[1], and

- average total chloride concentration in precipitation is on the order of 0.6 mg of Cl per liter of water and that all this infiltrates.

Under these conditions, about 930 atoms of ^{36}Cl were calculated to be created per year by spallation of ^{40}Ca per of cm^2 surface. This is added to 10^{17} atoms of stable Cl already in the infiltrating water (from precipitation), and perhaps more stable Cl from soil dissolution. The resulting ratio of ^{36}Cl to total Cl is about 9.3×10^{-15}, which is insignificant with respect to the 1250×10^{-15} threshold discussed above.

Increasing the calcite fraction to an unrealistic 100% would result in a chloride isotope ratio of 93×10^{-15}, which is still negligible. Increasing the amount of either infiltration or of the Cl content of infiltration would reduce the fraction being produced from ^{40}Ca. Dissolution of ^{36}Cl-free chloride minerals in the soil zone would also reduce the value of the ratio.

For these reasons, spallation of ^{40}Ca appears to be an insignificant source of ^{36}Cl in infiltrating waters. The panel has drawn the overall conclusion that waters with observed $^{36}Cl/Cl$ ratios in excess of the threshold value of 1250×10^{-15} can be reliably considered to contain a significant proportion of bomb water. But, again, the panel warns that ratios less than the threshold do not necessarily indicate the absence of a bomb contribution. Determining the presence of several cosmogenic/weapon-test isotopes of different half-lives and with different geochemical behaviors can help distinguish the proportion of these two waters in future sampling.

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[1] This precipitation rate is low, but the assumption in conservative with respect to the effect on ^{36}Cl from calcium spallation as discussed two paragraphs below.

3.4 *Interpretation of $^{36}Cl/Cl$ Variability (Jack Cornett)*

3.4.1 Conceptual Models of ^{36}Cl Variability

This section of the report addresses the sources of variability among measurements of $^{36}Cl/Cl$ ratios.

The $^{36}Cl/Cl$ ratios measured in samples from Yucca Mountain range from about 50 to 4100×10^{-15} (Fabryka-Martin et al., 1997). The $^{36}Cl/Cl$ ratio in individual samples can be interpreted by two general and different conceptual models (or combinations thereof):

1. The different ratios result from temporal variations in the input of ^{36}Cl and Cl into Yucca mountain and/or subsequent radioactive decay, or

2. The different ratios result from mixing of a bomb pulse input with different amounts of dilution by Cl and ^{36}Cl in another reservoir(s). The mixing and dilution could take place in any of several ways:

   - natural mixing of fast path water with water from non-fast path fracture sources

   - natural mixing of fast-path fracture waters with matrix pore water that entered the flow system before the bomb-pulse era,

   - artificial mixing with drilling and construction waters (whose effects on sampling should be minimized by recognizing bromide tracers in the introduced waters),

   - artificial mixing during sample collection and/or sample preparation of fast-path fracture Cl and ^{36}Cl with matrix pore waters, that are not bomb-pulse in origin, or with primary rock chloride in rock fluid inclusions.

3.4.2 Sources of ^{36}Cl

The inputs of ^{36}Cl and Cl have been reviewed by Fabryka-Martin et al. (1997). At Yucca Mountain, there are three sources of potential importance:

1. ^{36}Cl fallout from the atmosphere that is produced by natural processes,

2. ^{36}Cl fallout from the testing of nuclear weapons in the Pacific years.

3. ^{36}Cl fallout from the testing of nuclear weapons at the Nevada Test Site during the past ~50 years. This source is not considered to be distinct from the second source, that is, global fallout.

A fourth source, the leaching of ^{36}Cl produced in calcite residing in near surface soils, was reviewed carefully by the PRT (See section 3.3). Calculations presented in Appendix 4 show that the $^{36}Cl/Cl$ ratio of leachate from calcite is very low relative to inputs from both natural and bomb fallout, and the amount of stable Cl in calcite is also low. As a result, this source does not influence $^{36}Cl/Cl$ ratios in zones below the calcite rich soil and it will not strongly influence bulk soil samples that contain ^{36}Cl from weapons test fallout.

3.4.3 Chlorine-36 from Global Fallout as a Result of Nuclear Weapons Tests

^{36}Cl fallout from the testing of nuclear weapons in the atmosphere during the past ~50 years was much greater than natural inputs (Synal et al., 1990). This ^{36}Cl is still present in soils and is slowly moving through the biosphere. This peer review report concurs with the YMP team's conclusions that:

- The chloride in the soils at Yucca Mountain includes ^{36}Cl from weapons test fallout.

- Some deeper samples of groundwater from the saturated zone, pore water from the unsaturated zone, and rock extract from Yucca Mountain contain ^{36}Cl in excess of natural concentrations. Samples that contain more than about 1250×10^{-15} contain a component of Cl that has infiltrated into Yucca Mountain during the past 50 years. The ^{36}Cl/Cl ratios in these latter samples reflect the bomb pulse signal.

- The subsurface distribution of ^{36}Cl can be attributed to flow through fast pathways.

3.4.4 Natural ^{36}Cl Inputs

The natural flux of ^{36}Cl and Cl may have varied temporally due to changes in ^{36}Cl production as the cosmic ray flux to the earth's surface varies and due to smaller changes in stable Cl flux attributed to climatic changes. The variations in fallout have been measured using ^{36}Cl and several other isotopes (^{14}C, ^{10}Be, ^{26}Al), and they are broadly consistent with models that describe changes in the strength of the earth's magnetic field (Plummer et al., 1997). Members of the YMP have done a very thorough job of reconstructing the temporal variation in ^{36}Cl/Cl fallout ratios over the past 50,000 years. This peer review report supports their conclusion that two periods with significantly different <u>natural</u> fallout existed, namely:

1. ^{36}Cl/Cl fallout varied around 500×10^{-15} during the period from the present day to about 10,000 years before present, and

2. ^{36}Cl/Cl fallout ranged from about 600 to 1200×10^{-15} from 10,000 years before present to 50,000 years before present (Fabryka -Martin et al., 1997).

Radioactive decay of ^{36}Cl (half life 301,000 years) could further alter the ^{36}Cl/Cl ratio but this is only significant in samples that have been isolated for at least 50,000 years. This appears unlikely in the Yucca Mountain setting (Bodvarsson et al., 1997; Robinson et al., 1997).

3.4.5 Influence of Mixing and Dilution

One way to interpret variations in ^{36}Cl/Cl in samples from the unsaturated zone and from the saturated zone is to ascribe this variation to temporal variations in input of ^{36}Cl. A second way to interpret the different ^{36}Cl/Cl ratios in various samples is to postulate that they result from mixing of different Cl reservoirs and/or dilution of the ^{36}Cl input with ^{36}Cl and Cl from various sources. The additional sources could be derived from fracture pore water, matrix pore water, or from the rock itself (Andrews and Fontes, 1993). Since the $^{36}Cl/Cl$ ratio of the rock is very low ($\sim 50 \times 10^{-15}$, Fabryka-Martin et al. 1997) mixing with Cl from the rock matrix can dilute ^{36}Cl input from either natural sources or bomb test fallout. Figure 5 plots the strong inverse correlation between the $^{36}Cl/Cl$ ratio and the concentration of total Cl measured in the extract from the ESF samples. This figure illustrates that almost all of the samples with $^{36}Cl/Cl$ ratios >1000 have low Cl concentrations, which are less than 1 ppm.

Although YMP -taff have carefully designed their sampling program to minimize the component of rock ^{36}Cl in their samples, the hypothesis that ^{36}Cl is diluted by some Cl from the rock or from the pore water in the rock matrix cannot be dismissed.

The observations that are consistent with the dilution hypothesis include:

1. A reservoir of Cl exists in the rock because the mass of stable Cl in the rock is greater than that in the pore water. Leaching of about 10% of Cl from the rock in a 1-kg sample could dilute the $^{36}Cl/Cl$ ratio in the pore water by about 9 times and reduce the $^{36}Cl/Cl$ ratio from 5000 to 600×10^{-15}.

2. There is an inverse correlation between the $^{36}Cl/Cl$ ratio and the concentration of Cl leached from the ESF rock samples (Lehmann et al., 1993).

3. Dilution of ^{36}Cl from the atmosphere with Cl mixing with matrix pore-water chloride. Similar mixing effects have been suggested to occur in other geochemical systems such as ^{14}C (Sonnenthal and Bodvarsson, 1997).

4. The analysis of Sr isotope ratios in the pore water at Yucca Mountain indicates that these ratios have been affected by rock-water interaction (Marshall et al., 1998) so it is reasonable to argue that rock-water interactions could also influence $^{36}Cl/Cl$ isotope ratios.

5. Some samples that appear to contain bomb tritium also have ^{14}C ages that are far older than 50 years (Yang et al., 1996). This is consistent with mixing of waters (and the salts) of different ages.

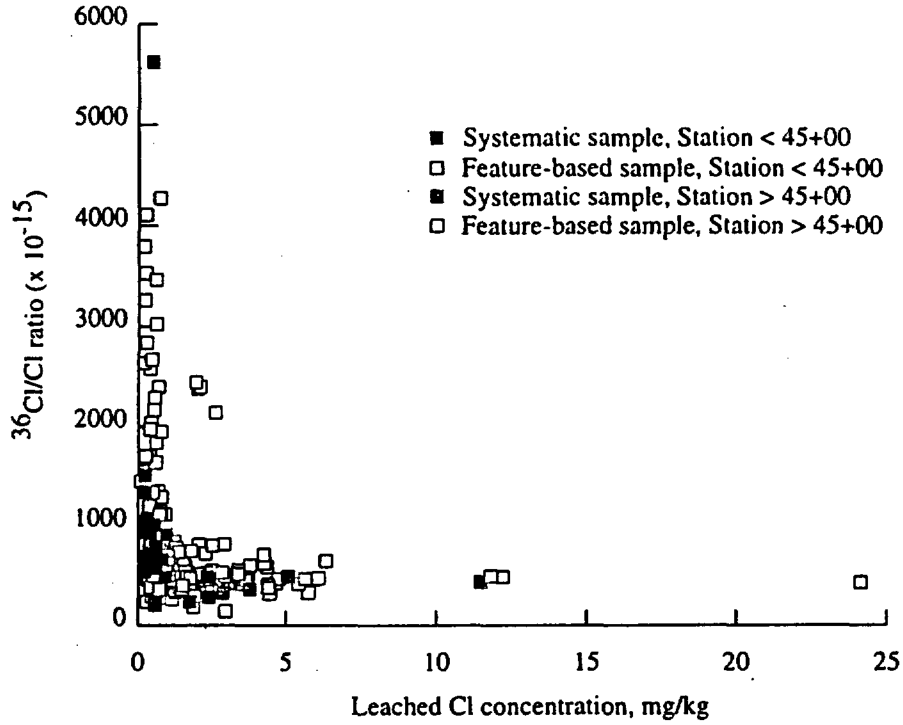

Figure 1. 36Cl/Cl ratios measured for ESF rock samples, as a function of location (north vs. south part of ESF) and sample type (systematic vs. feature-based)

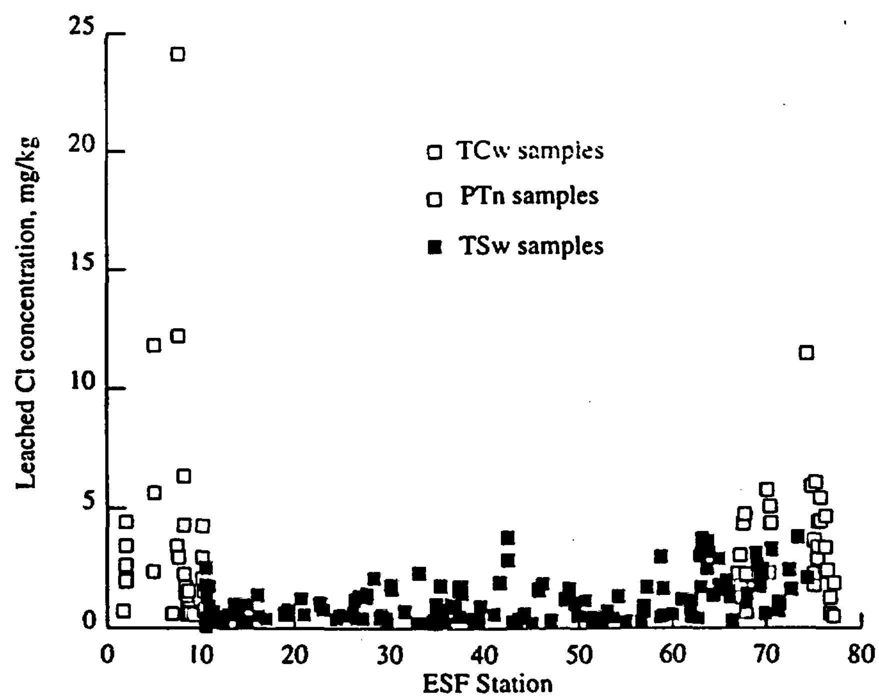

Figure 4. Leached Cl concentrations measured for ESF rock samples, as a function of location and lithology

Includes all ESF data available as of 22-Nov-98

Source: E. Sonnenthal, ^{36}Cl Peer Review Presentation.

FIGURE 5

RELATIONSHIP OF ^{36}Cl TO TOTAL Cl IN ESF PORE SALTS

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The observations that suggest that dilution is <u>not</u> important include:

1. The samples of rock were not crushed and ground and this will minimize the leaching of Cl from fluid inclusions in the rock.

2. The lowest $^{36}Cl/Cl$ ratios measured in samples from the ESF were several fold greater than the $^{36}Cl/Cl$ ratio that would be expected if most of the Cl and ^{36}Cl were derived from the rock rather than from the atmosphere.

3. Fluid inclusions are very stable under the conditions used to extract the Cl from the rock samples.

4. Variability in duplicate ^{36}Cl analyses is low relative to that to be expected following erratic leaching of fluid inclusions.

Variable amounts of dilution could bias the interpretation of the $^{36}Cl/Cl$ ratio data. For example, the observation that none of the ESF samples from the south ramp have $^{36}Cl/Cl$ ratios with a bomb pulse signature could be explained by more dilution in samples from that region of the ESF. To examine this question further the PRT requested a series of analysis which were performed by J. Fabryka-Martin using the ESF $^{36}Cl/Cl$ data set. (Fabryka-Martin, 1998 February, Pers. Comm).

These analyses examined a series of <u>potential</u> biases and variations in the amount of dilution that might occur as atmospheric $^{36}Cl/Cl$ ratios are diluted by mixing with rock, pore water, or drill water $^{36}Cl/Cl$. This analysis allows us to reject the hypothesis that the spatial variations in $^{36}Cl/Cl$ (e.g. the different ratios of $^{36}Cl/Cl$ in the North versus South ESF) can be explained by variable amounts of dilution in different parts of the ESF (Figure 3). Based on this analysis, it would appear that:

- On average, the amount of Cl leached from the systematic and feature based samples is not significantly different,

- There is no systematic difference in the amount of Cl leached from the systematic and feature based samples north or south of ESF station 45 so different amounts of dilution cannot explain the different $^{36}Cl/Cl$ ratios in the areas of the ESF,

- The Br/Cl ratio and the amount of Cl attributed to drill/wash water does not vary systematically among ESF stations or the north versus south regions,

- Stratifying the data by lithology does not change these conclusions,

- Several of the TCw samples from the south ramp area have low concentrations of leached Cl; but none of the samples that have been analyzed to date, (additional samples have been collected for analyses) has a high $^{36}Cl/Cl$ ratio ($>1000 \times 10^{-15}$), and.

- Omitting about 4 samples from Tiva Canyon tuff in the north ramp, all ESF samples with $^{36}Cl/Cl$ ratios greater than 1000×10^{-15} have concentrations of leached Cl that are less than 1 ppm. This could be used as a criteria to select samples for ^{36}Cl analysis.

The *simplest* interpretation of $^{36}Cl/Cl$ ratios at Yucca Mountain assumes that there is negligible interaction with the rock and all of the variations in $^{36}Cl/Cl$ ratios can be attributed to variations in the input of ^{36}Cl from the atmosphere (assuming that decay is not important). However one may conclude that:

- Samples with $^{36}Cl/Cl$ ratios less than 1250×10^{-15} may contain a component of bomb pulse ^{36}Cl that is less than 50 years old.

- Samples with elevated $^{36}Cl/Cl$ ratios provide conclusive evidence of recent waters but the low ratios do not imply an absence of bomb-pulse ^{36}Cl .

- Additional effort is needed to evaluate the mixing and dilution hypothesis so that the ^{36}Cl distribution can be interpreted in an unambiguous manner. The

YMP should investigate ways to define the amount of dilution of the $^{36}Cl/Cl$ ratio. This is especially important for samples that have less than $^{36}Cl/Cl$ atom ratios of 1200×10^{-15}.

3.5 Are ^{36}Cl Anomalies an Artifact of Sampling and Analysis? (3.5.1: Thomas Doe; 3.5.2 - 3.5.7: Jack Cornett)

The YMP interprets Cl^- and ^{36}Cl with the assumption that the chloride present in infiltrating water has an atmospheric source. These data are then used to evaluate models of water and radionuclide transport through the Yucca Mountain flow system. The major isotopes of Cl, $^{35}Cl/^{37}Cl$ and ^{36}Cl, are present ubiquitously in nature (Bentley et al., 1986), and the Cl (and ^{36}Cl) measured in a sample collected at Yucca Mountain could come from several sources:

- atmospheric inputs,
- leaching from fluid inclusions or Cl rich minerals in the rock,
- drill water or wash water used during the ESF or drilling operations
- mixing of Cl from different reservoirs (fracture and matrix water)
- a mixture of the first four sources.

In an ideal world, samples of *atmospheric* Cl in infiltrating water could be collected and analyzed. However there is insufficient infiltration to do this and the Cl from precipitation is not distinct from other sources. As water infiltrates through Yucca Mountain, it may pick up Cl from the rock, or isotope exchange may occur (Lehmann et al., 1993). As a result, it is important to evaluate the methods to ensure that the amount of atmospheric Cl can be determined from the total Cl or ^{36}Cl measurements. For example, these non-atmospheric sources, such as fluid inclusions of volcanic origin, may have distinctive stable Cl isotope ratios, $^{35}Cl/^{37}Cl$.

This section reviews the selection of sites, the sampling protocols used to collect samples for Cl and ^{36}Cl measurements, and the analytical techniques that were used to perform the measurements.

3.5.1 Collection of Samples for Cl and ^{36}Cl

If the project's conceptual model of fast-path flow and ^{36}Cl occurrence is correct, then sampling bomb-pulse ^{36}Cl should be a very difficult task. First, success in sampling would require finding a fast pathway fracture among a large populations of fractures that may not be fast pathways. Second not all fast pathways will carry the bomb pulse ^{36}Cl -- only those fast pathway fractures that are part of fracture networks connected to the surface via breach in the PTn. The chloride in the sample must be dominantly from the bomb pulse era and not reflect any significant mixing with older or younger waters, despite the likelihood that the fast pathways may have carried many similar pulses of water prior to the arrival of the bomb-pulse bearing flows. Finally, the sample must not contain significant amounts of non fast-path chloride from the rock matrix.

Even given the presence of bomb pulse ^{36}Cl, sampling it should be viewed as a major challenge. Systematic samples, taken without regard to geologic features, should be viewed as highly unlikely to contain any bomb-pulse chlorine. Even feature specific samples would require great care in collection to maximize the likelihood of selecting the fast-pathway fracture, assuring it has not been affected by construction waters, and avoiding sampling too much material away from the fracture surfaces.

Distinguishing characteristics for fast-path fractures have not been identified (other than by the measurement of $^{36}Cl/Cl$ ratios) and mineralogic correlations have proven negative (Fabryka-Martin, et al. 1997, Ch. 7). Furthermore, how chloride resides in the rock (i.e. evaporated onto fracture surfaces, in nearby matrix pores, or widely distributed in the matrix) is also not known. Despite careful and systematic selection of sample sites, until the distinguishing features of fast pathways can be identified and the nature of chloride occurrence is understood, sampling will be a hit-or-miss activity.

Several studies are underway which may help identify flowing fractures. First, efforts to isolate underground chambers and allow their air to build to 100% humidity should be very useful for revealing seeps that had been evaporated by the ventilation system. Second, in situ percolation and seepage tests should help considerably in identify potential fracture conductors. Finally , experiments aimed at finding and accounting for construction water from the ESF may be very instructive.

In summary, future sampling should focus upon features that are likely to be hydrologic pathways. In addition, multiple isotopic analyses should be performed at each sampling site (see Section 3.6). These recommendations are based upon the view that single measurements are always open to more interpretation than those of multiple techniques and recognition that fracture flow will be through features. Furthermore, the methods used to collect samples should be clearly documented so that a reproducible protocol is used at all locations.

3.5.2 Extraction of Cl and ^{36}Cl

There is insufficient water available in the majority of samples for fluids to be extracted for ^{36}Cl analysis. As a result most $^{36}Cl/Cl$ analyses have been made on Cl collected from soil or rock leached from the rock by making a slurry of distilled water and rock fragments. This has been an expeditious method; however a few selected tests would help (see below) to rigorously eliminate alternative interpretations of the extractions.

To minimize the leaching of Cl from the fluid inclusions and minerals in the rock, fragments of rock rather than finely ground rock flour were used in the analysis. Variable amounts of leaching of Cl from fluid inclusions could bias the results. In future work, the leaching procedure should be formalized to extract only those samples from a given particle size. This will ensure that small amounts of rock flour do not introduce relatively large amounts of Cl.

Recently some pore water samples have been used for stable Cl analysis. This is a commendable approach and more samples of pore water should be analyzed by pooling samples to obtain sufficient quantities of material for analysis. Furthermore, at a few selected sites where Cl and ^{36}Cl have been measured previously, water should be artificially injected into fractures and collected as it drains under gravity. These water samples should be analyzed for Cl and for ^{36}Cl, and these results compared to the previous measurements using laboratory-derived rock leachate. Additional sites should be chosen where ^{36}Cl from weapons test fallout was found and from sites where it was not found. Confirmation of the earlier results using this test will ensure that the dilution of weapons-test fallout by Cl from fluid inclusions or from matrix pore water, which is not associated with fast-path fractures, is not biasing the $^{36}Cl/Cl$ measurement results.

3.5.3 Analysis Methodology for Cl-

Stable Cl- has been analyzed by ion chromatography. This is an appropriate and very reliable method, particularly with continued inter-laboratory comparisons and other QA/QC activities.

3.5.4 Analysis Methodology for ^{36}Cl

^{36}Cl has been measured using accelerator mass spectrometry (AMS) (Phillips et al., 1986). AMS is very well established (Andrews et al., 1994). It is the only reliable technique for measurement of ^{36}Cl in the range of concentrations found in the environment at Yucca Mountain. This is an appropriate and a very reliable method. The precision (1 std) and accuracy of most analyses is about 5% (Andrews et al., 1990), which is adequate for the YMP program.

3.5.5 QA/QC Activities

YMP staff have examined the blanks, reproducibility, and efficiency of their extraction and measurement procedures very thoroughly (Fabryka-Martin, 1998). These tests are acceptable and the current technique used to extract Cl and ^{36}Cl is suitable to identify samples that may contain a component of ^{36}Cl from weapon's test fallout. The YMP should continue submitting blind samples to the analytical laboratories and supports their involvement in broader QA/QC activities.

3.5.6 Corrections to Cl and ^{36}Cl measurements

In some samples, a fraction of the Cl extracted from samples of drill chips or ESF rocks could contain Cl from drilling/washing fluids (Fabryka-Martin et al., 1997). Cl in these fluids is a potential source of contamination of the natural samples. YMP plan to label all introduced fluids with Br is an outstanding initiative. The program of Br measurements in the leachates is appropriate to determine the amount of drill or wash water contamination, and the YMP staff have done a very good job of correcting for this potential contamination problem.

Cl obtained from fluid inclusions has very low $^{36}Cl/Cl$ ratios. The extraction of this Cl could result in an erroneous interpretation of measurements with low $^{36}Cl/Cl$ ratios. The YMP staff should test the use of $^{35}Cl/^{37}Cl$ isotopic ratios as a means to measure, in the leachate, the proportions of Cl that have been derived from the volcanic rocks and from the atmosphere. If a clear distinction in isotope ratios is found between the two sources, then additional measurements of pore waters also would be justified.

3.6 Integration of Isotope Programs (Bridget Scanlon)

Integration of various isotope programs is essential to obtain a comprehensive understanding of unsaturated flow processes at Yucca Mountain. Each isotope has some limitations with respect to inferring travel time or ages; however, combining data from many isotopes to obtain a consistent conceptual model of the flow system is important.

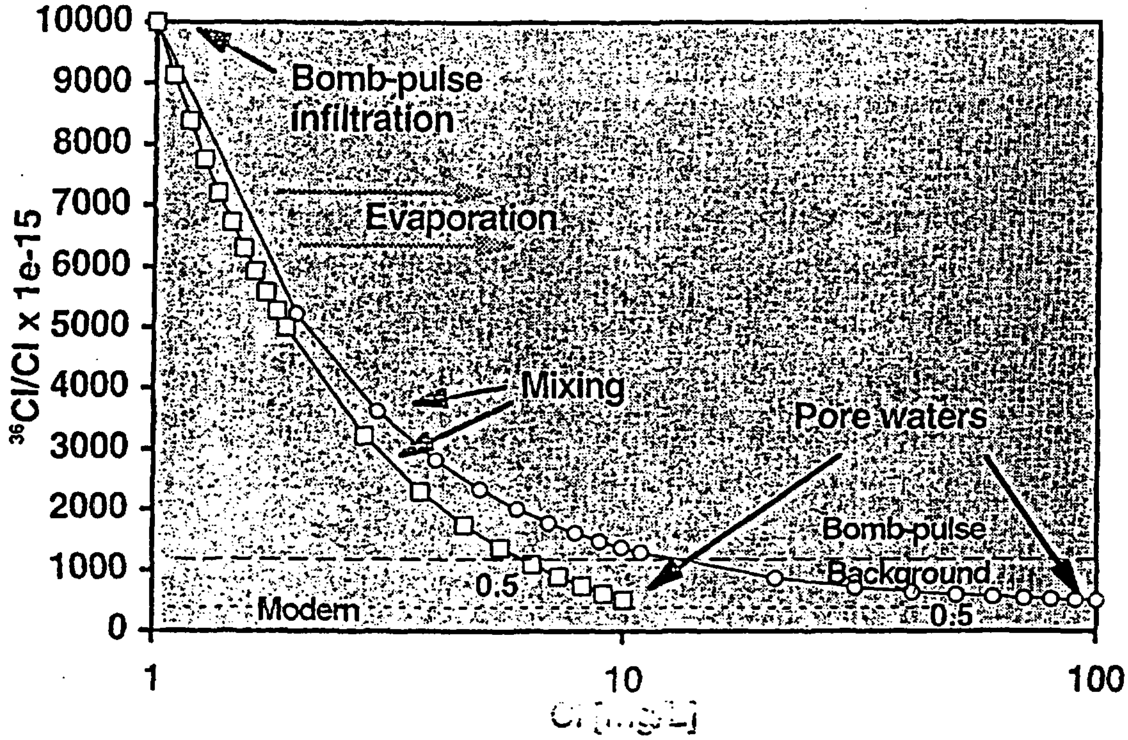
3.6.1 Limitations of $^{36}Cl/Cl$ Analyses

Much of the work at Yucca Mountain has focused on measurement of ^{36}Cl. High $^{36}Cl/Cl$ ratios provide unequivocal evidence of bomb pulse water at the sampled depths. Disadvantages with the use of $^{36}Cl/Cl$ ratios result from measurement of a ratio which is affected by chloride concentrations in the matrix and the large volumes of rock sample that need to be processed to obtain sufficient chloride for analyses. Both of these factors, i.e. measurement of ratios and large sample volumes, result in dilution of the bomb pulse signal. Therefore, the absence of high $^{36}Cl/Cl$ ratios does not necessarily indicate that there is no component of bomb pulse water in the sample. Figure 6 illustrates the dilution effect.

3.6.2 Analysis of Tritium

Bomb pulse tritium data play a key role in testing hypotheses used to explain the $^{36}Cl/Cl$ data, particularly for samples between 500×10^{-15} and 1250×10^{-15}. This ratio range can be explained either as unmixed Pleistocene water or as bomb-pulse waters diluted with recent non bomb-pulse water.. Unlike $^{36}Cl/Cl$ where the ratio is greatly affected by dilution, tritium is not as affected by mixing and dilution. Mixing of bomb pulse $^{36}Cl/Cl$ with pre-bomb water results in an exponential decrease in the $^{36}Cl/Cl$ ratio with increasing Cl concentrations in the matrix whereas with tritium the mixing relationship is linear. Tritiated water is volatile and therefore can move in the liquid and vapor phases. Therefore, tritium samples may be contaminated with air from the tunnel or drilling activities. Whereas the bias for $^{36}Cl/Cl$ sampling of potential bomb-bearing water is

EFFECTS OF MIXING AND EVAPORATION ON 36Cl/Cl



Source: E. Sonnenthal, ^{36}Cl Peer Review Presentation.

FIGURE 6
EFFECTS OF MIXING AND EVAPORATION ON ^{36}Cl / Cl RATIO
YUCCA MT/PEER REVIEW/NV

PROJECT NO. 974 1009.006.001 DRAWING NO 77202 DATE 3/6/98 DRAWN BY EA generally towards false negatives, the opposite is probably true for tritium where potential contamination of tritium samples with drilling air and air from the tunnel could result in false positives. The YMP's investigators have taken commendable steps to minimize contamination effects in tritium measurements. For example, using SF_6 as a drilling tracer by the YMP (Yang, personal communication, 1998) is an excellent measure, but not a complete solution to the problem, as the tracer is considerably less soluble in water than tritium. To evaluate the potential of obtaining false positives, procedural blanks should be used in the field during sample collection for tritium. Such blanks could consist of rock core that had previously been oven dried and then wetted with dead tritiated water. This would provide a blank for liquid water tritium samples and should be subjected to the same procedures in the field as the regular samples. A sufficient number of blanks should be used to allow estimation of uncertainties in blank values.

Tritium results from Alcove 2 in the ESF indicate unequivocal bomb pulse water with tritium values several times the detection limit (20 to 144 TU). These data are consistent with the presence of bomb pulse ^{36}Cl. High tritium levels have also been measured in samples from surface based boreholes (NRG-6 [≤145 TU], NRG-7a [≤30 TU], SD12 [≤30 TU]). Interpretation of the remaining tritium analyses from the ESF is equivocal. Analysis of tritium by direct liquid scintillation without pre-enrichment resulted in a high detection limit (2 standard errors, 8 TU) (Patterson, pers. comm.); therefore, levels ≤16 TU cannot be interpreted to be bomb pulse at the 95% probability level.

It is important to evaluate tritium relative to $^{36}Cl/Cl$ ratios. Although many of the samples have been analyzed from the north ramp, samples are available for analysis of the south ramp. These samples should be enriched prior to analysis and gas proportional counting should be evaluated because it may result in lower detection limits than liquid scintillation. Tritium/Helium ingrowth should also be investigated although the small sample volumes may require inordinately long times to decrease standard errors using this analysis procedure. Samples should be combined where necessary to increase sample volume.

The ambiguous interpretation of many existing tritium measurements will continue to impact negatively the entire hydrogeologic program in the unsaturated zone. The YMP should rigorously evaluate all existing tritium measurements to define uncertainties in the measurements and to define the lower limits of detection in the analyses.

In addition to extraction of water from core samples for tritium analyses, tritium can also be analyzed in water condensed from gas samples. Such an approach was used by Yang et al. (1985), however it was applied to a 30-ft interval of UZ-1 and did not clearly sample specific fractures. It is difficult to sample preferred pathways or fractures directly because most tracers require water extraction from a large volume of rock and such samples basically reflect the resident averaged concentration. Evaluation of preferential flow requires sampling of the flux averaged concentration which is extremely difficult or impossible in most cases because preferred pathways are dry most of the time.

Tritiated water is volatile; therefore, water can be condensed from gas samples extracted from fractures. There are limitations in sampling gases from fractures, such as contamination with air from the tunnel boring machine. However, this sampling procedure should be evaluated because it may provide more a more direct sampling of the preferred pathways than water extraction from cores. Procedural blanks should also be used to evaluate any biases or problems associated with sampling and analyses.

3.6.3 Analysis of Other Isotopes

Information on stable isotopes of oxygen and hydrogen may prove useful in discriminating old water, that entered the system during the Pleistocene when the climate was cooler and wetter (less enriched in stable isotopes), from young waters (Holocene, warmer and drier) to test the hypothesis that the low $^{36}Cl/Cl$ ratios represent old water (> 10,000 years). The database on stable isotopic compositions of rainfall is large and can be used to distinguish present-day winter versus summer precipitation relative to Pleistocene recharge. Results of previous studies on the Nevada Test Site that interpreted deeper samples as representing Pleistocene water can be used to guide the interpretation of the results at Yucca Mountain. Although some have suggested that depleted stable isotope levels reflect recharge from higher elevations, numerical modeling and other studies indicate that recharge from higher elevations is unlikely as most water above the Calico Hills non-welded tuff at Yucca Mountain mainly moves vertically.

Some advantages with use of stable isotopes include the small amount of water required for analyses (~ few milliliters) and the low standard errors for these analyses. The small sample size requirements would allow more discrete feature based sampling to be conducted at Yucca Mountain. Different extraction procedures have been used at Yucca Mountain to obtain water for stable isotope analyses including compression and vacuum distillation. Care should be taken when analyzing clay rich or zeolitized samples because vacuum distillation could result in isotopic fractionation during water extraction (Yang et al., 1996).

^{14}C and $^{12}C/^{13}C$ ratios of dissolved inorganic carbon in the pore water should also be measured. Although carbon isotopes may be affected by vapor-phase mixing, they nonetheless provide data which corroborate the $^{36}Cl/Cl$ ratios to evaluate flow and transport. Previous studies by Yang et al. (1996) demonstrate the potential uses of such data in evaluating ages of pore water. Old ^{14}C ages (> 10,000 years) would support the hypothesis that $^{36}Cl/Cl$ ratios between 500 and 1250×10^{-15} reflect older Pleistocene water.

$^{87}Sr/^{86}Sr$ ratios in water reflect water rock interactions and have been used to evaluate water movement through the unsaturated zone. Whereas ratios of $^{87}Sr/^{86}Sr$ have been interpreted to represent differing residence times in the PTn unit, $^{87}Sr/^{86}Sr$ ratios may be capable of differentiating waters that have traveled fast, non-reactive paths from those which have followed slower, more reactive paths. Water that travels a fast path through the PTn should have lower $^{87}Sr/^{86}Sr$ ratios more reflective of the TCw units overlying the PTn than higher Sr ratio waters that had more opportunity for rock interaction in the PTn. Measurements of $^{87}Sr/^{86}Sr$ should be continued to provide corroborative data to test alternative conceptual models of $^{36}Cl/Cl$ ratio distributions, and it would be useful to compare $^{87}Sr/^{86}Sr$ ratios between samples with and without bomb pulse $^{36}Cl/Cl$ signatures .

Analysis of Technetium-99 may prove useful in evaluating the presence or absence of a component of bomb pulse water. ^{99}Tc is an anthropogenic tracer with very low background levels. Preliminary studies demonstrated the technical feasibility of using ^{99}Tc (Fabryka-Martin et al., 1997). However, sample preparation is complicated and should be thoroughly tested prior to using this tracer. Measurements of ^{99}Tc should be conducted initially in the soil zone and at selected sites in the ESF where ^{36}Cl measurements were made previously.

In addition to isotopic measurements, stable chloride concentrations in pore water should also be measured to evaluate the possibility that low $^{36}Cl/Cl$ ratios reflect damping with high matrix chloride concentrations. Such analyses should be routinely done in all samples. YMP investigators should also test the use of $^{35}Cl/^{37}Cl$ ratios to distinguish between chloride from different sources such as the atmosphere and rock matrix.

3.7 Interpretation of the $^{36}Cl/Cl$ and other Environmental Isotope Data in the Context of the Conceptual and Numerical Flow Models (Thomas Doe)

The modeling studies for the Yucca Mountain site serve several purposes. As discussed in section 3.1, the modeling goals include:

- Testing of the consistency of fast past hypotheses with currently accepted conceptual models and hydraulic parameters,
- Estimation of total water flux across repository horizon,
- Simulation of flow and transport below the repository, and
- Simulation of travel time.

3.7.1 Simulation of ^{36}Cl Fast Paths

Dual permeability formulations of the site numerical models have reproduced fast pathways for ^{36}Cl transport (Fairley and Wu, 1997; Fabryka-Martin et al, 1997). As discussed in section 2, the criteria for creating a fast pathway include connection to fracture paths across the PTn, low surficial storage, and sufficient local infiltration to sustain flow. As Fairley and Wu point out, the modeling uses "stock" parameters, that is, there were no major or unrealistic adjustments of the hydraulic parameters to create such pathways.

This modeling does not prove the fast-path hypotheses, but it provides confidence that the fast pathways and the bomb-pulse measurements are real -- or at least more likely to be real than if the models had required severely large deviations of properties or geometries to produce the results.

The numerical models reproduce in general the spatial pattern of bomb pulse ^{36}Cl/Cl, particularly for the Sundance Fault (figure 3). Other faults, however, do not appear to relate clearly to ^{36}Cl/Cl ratios. Furthermore, higher ^{36}Cl/Cl ratios in the North ramp rather than the South ramp is contrary to an expectation that a thinner PTn in the south should lead to more bomb pulse ^{36}Cl/Cl ratios in that area. Sonnenthal and Bodvarsson (1997, Ch.15) point out that the model predicts higher total chloride concentrations near some faults such as the Ghost Dance fault which can mask the bomb pulse signatures (Figure 6).

There is not a major discrepancy between the model predictions of fast-path occurrence in the ESF and the measurement results. There are numerous reasons why bomb pulse ^{36}Cl /Cl should be difficult to sample, as discussed in section 3.5. Furthermore, there are reasons to expect tracers to move significantly from their source at the PTn. Fracture network geometry may play a role in moving tracers away from faults which breach the PTn and into fractures that do not lie in fault zones or directly beneath fault intersections with the PTn..

The programs to predict and sample $^{36}Cl/Cl$ are an essential part of future studies in the planned E-W drift. These programs will be particularly useful for testing conceptual models of the role of PTn by sampling beneath areas where that unit is absent. The predictions of other anomaly locations should be viewed stochastically rather than deterministically due to uncertainty in the exact nature of fracture flow paths in the TSw. Furthermore, given the dilution and sampling problems discussed in sections 3.4 and 3.5, the absences of $^{36}Cl/Cl$ anomalies in the measurements in the E-W drift should be treated carefully with respect to their use in validation.

3.7.2 Flux Estimation Using Environmental Isotopes and Other Complementary Lines of Evidence

Unfortunately, there is no single method for directly measuring water flux at Yucca Mountain. Thus, computer simulations, constrained by calibrations to measurements and observations from a variety of physical and chemical sources, have been the primary tool for flux assessment. Unsaturated flow models of the site have been developed by two major groups, Lawrence Berkeley National Laboratory (LBNL) and Los Alamos National Laboratory (LANL). These groups have adopted somewhat different numerical approaches -- integrated finite difference versus finite element -- however, the overall modeling approach has been very similar in terms of properties and processes. The scope of the modeling studies, the scientific quality, and the thoroughness of the work produced by both modeling groups are impressive.

Modeling the Yucca Mountain site is a significant technical challenge. Due to the scale and complexity of unsaturated properties and processes, the unsaturated zone models, by necessity, use little in the way of field measurements as direct input parameters. Critical parameters such as the infiltration rates near the surface (Flint et al., 1996) and the hydrologic parameters (Banduragga and Bodvarsson, 1997) themselves rely on sophisticated numerical models which are calibrated to limited data bases.

Finding unique, constrained answers using numerical models also has been a major challenge. This non-uniqueness comes in large part from a reliance on inverse modeling for the site model input parameters. In the past, numerical models of fluid flow have proven to be highly adaptive to fundamental changes in the conceptual model of the site. This flexibility indicates the difficulties of constraining model calculations. Major validation criteria, such as matrix saturation profiles and perched water locations, have been met both by single permeability and dual permeability models, the dual permeability models having been introduced to accommodate the possible presence of fast pathways. For example, the application of dual permeability models allowed an increase in the infiltration estimates from the 0.1 mm/year rate used in Wittwer et al.'s (1995) single porosity model to about 5 mm/year with a dual permeability model (Robinson et al., 1997), while still matching the matrix saturations. As noted in Fabryka-Martin, et al. (1997, Section 8.1), "...minor changes in hydraulic parameters lead to different flow distributions in the PTn matrix and fractures, that in turn affect predictions of the locations at which fast paths exist...".

The fracture properties, and particularly the fracture-matrix interaction terms, are uncertain (Doughty and Bodvarsson, 1997, Ch. 5, p. 5-29). As with the previous single permeability models, the dual permeability models depend heavily on calibration to the matrix saturation profiles. Given the lack of constraint on the fracture-matrix interaction terms, a wide range of fracture properties can be inverted and still satisfy the calibration criteria. Ideally, saturation data for the fracture network would considerably improve the calibration, however, these would likely be extremely difficult if not impossible to obtain.

In addition to the uncertainties in the fracture parameters, flow processes in unsaturated fractures have not yet been resolved. The current model formulations rely on continuum approaches which determine the permeability for each of the two phases, air and water, in terms of relative permeability, where the relative permeability approaches the single phase value as the saturation of that phase approaches 100%. Thin-film flow is an alternative process (Tokunaga and Wan, 1997). Thin films may have significantly greater flow capacity than equivalent porous media particularly for vertical flow. Tokunaga and

Wan found that vertical fractures could have as much as 1000 times the water flow capacity as an equivalent saturated continuum operating with a gradient of one.

These limitations of site-scale numerical models are well recognized among the Yucca Mountain Project investigators. Hence, considerable effort has gone into the development of multiple lines of evidence to constrain the interpretive range of model results. These lines include temperature profiles, gas flow responses to ESF construction, the interpretation of gas pressure interference tests, and studies of environmental tracers, which are the subject of this peer review report. As discussed in Fabryka-Martin, et al. (Chapter 8, 1997), environmental tracers reflect scales of time and distance that other forms of experimentation (such as pneumatic tests or human-induced tracer tests) cannot duplicate. Environmental tracers reflect the "...bulk response of the system, particularly the combined influence on fracture-matrix interactions on transport processes in the liquid phase".

Given the possibility of under-constraint of the numerical models, the it is clear that data from environmental tracers are essential for constraining model performance. Without such constraints, the utility of the models is highly questionable. The remaining questions for this discussion are which tracers should be used and for what purposes in constraining model development.

Environmental tracers have been used to estimate water flux. Total chloride and strontium isotope simulations provide independent means of estimating flux, the chloride based on mass balance and the strontium reflecting retardation effects in the PTn. Both approaches give reasonable and consistent values of <5mm/yr (Sonnenthal and Bodvarsson, 1997) and 0.5-5 mm/year (Sonnenthal, et al., 1997) respectively.

It should be noted that both the total chloride and strontium simulations use the older single permeability models, which do not reflect the dual permeability modifications that were made in response to the fast-path hypotheses. While such calculations may approximate average flux, neglect of the fracture pathways loses detail in the extent that the flux may be concentrated locally.

A consequence of adopting a dual permeability model is that the site may also have dual saturation, dual flux, and dual chemistry behaviors as well. It is vital that the project differentiate the fracture and matrix fluxes (particularly for the non-welded units). Furthermore, sampling efforts in the future should attempt to distinguish chemical components in the matrix and in the fractures.

The value of ^{36}Cl as compared with other geochemical indicators for estimating flux has been questioned within the YMP. Fairley and Wu (1997) suggest that ^{36}Cl would be less useful as a means of obtaining water flux for several reasons including (1) limited temporal duration of flows, (2) limited flow area as few fractures are expected to fast-path conductors, and (3) masking of ^{36}Cl by dilution with other chloride bearing waters. [2] This assessment of 36Cl/Cl ratios may be premature because it is not known whether $^{36}Cl/Cl$ ratios between 500×10^{-15} and 1250×10^{-15} represent a component of bomb pulse ^{36}Cl. In addition, the ESF is located below a low infiltration zone according to the infiltration model developed by Flint et al. (1996) whereas the E-W drift will be located under an area where infiltration is 3 to 10 times higher). $^{36}Cl/Cl$ ratios measurements from the E-W drift should test concepts of the relationships between infiltration, flux, and the development of preferred pathways.

Despite multiple lines of evidence for total flux, knowing the distribution and localization of flux, both temporally and spatially, may be as important as knowing a single, averaged-flux number. The bomb-pulse studies have something of a chicken-and-egg relationship to other studies on defining localization of flux. A better understanding of higher flux pathways will improve the sampling of bomb-pulse and other fast-path nuclides, while fast-path materials should help in understanding the frequency of higher flux conductors and their characteristics.

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[2] The use of terminology in Fairly and Wu is confusing, in that "fast path" really means paths that are not retarded by the PTn. At the depth of the ESF flow may be equally likely to be fast whether or not the conducting fractures tap breaches of the PTn and carry bomb pulse signatures.

Current and planned experiments to better define percolation in the ESF should greatly aid in a better understanding of flux and pathway localization. Experimental programs to study the fate of construction waters, both from the ESF construction and the planned E-W drift construction, should also receive strong attention.

3.7.4 Use of Flow through the Paintbrush Tuff as an Analog for Flow through the Calico Hills Tuff

A major concern for performance assessment will be the integrity of the non-welded units for retarding flow from the repository to the saturated zone. A major consequence of the discovery of fast paths through the PTn is to raise concerns about the integrity of these deeper transport barriers. At present, the bomb pulse sampling results are a primary source of data on flow through the PTn in particular and possibly non-welded tuffs in general. Hence , flow across the PTn may have value as an analog for flow across the CHn particularly with respect to the frequency and characteristics of fracture pathways through non-welded units. The presence of perched water above parts of the CHn does provide some constraint on the total flux across the unit in those areas. Nonetheless, the use of $^{36}Cl/Cl$ data to understand transport across the PTn may complement information from saturation profiles.

3.7.5 Simulation of Water Travel Times from the Surface to the ESD and Below

As pointed out by Fabryka-Martin et al. (1997) and others in the YMP, fast travel times do not necessarily mean those pathways are carrying the major portion of the flux. $^{36}Cl/Cl$ ratios are mainly indicators of travel time, and by themselves may not indicate total water flux, which has been a major goal of the numerical modeling. However, fast travel times *do not preclude* the possibility of a major portion of the flux moves on the fast pathways, particularly if transient events dominate the water flow through the site. Travel time is one of several calibrations that bring confidence to the modeling results. Furthermore, travel time provide a check on the validity of the flow processes used by the

numerical models, such as two-phase, porous continuum flow versus thin-film fracture flow.

4. Conclusions and Recommendations

This section presents the peer review findings in two parts. The first part consists of the major conclusions of the PRT. The second part includes specific recommendations.

4.1 Conclusions

The staff involved in the Yucca Mountain Project have an outstanding program studying water movement using ^{36}Cl. This program has contributed significant new understanding of the Yucca Mountain flow system (Fabryka Martin et al 1997). Simultaneously the program staff and their collaborators have developed new, more fundamental techniques to reconstruct cosmogenic nuclide deposition during the past 40 years (Plummer et al., 1997). The significance of this publication in SCIENCE demonstrates the high quality and world class nature of the YMP ^{36}Cl program. This is leading edge science, and by its nature, it will therefore be more controversial.

The peer review reached five major conclusions which are stated below and elaborated in subsequent sections of this report. These conclusions are the following:

1. Bomb-pulse sources are currently the only plausible explanation of the elevated ($>1250\times10^{-15}$) $^{36}Cl/Cl$ ratios observed in the ESF.

2. $^{36}Cl/Cl$ has provided the main evidence for fast paths, and continued effort in sampling $^{36}Cl/Cl$ ratios, particularly in the E-W drift and in accessible portions of the CHn such as at Busted Butte, are essential for understanding site hydrogeology.

3. Current sampling programs almost certainly underestimate the numbers of occurrences of bomb-pulse $^{36}Cl/Cl$ in the ESF due to dilution and mixing with stable chloride and challenges in identifying and sampling of likely fast-flow paths.

4. Measurement programs for $^{36}Cl/Cl$ require strong coordination with sampling efforts for other isotopes and environmental tracers.

5. The numerical modeling programs require geochemical data, including $^{36}Cl/Cl$, to constrain the ranges of input parameters, particularly as the geology and flow processes of fast paths may not be understood well.

4.1.1 The Bomb Pulse is the Plausible Source of $^{36}Cl/Cl$ Ratios in the ESF (Anthony Muller)

The opinion of this report is that the elevated $^{36}Cl/Cl$ ratios found in the ESF and other samples originate from the infiltration of ^{36}Cl from weapons tests fallout during the past 50 years. Furthermore the 1250×10^{-15} $^{36}Cl/Cl$ threshold is conservative in identifying bomb waters. It is very important to note, however, that the corollary is not necessarily true -- waters below that threshold cannot (and have not) been claimed to be free of bomb ^{36}Cl and could well be a mixture of non-bomb and bomb waters.

Other explanations of the elevated $^{36}Cl/Cl$ ratios such as in-situ formation of ^{36}Cl in surface exposures containing calcite cannot explain the elevated $^{36}Cl/Cl$ ratios. Further scientific effort need not be expended on this alternative explanation, and the issue should be closed so that YMP staff can focus their attention on the application of the ^{36}Cl technology.

Flow simulations further support the bomb-pulse origins of ^{36}Cl at the ESF. While models are not necessarily perfect representations of reality, the fact that fast paths could be generated without major property adjustments in a dual permeability model boosts the plausibility of bomb-pulse hypotheses.

This report concurs with YMP Principal Investigators that the elevated $^{36}Cl/Cl$ ratios are consistent with unsaturated zone models that contain fast paths and these pathways can only be attributed to water movement through fractures that occurs with minimal matrix interaction in the Paintbrush Tuff.

Members of the YMP have done a very thorough job of reconstructing the temporal variation in $^{36}Cl/Cl$ fallout ratios over the past 50k years. This report supports their conclusion that two periods with significantly different natural fallout existed -- a 500 $\times 10^{-15}$ $^{36}Cl/Cl$ period back to about 10,000 years before present, and a 600 to 1200 $\times 10^{-15}$ $^{36}Cl/Cl$ period from about from 10k years to 50 k years (Fabryka Martin et al., 1997).

4.1.2 $^{36}Cl/Cl$ Studies are Important for Understanding Flow and Transport at Yucca Mountain (Bridget Scanlon)

The $^{36}Cl/Cl$ data are critical to understanding flow and transport at Yucca Mountain. $^{36}Cl/Cl$ studies provide field observations of flow and transport over large space and time scales and provide valuable information on travel times. Studies at the ESF unequivocally demonstrate fast flow to this level that has been attributed to bypassing of the PTn by faulting and increased fracturing associated with faults. Lower $^{36}Cl/Cl$ ratios (500 to 1250 $\times 10^{-15}$) have been interpreted to represent older water (10,000 years). Integration with other isotopic data (^{3}H, ^{99}Tc, and ^{2}H and ^{18}O) will be very important to confirm or refute the hypothesis that $^{36}Cl/Cl$ ratios between 500 and 1250$\times 10^{-15}$ reflect old water or a component of bomb pulse water. Because of the potential for false negatives with the $^{36}Cl/Cl$ analyses as a result of mixing and dilution, it is possible that there are more fast pathways than suggested by the conceptual model.

Although it has been suggested that the volume of water flowing through fast pathways is negligible (~ 1%), this volume has been estimated based on the current conceptual model for ^{36}Cl that has yet to be confirmed. ^{36}Cl ratios between 500 and 1250$\times 10^{-15}$ could represent a component of bomb pulse that would also increase the volume of water flowing in fast pathways. The volume of flow in fast pathways could be much greater beneath areas of higher infiltration. This hypothesis will be tested when the E-W drift is constructed because infiltration rates based on Flint et al. (1996) are 3 to 10 times greater above the E-W drift. Until information on these issues are obtained, (fast flow under higher infiltration, interpretation of $^{36}Cl/Cl$ ratios between 500 and 1250 $\times 10^{-15}$), it is premature to estimate quantities of water involved in fast flow or to evaluate fluxes associated with fast flow.

In addition to providing information of flow processes (i.e. fast pathways) the $^{36}Cl/Cl$ data have played a key role in determining the type of model to be used at Yucca Mountain and in refining fracture-matrix parameter sets for the numerical simulations. Initially equivalent continuum models were used to simulate flow and transport through Yucca Mountain (Wittwer et al., 1995); however, such models were unable to reproduce the distributions of $^{36}Cl/Cl$ found at the level of the ESF. The occurrence of bomb pulse $^{36}Cl/Cl$ at the ESF forced the modelers to use a dual permeability model to simulate fast pathways (Robinson et al., 1995). Therefore, the $^{36}Cl/Cl$ data effected a major change in the modeling process. Several parameter sets have been developed during the course of the Yucca Mountain program which demonstrate that calibrations of these parameters to matrix saturation are not unique (Bandurraga and Bodvarsson, 1997). The $^{36}Cl/Cl$ data provide a critical constraint to these parameters sets for the current flow (Robinson et al., 1997) and transport model and will continue to play a role in refining these parameter sets as new data becomes available. The infiltration map developed by Flint et al. (1996) indicates that the ESF is located under a zone of low infiltration whereas the E-W drift will be located under a zone of ~ 3 to 10 times higher infiltration. $^{36}Cl/Cl$ and Cl data from the E-W drift will help evaluate whether fracture flow is much more prevalent through the PTn under wetter conditions. For example, if bomb pulse $^{36}Cl/Cl$ ratios are found in many areas of the E-W drift, this would confirm the higher infiltration rates and indicate that fracture flow can be sustained through the PTn under wetter conditions. Such results could lead to further major changes in the hydrologic parameter set to be consistent with such data.

While most studies focus on flow through the PTn, studies are also required to evaluate flow through the CHn unit. Tests should be conducted to determine if there are bomb pulse tracers associated with faults in the CHn in the Busted Butte program. These data will help constrain hydrologic parameters for modeling this unit. In summary, the $^{36}Cl/Cl$ data provide a valuable tool to (1) evaluate flow processes through various units, (2) constrain hydrologic parameter sets for numerical simulations and (3) test the surface infiltration model.

4.1.3 Bomb Pulse $^{36}Cl/Cl$ Occurrences are Probably Underestimated Due to Dilution with Stable Chloride and Difficulties in Sampling (Jack Cornett and Thomas Doe)

Sampling for $^{36}Cl/Cl$ ratios poses major challenges. Given several reasons why elevated $^{36}Cl/Cl$ ratios could be masked or missed -- such as dilution, mixing, and a lack of other geologic indicators for fast paths -- one might be impressed that bomb pulse $^{36}Cl/Cl$ was recognized at all in the ESF.

Several factors contribute to the sampling challenges. Because fractures have considerable flux-bearing potential, only small numbers of fractures are necessary to accommodate the fluxes through the unsaturated zone at Yucca Mountain. Pore waters in the fractures of the Topopah Springs tuff may have little interaction with the matrix, hence one should expect bomb-pulse $^{36}Cl/Cl$ and other fast-path indicators to lie in or close to the fracture planes.

Mixing and dilution with stable chloride almost certainly mask bomb-pulse $^{36}Cl/Cl$. Many chlorine-bearing pulses of water are likely to have passed through the fractures at the ESF level. Current $^{36}Cl/Cl$ ratios should represent a mix of sources from both before and after the nuclear testing era. Further mixing and dilution may occur during sampling from the inclusion of significant volumes of rock in addition to the fracture surfaces.

This report concurs with conclusions of YMP Principal Investigators that dilution and mixing could seriously affect the ability to detect bomb-pulse $^{36}Cl/Cl$. Bomb-pulse chlorine is an indicator of a fast paths, however, *the absence of bomb pulse $^{36}Cl/Cl$ does not preclude the presence of a fast path.*

Effective sampling will require a better understanding of the fracture flow and transport processes. The geologic nature of the fast paths is not well enough understood to guide sampling efforts to likely locations with elevated $^{36}Cl/Cl$ ratios. Bomb-pulse bearing paths are not mineralogically distinctive. Although there may be a relationship of pathways to faults, fracture pathways in the welded tuffs may guide fast-moving fluxes well away from fault locations. In summary, sampling approaches require improvement to better define fast-path locations. Sampling needs to be feature-specific as systematic samples are unlikely to yield useful results.

4.1.4 Measurement Programs for $^{36}Cl/Cl$ Require Strong Coordination with Sampling Efforts for Other Isotopes and Environmental Tracers (Bridget Scanlon)

Measurements of $^{36}Cl/Cl$ ratios can be used most effectively by combining these $^{36}Cl/Cl$ ratio measurements with measurements of other isotopes (^{3}H, Cl, $^{87}Sr/^{86}Sr$), that are performed on sub-samples of the same sample, or where this is not possible, samples from the same location.

The YMP site characterization program can be most effective and add value to the overall Yucca Mountain Project by integrating all field, isotopic measurements, and modeling work so that staff continue to rigorously test their conceptual model of water and radionuclide movement through the unsaturated zone. This report concurs with the recommendation of the Thermohydrologic Modeling Peer Review (Witherspoon et al., 1996) that the ^{36}Cl program is a key element in the overall hydrochemistry and model development programs.

4.1.5 $^{36}Cl/Cl$ and Other Environmental Tracers are Required to Constrain Parameter Ranges in Unsaturated Flow Models (Thomas Doe)

Unsaturated zone modeling and site simulation are very challenging, and the Principal Investigators and the modeling groups should be commended for outstanding effort in this area.

Without hydrogeochemical data, however, the site models are highly under-constrained. The matrix saturation, which is the main calibration criterion, can be matched by a wide range of models and data sets, including the now-rejected single permeability models. Although the dual permeability formulations represent a major conceptual improvement, major uncertainties remain in properties such as the fracture-matrix interaction terms and in the in situ fracture saturations. Furthermore, fundamental areas such as the process of unsaturated flow in the fractures remain uncertain. The introduction of thin-film flow concepts (Tokunaga and Wan, 1997) may indicate flux capacities in excess of those theorized by more conventional two-phase porous media approaches.

Given these uncertainties, the isotopic data, including ^{36}Cl become key components in constraining model interpretations. ^{36}Cl may be less effective than other chemical constituents, such as total Cl, for estimating total flux at the repository level. However, understanding the spatial and temporal variations of flux is equally important as having an averaged flux value, and bomb-pulse isotopes have been critical for recognizing flux localization. At this time, the YMP is still developing an understanding of the proportion of the total flux being borne by various pathways in the unsaturated zone. These pathways would include bomb-pulse fracture networks and non-bomb pulse fracture networks in welded tuffs, and in the non-welded tuffs the relative proportions of matrix and fast-path fracture flow.

Statements that fast-path flux is a minor component of total flux may be true, but also may require more thought and substantiation. For example, if the $^{36}Cl/Cl$ ratios between 500 and 1250 x 10^{-15} represent a component of bomb pulse water there may be much more water flowing along fast paths than is currently thought. The ESF underlies an area of low infiltration according to the infiltration map developed by Flint et al. (1996); however, infiltration rates over the area of the E-W drift are 3 to 10 times higher; therefore, there could be substantially more water flowing along fast paths in the E-W drift.

In addition to the flux calculations, using hydro-geochemistry to study the pathways through the PTn to the ESF is a useful analog for the flow from the repository through the Calico Hills tuff to the saturated zone.

4.2 Recommendations

4.2.1 Need for $^{36}Cl/Cl$ Ratio Studies and Improvement of Sampling Strategies (Jack Cornett)

4.2.1.1 The analysis of ^{36}Cl should be continued for reasons discussed in section 4.1.2. Future field work should focus upon feature based sampling and should include provision for the analysis of the multiple suit of isotopes mentioned in this set of recommendations.

4.2.1.2 The method to select feature-based samples for ^{36}Cl analysis should be reviewed and clearly documented. Greater effort should be made to geologically distinguish potential fast paths (though mineralogy has not been successful in this regard). Also methods should collect sufficient sample material for analyses of other isotopes.

4.2.1.3 The total concentration of stable Cl should be measured in each rock type so that corrections can be made for the amount of dilution introduced during the leaching of the samples.

4.2.1.4 A method should be developed to estimate the amount of Cl (and ^{36}Cl) extracted from the rock and fluid inclusions during the pore water extraction. Possible approaches that could be explored are differences in the pore water and rock in $^{35,37}Cl$, ^{87}Sr, and anion ratios in the extract. Such a correction would increase the power of the ^{36}Cl method so that the influence of dilution is minimized.

4.2.1.5 The additional analyses referenced above should be used to examine alternative conceptual models of the ^{36}Cl distribution. These models should include geologic features, dilution, proximity to fractures, the pneumatic characteristics of the rock mass.

4.2.2 Integration with Other Isotopic and Environmental Tracer Methods (Bridget Scanlon)

4.2.2.1 The panel recommends that further work in isotope hydrology should use multiple isotopes (^{3}H, ^{99}Tc, $^{13,14}C$, ^{36}Cl, and stable isotopes, ^{18}O and D). Samples should be enlarged so that splits of the same sample could be used for multiple isotopes.

4.2.2.2 ^{99}Tc should be measured in a suite of about 10 ESF samples that have already been analyzed for ^{36}Cl (with appropriate blanks and standards). Sr isotope ratios, which reflect rock water interaction, should be measured to assist in distinguishing waters passing through fast paths from those traveling matrix pathways.

4.2.2.3 The presence or absence of tritium concentrations greater than ~ 1 TU in pore waters provide key corroboration of recent or post bomb water recharge. However the reliability of past low level tritium measurements (<~16 TU) is not clear because field blanks have not been performed routinely and lower limits of detection are often not clearly stated. Better detection limits are needed for this work. The panel recommends that the program investigate the use of isotope enrichment and gas flow proportional counting to achieve an acceptably low background detection threshold. The panel recommends also that the program develop and use a method to determine the tritium concentration in field blanks.

4.2.3 Integration of Environmental Tracer Studies with Testing and Modeling (Thomas Doe, unless otherwise noted)

4.2.3.1 The surface infiltration studies and related model development provide a key constraint on the flow through the unsaturated zone. The amount of infiltration drives flow and transport in the PA models. Work on the infiltration model should be continued with an emphasis on refining the upper limits on infiltration under wetter climates.

4.2.3.2 The Yucca Mountain Project should develop and test, through experimentation, alternative conceptual models of the flow processes, such as thin-film process models.

The results of percolation tests on natural fractures and observations of re-wetting of water bearing fractures under controlled humidity conditions underground should contribute greatly to understanding conductive fracture frequencies and flux localization. These data should be critical to focused sampling for hydrochemistry.

4.2.3.3 The premise of fracture flow through the PTn is a key assumption that must be invoked to decrease travel times so that bomb pulse nuclides can rapidly penetrate into underlying units. The YMP should investigate ways to locate and quantify flow through this breach.

Transport rates and water fluxes through major faults are key parameters in the YMP PA work. This report recommends that (R. Jack Cornett)

> *4.2.3.4* Determining the hydraulic properties of fractures (as opposed to matrix flow) should receive proportionately more effort in future work.
>
> *4.2.3.5* All appropriate aspects of the program should test their understanding of the flow system by documenting their predictions of the hydrologic properties of the Solitario Canyon fault before it is crossed in the E-W drift. The value of predictions of bomb-pulse occurrence will be increased greatly if uncertainty in sampling methods and sampling site selection (i.e. targeted fractures) can be decreased.

Transport rates and water fluxes through major faults in units below the repository are key parameters in the YMP performance assessment work. The panel believes that the test program at Busted Butte provides an excellent way to learn about fast-path potential in the CHn and to test predictions and isotopic techniques developed in the ESF studies. This report further recommends (Bridget Scanlon, Jack Cornett, and Thomas Doe) that:

> *4.2.3.6* The Busted Butte program plan should be reviewed to integrate more isotopic and environmental tracer work.

4.2.3.7 The hydraulic properties of fractures through Busted Butte should be investigated.

4.2.3.8 All appropriate aspects of the program should test their understanding of the flow system by documenting their predictions of the hydrologic properties of the Busted Butte system before it is instrumented.

4.2.3.9 The Busted Butte flow system should be investigated using multiple isotopes (^{3}H, ^{99}Tc, $^{13,14}C$, ^{36}Cl) and stable isotopes (^{18}O-and D). in order to test the predictive power of the techniques.

4.2.3.10 A multiple isotope approach should be used to test for fast pathways and to estimate water fluxes in the unsaturated zone in geologic units below the repository horizon.

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Appendix 1. Peer Review Plan

PLAN FOR PEER REVIEW OF CHLORINE-36 STUDIES AT YUCCA MOUNTAIN

This Peer Review Plan has been prepared in accordance with existing project requirements for quality assurance and with the YMP Office Quality Management Procedure, QAP 2.5, Peer Review. The outline of the Plan follows section 6.2 of QAP 2.5.

A. Statement of Work Product

The purpose of this peer review is to evaluate the sampling, analytical, and interpretational aspects of the Chlorine-36 (Cl-36) and other environmental isotope data collected by the Yucca Mountain Project. The peer review panel will also examine the adequacy of the approach which the project is using to incorporate this data with the site-hydrogeologic characteristics to develop the predictive models of groundwater flux and its distribution in the potential repository horizon. The use of these data in performance assessment is specifically not included within the scope of this review.

The panel will perform the following tasks:

- Review the most recent synthesis reports on Cl-36 and other environmental isotopes in the context of the 3D geologic framework model and updates of the UZ flow and transport models.
- Evaluate the adequacy of sampling approach and locations utilized.
- Evaluate the adequacy of the fluid-extraction and isotopic analytical approach, precision and accuracy of the data.
- Evaluate the adequacy of the background definition and accuracy of the half lives of the isotopes.
- Evaluate the adequacy of interpretation of the Cl-36 and other environmental isotope data in the context of the current conceptual flow models and modeling approach that incorporate the site-hydrogeologic features, rock heterogeneity, alteration and fracture mineralogy, and geochronologic data on the fracture-lining minerals.

B. Size and Technical Composition of Peer Review Group

The Peer Review Panel will consist of the four reviewers who are the following:

Dr. Thomas W. Doe, chair, Fracture flow

Dr. Anthony B. Muller (Booz-Allen). Isotope geochemistry

Prof. Fred M. Phillips (New Mexico Institute of Technology), Chlorine isotopes in geologic media[1]

Dr. Bridget Scanlon (University of Texas/Texas Bureau of Economic Geology), Unsaturated zone transport

Dr. Ronald Linden of the Management and Technical Support organization of the Yucca Mountain Project Office will serve as Technical Contact in Las Vegas. Questions, correspondence, materials, or requests for clarifications will be directed to the Chair and the Technical Contact, who will prepare the materials and distribute them to all the Panel members.

C. Method of Documenting, Controlling and Resolving Comments, Concerns, and Conclusions

The peer reviewers will maintain written minutes of meetings, deliberations, and peer review activities. The Peer Review Report will be prepared in sections by the various reviewers, and each reviewer will review all drafts of the Peer Review Report. All comments, resolutions to comments, and acceptances to resolutions will be documented using a formal Document Review Record. If the comments of the reviewers cannot be resolved, the Peer Review Report will note the absence of concurrence and provide an explanation of each opinion.

A draft Peer Review Report will be submitted to the Yucca Mountain Project Office. The Project Office will comment on Peer Review conclusions, primarily for the purpose of noting opinions or conclusions that may have been based on misinterpretation of information or incomplete information. All comments, resolutions to comments, and acceptance of resolutions relating to the Project Office review will be documented using a formal Document Review Record. If resolution of Project Office comments cannot be attained, the Peer Review Report will note the absence of concurrence with the Project office and provide an explanation of Panel members' opinions.

---

[1] Dr. Fred Phillips withdrew due to university schedule conflicts and was replaced by Dr. Jack Cornett of Atomic Energy of Canada, Ltd.

D. Schedule for Conducting and Reporting Results of the Peer Review

The Peer Review will be completed in a series of tasks. The tasks and key schedule dates are given in Table 1.

Task 1. *Peer Review Plan*: The Peer review chair has prepared this Peer Review Plan prior to distribution of review materials

Task 2. *Peer Review Preparation* will include selection and distribution of materials for the peer review by September 15, 1997. Additional FY 1997 deliverables may be added as they become available. Materials will include:

> Summary Report of Chlorine-36 Studies: Sampling, Analysis, and Simulation in the Exploratory Studies Facility, by J. Fabryka-Martin, A. Wolfsberg, P. Dixon, S. Levy, J. Musgrave, and H. Turin, Los Alamos National Laboratory Report, LA-CST-TIP-96-002, Milestone 3783M (29 August, 1996)

> Investigations on Structural Controls and Mineralogic Associations of Chlorine-36 Fast Pathways in the ESF (DRAFT) by S. Levy, D. Sweetkind, J. Fabryka-Martin, P. Dixon, J. Roach, L. Wolfsberg, D. Elmore, P. Sharma, Los Alamos National Laboratory Report LA-EES-1-TIP-97-004, Milestone AP2301M4 (12 March, 1997)

> Modeling of Flow, Radionuclide Migration and Environmental Isotope Distribution at Yucca Mountain (DRAFT) by B. Robinson, A. Wolfsberg, H. Viswanathan, C. Gable, G. Zyvoloski, H. Turin, Los Alamos National Laboratory Report, Milestone 3672, (29 Aug, 1996)

> Summary Report of Chlorine-36 Studies (DRAFT) by J. Fabryka-Martin, H. Turin, A. Wolfsberg, D. Brenner, P. Dixon, and J. Musgrave, Los Alamos National Laboratory Report, Milestone 3782, LA-CST-TIP-96-003 (30 Aug., 1996)

Task 3. The *Preparation of for the Peer Review Meeting* will involve reading and review of the distributed materials.

Task 4. *The First Peer Review Meeting* will be held in Las Vegas and at the Yucca Mountain site and will include presentations and discussions with relevant Yucca Mountain investigators, a field trip to the site, and a Peer Review team executive session with preliminary observations to DOE.

Task 5. *Peer Review Report*: The peer review panelists will prepare the peer review report.

Task 6. *Compilation and Reviewer Check*: The peer review chair will compile the individual contributions into a single peer review report. The draft report will be reviewed by the individual panelists. The report will note areas of consensus and document alternate views and opinions.

Task 7. *Comment Response and Revision*: After submission of the draft report, the Peer Review Panel will prepare responses to DOE comments and revise the report appropriately.

Task 8. *Final Presentation*: A final presentation and meeting will be held with DOE by the Peer Review Chair. Additional peer review members will be involved as necessary to resolve final issues.

E. Criteria to be Evaluated

The criteria to be applied in the carrying out the Peer Review will include:

1. validity of basic assumptions
2. alternate interpretations
3. adequacy of requirements
4. appropriateness and limitations of methods and implementing documents used to complete the work product under review
5. adequacy of application
6. accuracy of calculations
7. validity of conclusions
8. uncertainty of results of impact if incorrect.

Acceptance Page

Thomas W. Doe Golder Associates Inc. Peer Review Chair	Steven J. Brocoum Yucca Mountain Project Assistant Manager for Licensing

cc:

F. Harvey Dove, MTS/GFS

Larry R. Hayes, M&O/TRW

Ronald M. Linden, MTS/GFS

Bimal Mukhopadhyay, MTS/BAH

Anthony B. Muller, BAH

Russell L. Patterson, DOE

Fred M. Phillips, NMT

Bridget R. Scanlon, UT/Austin

William B. Scott, USGS

Dennis R. Williams, DOE

Table 1. Chlorine-36 Peer Review Schedule

Distribute to PRT the background products to be reviewed	9/15/97
Distribute the FY97 milestones to PRT for review	10/10/97
First Peer Review meeting (4 days)	11/15/97
Peer Review members provide results to the chairperson	12/15/97
Peer Review report received by DOE	1/12/97
DOE issues Peer Review Record Memorandum with responses	2/27/98
Peer Review chair responds to comments by DOE/revises report	4/13/98
DOE issues Peer Review Record Memorandum	5/8/98

Appendix 2. Peer Review Meeting Agenda

CL-36 PEER REVIEW TECHNICAL PRESENTATIONS AGENDA

Yucca Mountain Site Characterization Project

Fiesta Hotel and Casino   Las Vegas, NV

January 13th and 15th, 1998

Morning of January 13th: Session will start at 8:00 a.m.

1. **Introductory Remarks** (15 minutes)

   Presented by: Russ Patterson (Department of Energy)

2. **Conceptual Model of Flow and Transport at Yucca Mountain** (30 min./20 min.)

   Presented by: Bo Bodvarsson (Lawrence Berkeley National Laboratory)

3. **Infiltration Studies at Yucca Mountain** (30 min./20 min.)

   Presented by: Alan Flint (United States Geological Survey)

   Break - 15 minutes

4. **Chlorine-36 Studies at Yucca Mountain** (60 min./30 min.)

   Presented by: June Fabryka-Martin (Los Alamos National Laboratory)

   Lunch - Approximately 1 hr. 20 min. Presentations will resume at 1:00 p.m. and run to approximately 5:30 p.m.

5. **Thoughts on Alternative Models from a Geologic Perspective** (20 min./15 min.)

Presented by: Mark Tynan (Department of Energy)

6. **Considerations for Peer Review Panel on Cl-36** (15 min./15 min.)

Presented by: John Stuckless (United States Geologic Survey)

7. **Carbon-14 and Tritium Studies at Yucca Mountain** (25 min./20 min.)

Presented by: Gary Patterson (United States Geological Survey)

Break - 15 minutes

8. **Strontium Isotope Studies at Yucca Mountain** (25 min./20 min.)

Presented by: Brian Marshall (United States Geological Survey)

9. **Stable Isotopes Studies at Yucca Mountain (deuterium, O-18, C-13)** (25 min./20 min.)

Presented by: John Stuckless (United States Geological Survey)

10. **Major/Minor Anions and Cations - General Geochemistry** (25 min./20 min.) presented by: Arend Meijer (Los Alamos National Laboratory)

Adjourn for the day

Note: The panel and selected PIs will be participating in a field trip to the site on January 14th.

The technical presentations will resume on Thursday, January 15th.

-<u>Morning of January 15th</u>: Session will start at 8:00 a.m.

11. Solute Transport Simulations of Cl-36 Migration and Distribution (30 min./20 min)

Presented by: Andy Wolfsberg (Los Alamos National Laboratory)

12. Modeling of Chloride Geochemistry in the Unsaturated Zone (30 min./20 min.)

Presented by: Eric Sonnenthal (Lawrence Berkeley National Laboratory))

13. Modeling of Fast Flow and Transport Pathways Using Environmental Isotopic Tracers (30 min./20 min.)

Presented by: Jerry Fairley (Lawrence Berkeley National Laboratory)

14. Use of Isotopic Data in the UZ Transport Model of Yucca Mountain (30 min./20 min.)

Presented by: Bruce Robinson (Los Alamos National Laboratory)

14. Summary of Peer Review Presentations (20 minutes)

Presented by: Ron Linden (Golder Federal Services)

Lunch - Approximately 1 hr. 20 min. Panel/PIs will reconvene at 1:30 p.m.

If deemed necessary, the afternoon will be devoted to a **Round-Table Discussion of Items of Interest to the Cl-36 Peer Review Panel** (No set time or duration)

Appendix 3. Major References Reviewed

Bodvarsson, G.S., Bandurraga, T.M., and Wu, Y.S. (1997). "The site-scale unsaturated zone model of Yucca Mountain, Nevada, for the viability assessment," Lawrence Berkeley National Lab. Rep. 40378 (selected chapters).

Fabryka-Martin, J.T., Flint, A.L., Sweetkind, D.S., Wolfsberg, A.V., Levy, S.S., Roemer, G.J.C., Roach, J.L., Wolfsberg, L.E., and Duff, M.C. (1997). "Evaluation of flow and transport models of Yucca Mountain, based on chlorine-36 studies for FY97," YMP Milestone Report SP2224M3.

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Robinson, B.A., Wolfsberg, A.V., Viswanathan, H.S., Bussod, G.Y., Gable, C.W., and Meijer, A. (1997). "The site-scale unsaturated zone transport model of Yucca Mountain," Los Alamos National Lab. Yucca Mountain Project Milestone SP25BM3.

Appendix 4. Calculation of amount of ^{36}Cl generated in soils at Yucca Mountain by ^{40}Ca spallation in soils

^{36}Cl is created by the spallation of ^{40}Ca by cosmogenic neutrons in surface soils. Production decreases exponentially with depth because these neutrons do not penetrate deeply into the subsurface. In soils with a bulk density of 1.5 gm/cm^3, production falls to one half of the surface rate at a depth of 1m (assumption 2). Thus, taking production to be equal to the surface rate over the first meter and taking no production to occur below it (as has been done here) is equivalent to taking the integrated production decreasing with depth. Thus

$$D = 1\text{m}.$$

where D is the half-depth of neutrons penetration.

The amount of calcium per square cm of surface, C, to which these neutrons have access in the first meter can be calculated by

$$C = (D \times 1\text{cm}^2)\rho f m = 6\text{g Ca}$$

where

> $(D \times 1\ \text{cm}^2) = 100\ \text{cm}^3$ = equivalent volume of soil producing ^{36}Cl at 100% of the surface rate per cm^2 of surface
>
> $\rho = 1.5$ g soil/cm^3 = bulk soil density
>
> $f = 0.1$ g calcite per g of soil = calcite fraction
>
> $m = 0.4$ g Ca per g calcite $= \dfrac{40\ \text{g / mole Ca}}{100\ \text{g / mole CaCO}_3}$

While it can be argued that f is greater than 0.1 at many Yucca Mountain locations, 0.1 is well above the average at the site. Also, f can not exceed 1 and is unlikely to approach it. f =1 would increase C by a factor of 10, which will be used in the sensitivity analysis below.

The amount of ^{40}Ca-origin ^{36}Cl produced per cm^2 of soil surface is then P,

$$P = Cp = 930 \text{ atoms } ^{36}\text{Cl per cm}^2 \text{ of surface per year}$$

where p = 155 atoms of ^{36}Cl per g Ca (assumption 1) per year, and C = 6 g Ca, from above.

These atoms are added to the "background" of ^{36}Cl in infiltrating water. Assuming (assumption 3) that all rain either infiltrates or evaporates from the site, the Cl fallout, F, can be calculated (in atoms of Cl/cm^2 of surface/year) as

$$F = (i \times 1\text{cm}^2)c\frac{1}{fwt_{cl}}\text{A} = 10^{17} \text{ atoms Cl/cm}^2 \text{ soil surface per year}$$

where i = depth of infiltrating water per cm^2 of surface (so $i \times 1$ cm^2 = volume of water infiltrating per cm^2 of surface)

c = 0.6 mg Cl per L (assumption 4)

fwt_{Cl} = 35.5 g Cl / mole Cl

A = 6.02 x 10^{23} atoms / mole (Avogadro's number)

The potential ^{36}Cl contribution from Ca is thus

$$\frac{P}{F} = 9.3 \times 10^{-15}\ {}^{36}\text{Cl}/\text{Cl} = 9.3 \times 10^{-15}$$

Note that this relationship is true independently of how much ^{36}Cl there is in the infiltrating water, since this is the <u>additional</u> ^{36}Cl from the Ca source.

Assuming that the infiltrating water has 500×10^{-15} (assumption 4) then 5 x 10^4 atoms of ^{36}Cl (= 500 x 10^{-15} ^{36}Cl/ΣCl × 10^{17}ΣCl) are in the fallout. An additional 930 atoms represents <2% of the fallout quantity. Both of these perspectives show a calcium source of ^{36}Cl to be insignificant to hide or appear like bumb pulse Cl at the 1200 $\times 10^{-15}$ threshold level.

Even if $f = 1$ were used, this conclusion still holds, since a ratio of 93 $\times 10^{-15}$ is not significant either.

It could be argued that ^{40}Ca-origin ^{36}Cl could accumulate over several years in soil and be released in pulses to infiltrating water. This is consistent with the periodicity of rainfall in desert regions and with pedologic evidence that soil calcite is occasionally reworked by water. The nature of calcite dissolution/reprecipitation in soil suggests that only a small mass fraction of caliche is mobilized in any event, even if the event is large. If one assumes that every 1000 years, 10% of calcite releases its accumulated ^{40}Ca-origin ^{36}Cl, the calcite contribution would be an average input of 930 $\times 10^{-15}$ (rather than 9.3 $\times 10^{-15}$). However the stable Cl will also have accumulated in the soil between these hydrogeochemical events. All of the Cl and ^{36}Cl would be mixed and undergo appreciable dilution as infiltration is integrated over time and location (by a factor of at least 100). The periodicity and fraction assumptions underlying this assumption are very conservative since they can not be sustained over time. That is, steady state would be achieved at about 10,000 years with mean ^{40}Ca-origin ^{36}Cl in soil of about 50,000 $\times 10^{-15}$, which is far above what has been observed in soils at Yucca Mountain. Shorter periods, which are more reasonable, would not allow enough accumulation for the pulse to have impact while larger discharge rates would drain the soil reservoir with similar effect.

<u>Assumptions</u>

1. At elevation and latitude of Yucca Mt., ^{40}Ca spallation production rate is about 155 atoms of ^{36}Cl yr^{-1} g^{-1} ^{40}Ca (FM, et. al., 1997)

2. Cosmogenic neutron attenuation has a half-length of about 155 g/cm^2 of soil which corresponds to about a production flux 1/2 of the surface flux at 1m in a soil with bulk density of 1.5 g/cm^3 (FM, et. al., 1997)

3. This is based on a mean infiltration rate of 10 cm and the assumption that all Cl in rainfall will infiltrate. Greater infiltration rates are less conservative (*i.e.* decrease the importance of ^{40}Ca as a source) as are higher Cl concentrations in precipitation.

4. Assumes background (non-bomb) precipitation to contain 500×10^{-15}.