



U.S. Department of Energy  
Office of Civilian Radioactive Waste Management



# Los Alamos National Laboratory Chlorine-36 Validation Studies at Yucca Mountain, Nevada

Presented to:  
**Nuclear Waste Technical Review Board**

Presented by:  
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**Amargosa Valley, Nevada**



# Changes in Los Alamos National Laboratory $^{36}\text{Cl}$ Program

- Robert Roback joined project in 2000, assumed Principal Investigator (PI) role in FY 2001
- Complete changeover in technical support (sample processing) occurred after 3 month overlap
- New laboratory for sample processing established; located in non-rad area of Los Alamos National Laboratory (LANL)
- Modifications made to original sample processing procedure
- Many samples submitted to Lawrence Livermore National Laboratory (LLNL) in addition to Purdue Rare Isotope Measurement Laboratory for Accelerator Mass Spectrometry analysis

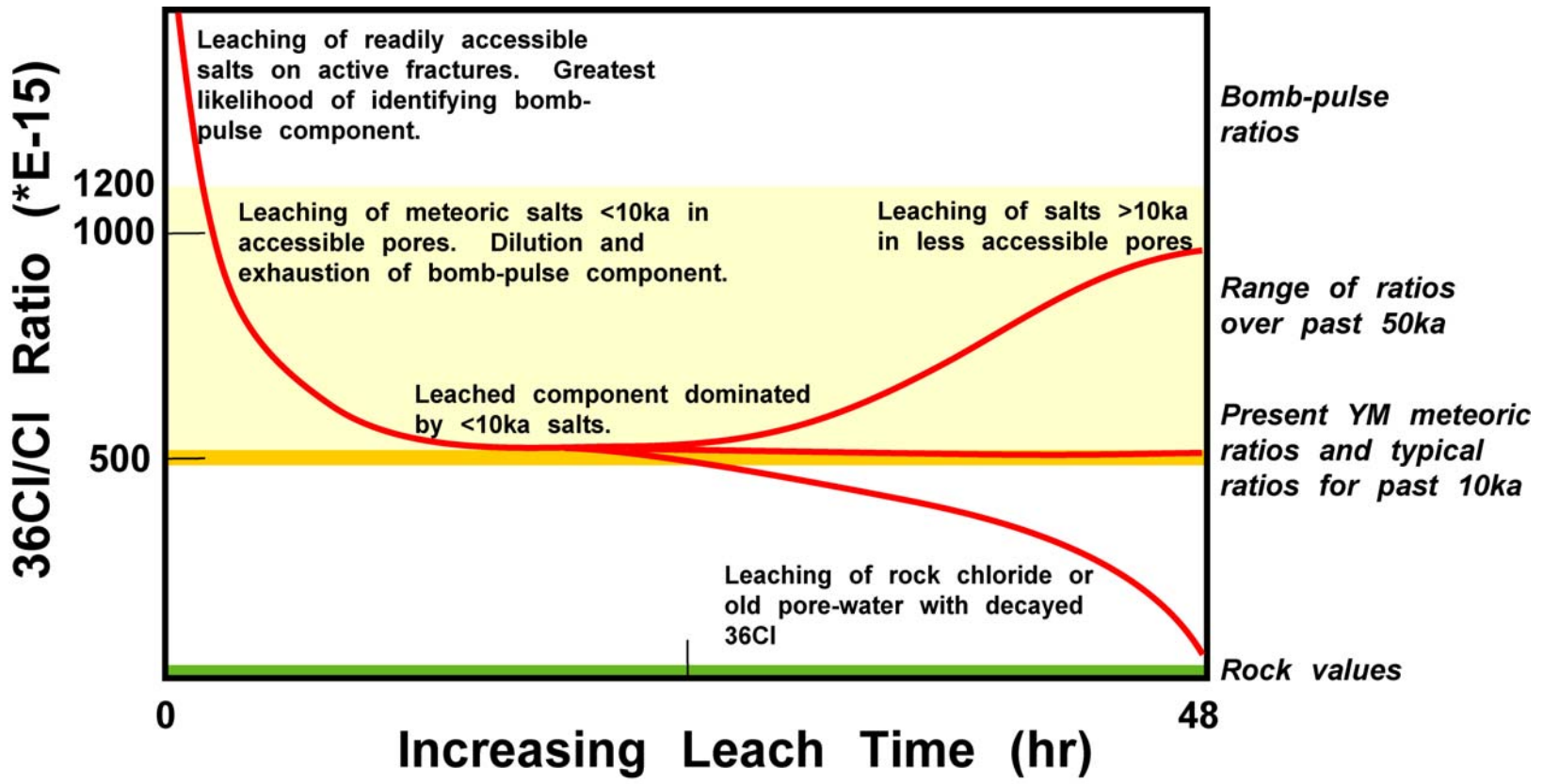
***Current LANL  $^{36}\text{Cl}$  Project satisfies most criteria for independent validation***



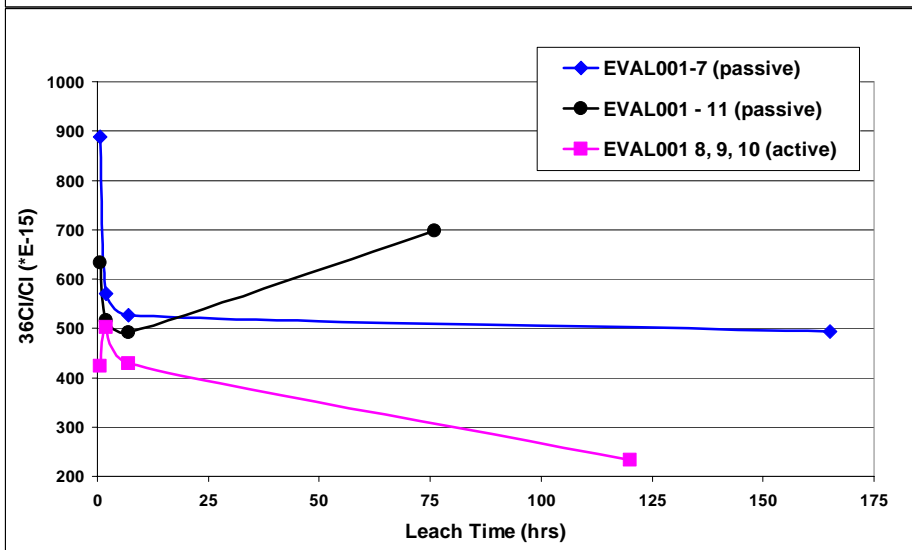
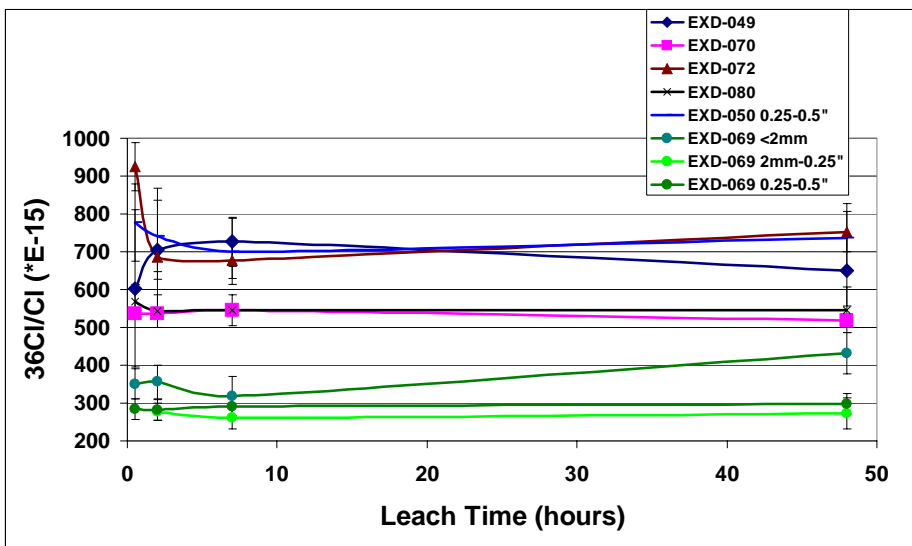
# Summary of Data Produced

- **Set of samples from Enhanced Characterization of the Repository Block (ECRB) processed using “traditional” methods**
- **ECRB samples to evaluate effects of sample processing on  $^{36}\text{Cl}/\text{Cl}$  ratios. Variables evaluated include:**
  - **Leaching method**
    - ◆ **Active leach (i.e., sample is agitated during leach)**
    - ◆ **Passive leach (i.e., sample is left undisturbed during leach)**
  - **Leaching time**
    - ◆ **0.5 hrs to 165 hrs**
  - **Particle size**
    - ◆ **Dust to 1/2”**
- **Validation samples leached at U.S. Geological Survey (USGS)**
- **Niche 1**
- **Blanks and Standards**

# Conceptual Model of Effects of Progressive Leaching on $^{36}\text{Cl}/\text{Cl}$ Ratios



# Results of Leaching Experiments – Enhanced Characterization of the Repository Block Samples



- Samples span length of Cross Drift; include all lithologic units in Cross Drift: TpTpll, TpTpul, TpTpln
- All passive leach; one sample with three size fractions
- Reference sample (EVAL #1): passive and active leach

# Summary of All Leach Experiments

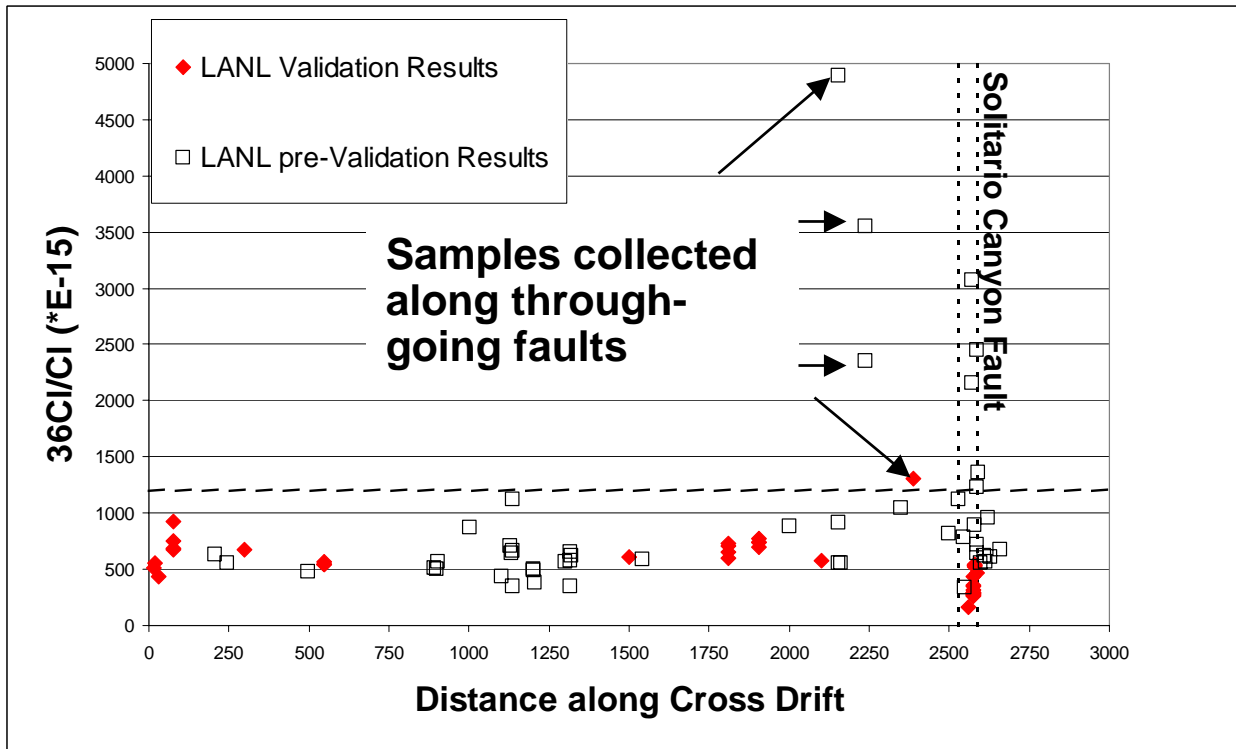
- **7 of 10 passive-leach samples show uniform  $^{36}\text{Cl}/\text{Cl}$  ratios with time**
  - Most are consistent with meteoric salts <10 ka, between 10 ka and 50 ka, or a mixture of the two
- **3 samples show decrease in  $^{36}\text{Cl}/\text{Cl}$  with time**
  - May reflect small component of bomb-pulse signal
- **11 fractions (3 size fractions with sequential leach times) from the same sample have small  $^{36}\text{Cl}/\text{Cl}$  ratios**
  - May reflect uniform addition of rock Cl; or partially decayed  $^{36}\text{Cl}$
- **The active-leach sample shows decrease in  $^{36}\text{Cl}/\text{Cl}$  with time due to dilution by rock Cl**



# Implications for Previous Data

- **Most data do not reflect significant addition of rock Cl**
- **Data likely reflect deposition from meteoric sources <10ka and mixtures of <10ka – 50ka meteoric sources**
- **Cannot rule out possibility that some samples contain small component of bomb-pulse  $^{36}\text{Cl}$  that was not identified**

# Comparison of Los Alamos National Laboratory $^{36}\text{Cl}$ Results For Enhanced Characterization of the Repository Block



**Most samples have values between 500 and 1000 (\*E-15) along entire length of ECRB and in all lithologic units, despite different processing methods**

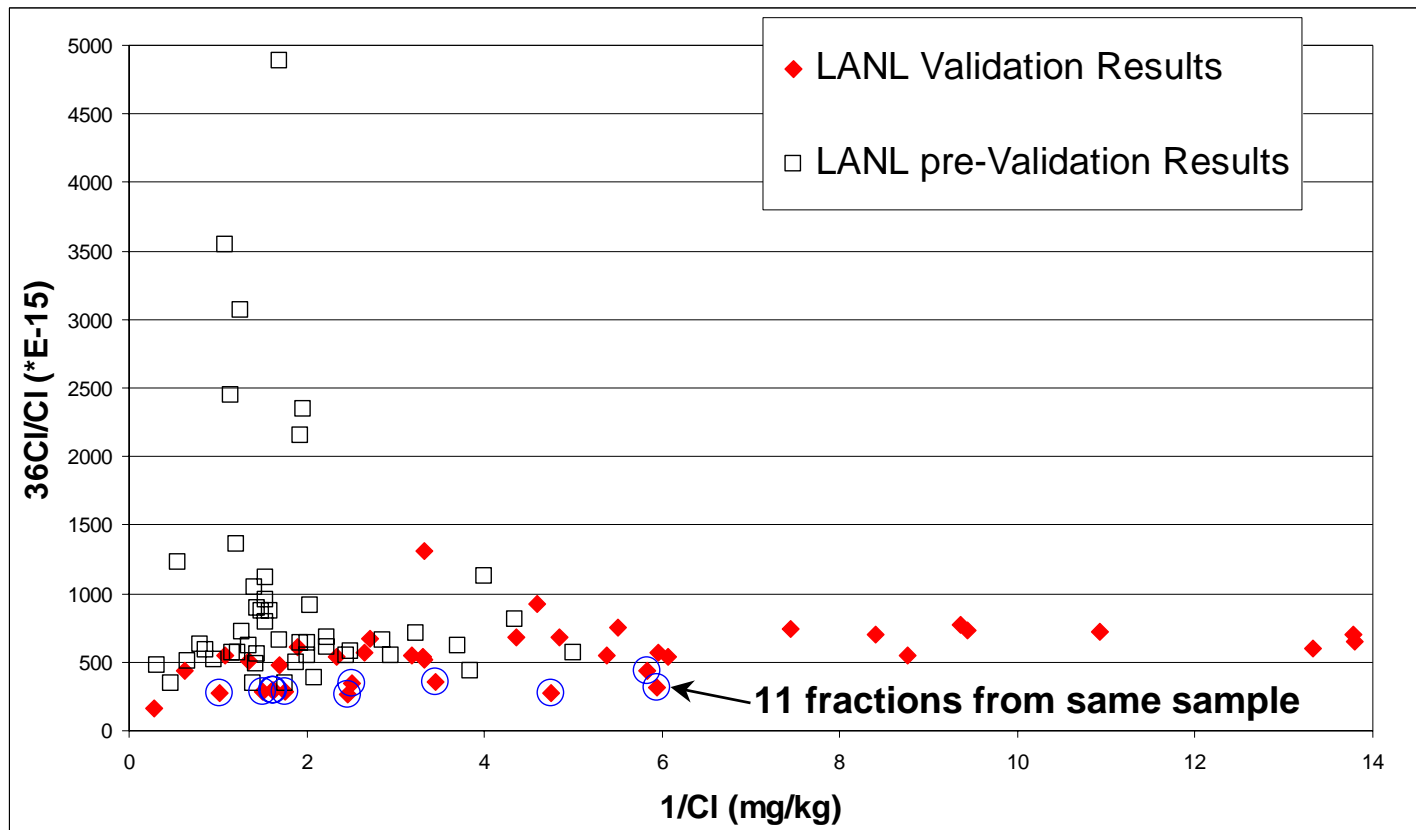
**Samples processed using earlier methods and new methods**

**Results of all leach experiments shown**



# Comparison of Los Alamos National Laboratory $^{36}\text{Cl}$ Results for Enhanced Characterization of the Repository Block

(Continued)



# Discussion of Los Alamos National Laboratory $^{36}\text{Cl}$ Blanks

- Laboratory Blanks
- Laboratory swipes of countertops and hood
  - $2.9 \times 10^{-14}$  mg to  $5.6 \times 10^{-14}$  mg  $^{36}\text{Cl}$
- Process blanks with each group of samples – ~10% of all samples
  - $2.9 \times 10^{-15}$  mg to  $4.5 \times 10^{-14}$  mg  $^{36}\text{Cl}$

## *Blanks are small relative to sample size*

- Typically <15% of small samples, <7% typical sequential leach samples
- Niche 1 samples: 5% or less for all; for 4 samples with bomb pulse blank is between 1.6% and 0.2% of sample
- Earlier LANL  $^{36}\text{Cl}$  samples are larger, measured laboratory blanks that typically account for <1% of sample



# Discussion of Los Alamos National Laboratory $^{36}\text{Cl}$ Blanks

(Continued)

- **Lack of LANL Crushing Blanks**

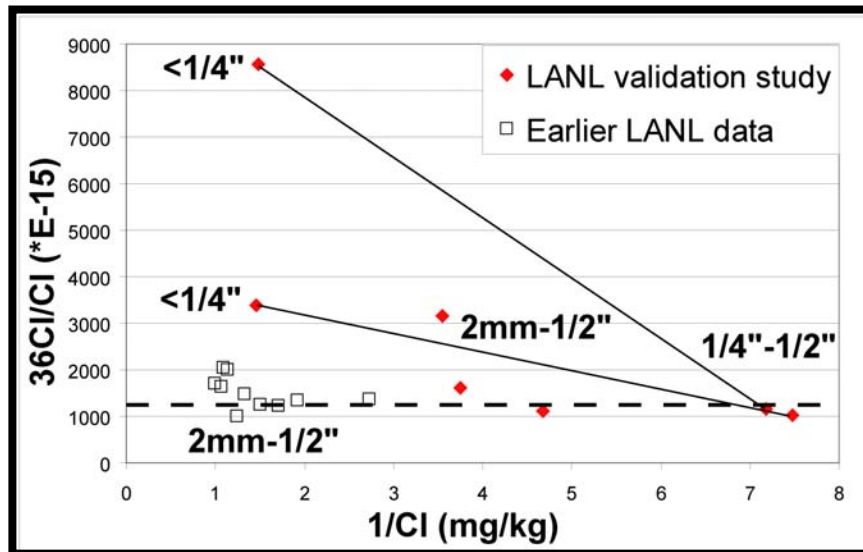
- All crushing and sample processing equipment is thoroughly cleaned prior to use and between samples with final rinse with deionized water
- Sample crushing takes one to a few hours compared to 3-10 days for sample purification

# Discussion of Los Alamos National Laboratory $^{36}\text{Cl}$ Blanks

(Continued)

- **Systematic variations among sample groups**

- Earlier LANL data for feature-based vs. systematic samples
- Generally a good agreement between earlier results and results of this study
- Systematic and reproducible differences among different size fractions and leach times for ECRB and Niche 1 samples



## Niche 1 Results

Tie lines join different size fractions from same sample

# **$^3\text{H}$ and $^{36}\text{Cl}$ Data in Enhanced Characterization of the Repository Block: *Validation of Bomb-Pulse Signal(?)***

- Of 22 samples, 11 yield values  $> 1.0$  Tritium Units (TU) and 8 yield values  $> 2.0$  TU, with a maximum of 10.3 TU
  - Any “valid” analysis  $> 0.2 \pm 0.1$  TU is indicative of recent infiltration
- Most  $^3\text{H}$  and  $^{36}\text{Cl}$  data from samples co-located within a few meters agree ( $^3\text{H}$  below detection and  $^{36}\text{Cl} < 1200 \cdot \text{E}^{-15}$ )

## ***Except***

- One sample pair collocated within 4m that shows second largest  $^3\text{H}$  (9.8 TU) and largest  $^{36}\text{Cl}$  ( $4890 \cdot \text{E}^{-15}$ ) values measured in ECRB
- Of other samples with either  $^3\text{H}$  or  $^{36}\text{Cl}$  bomb-pulse signature, none are collocated within 12 m

***Emphasizes need for coordinated analysis of same sample for  $^3\text{H}$  and  $^{36}\text{Cl}$***

