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

YUCCA MOUNTAIN PROJECT

Chlorine-36 Validation Studies at Yucca Mountain, Nevada

Presented to:
Nuclear Waste Technical Review Board

Presented by:
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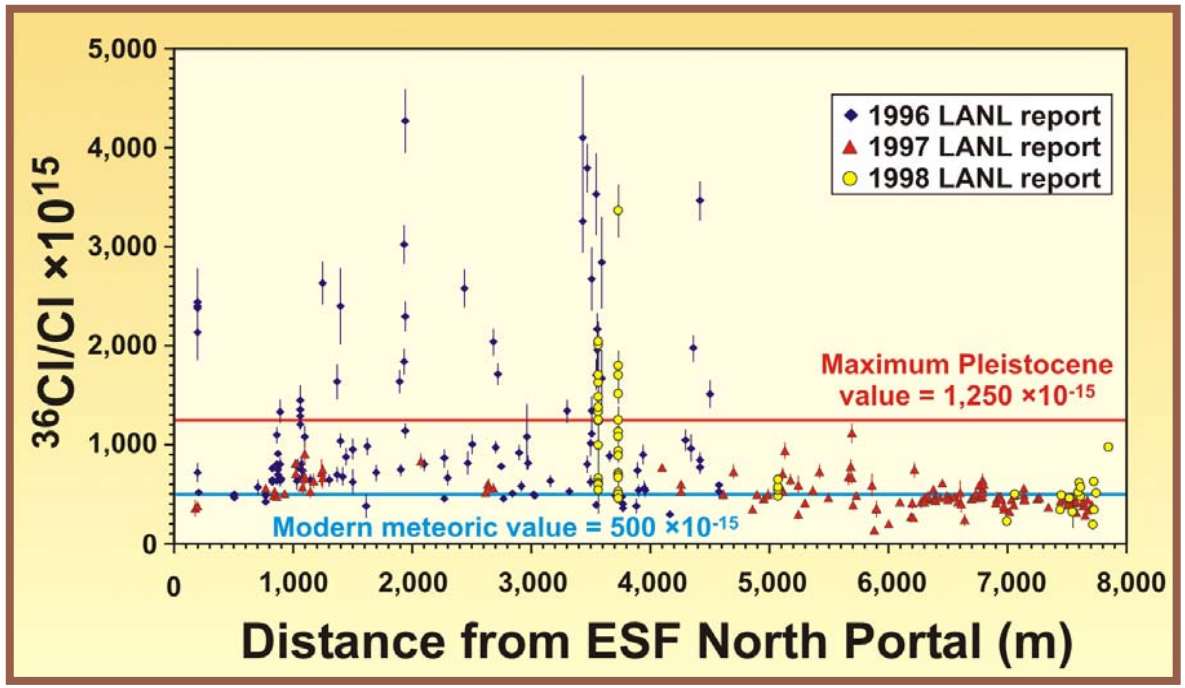
Amargosa Valley, Nevada

Original Los Alamos National Laboratory Data Set

- **Sampling followed Exploratory Studies Facility (ESF) construction and yielded surprising results**
  - **Abundant "bomb-pulse" $^{36}Cl/Cl$ at depth in TSw**
  - **No "bomb-pulse" $^{36}Cl/Cl$ values after ESF station 44+00**
- **Explanation of data required rapid percolation down PTn-cutting faults, as well as variations in infiltration and PTn thickness**
- **Elevated $^{36}Cl/Cl$ ratios have been difficult to reproduce**

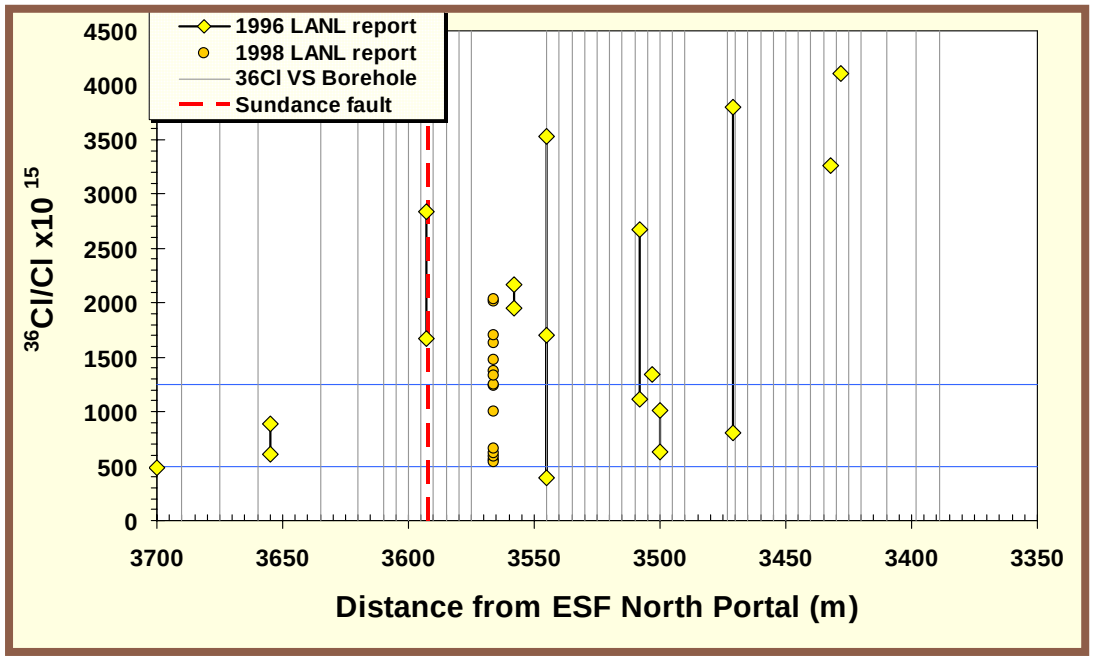


Chlorine-36 Validation Study

- **January 1999 – DOE requests ^{36}Cl Validation Study**
  - **U.S. Geological Survey (USGS) responsible for organizing study and ^{3}H analyses**
  - **Lawrence Livermore National Laboratory (LLNL) responsible for ^{36}Cl analyses**
  - **Atomic Energy of Canada Limited (AECL) responsible for $^{234}U/^{238}U$ analyses**
  - **Los Alamos National Laboratory (LANL) responsible for limited ^{36}Cl analyses in oversight role**
- **Goal of Validation Study was to verify presence of elevated $^{36}Cl/Cl$ over a limited area where it had been reported previously**

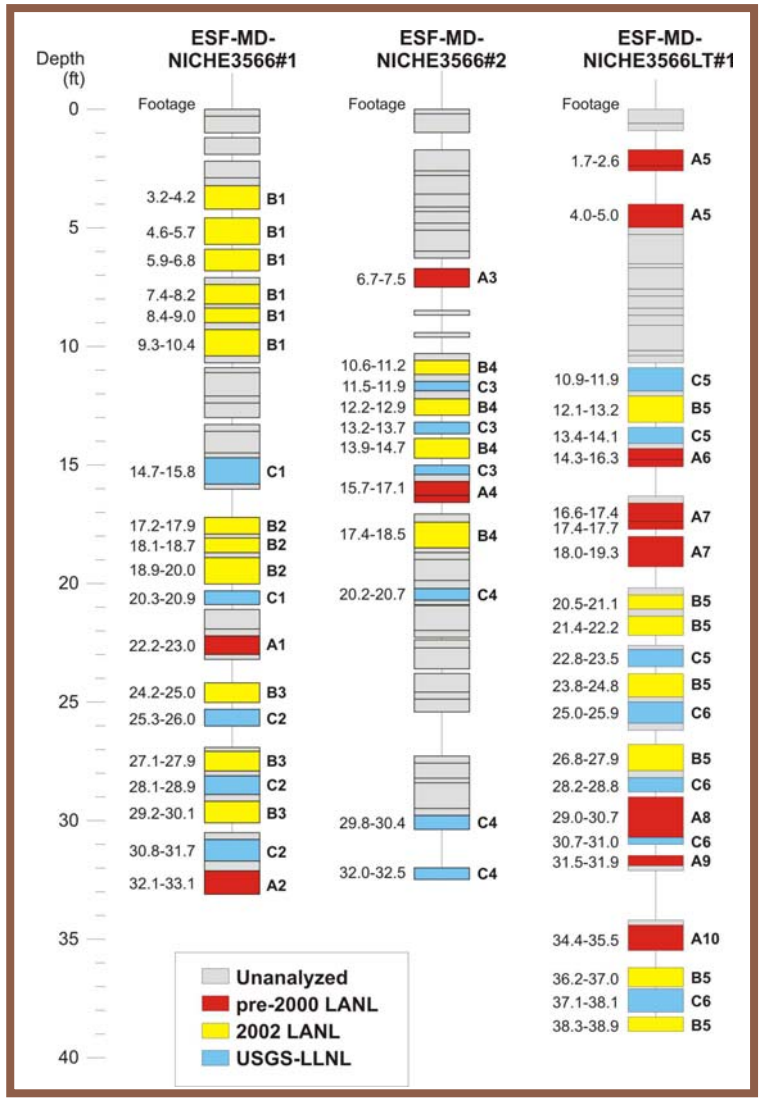
Study Area: Validation-Study Boreholes

- **Sundance fault zone chosen as primary target**
  - **165-m zone from which a large percentage of "bomb-pulse" $^{36}Cl/Cl$ values were previously reported**
  - **Maximizes probability of reproducing "bomb-pulse" signal**
  - **Project drilled 40 4-m coreholes across zone**



Study Area: Niche #1 Core

- **Original LANL data identified "bomb-pulse" $^{36}Cl/Cl$ in 8 of 10 drill core samples from 3 Niche # 1 boreholes**
  - **Supports use of drill core in Validation-Study**
- **Remaining core from the same 3 boreholes was split between USGS-LLNL and LANL labs**
  - **Multiple intervals were combined**
  - **USGS and LANL intervals overlap**
- **Considered critical samples in Validation Study because they represent nearly identical material analyzed at both labs**

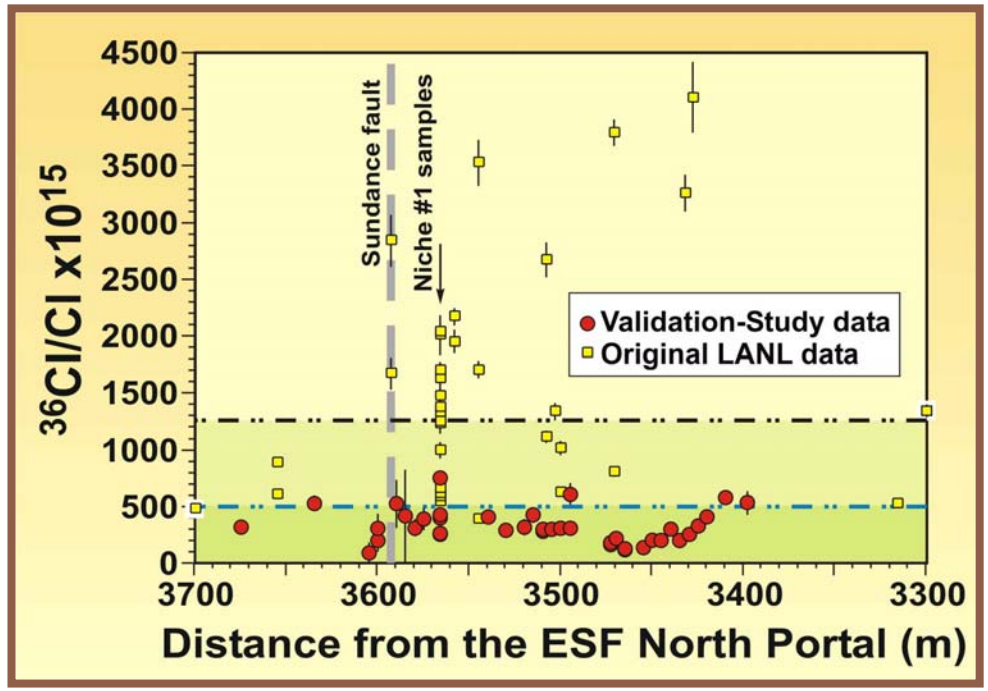


Initial Results

- Samples crushed and leached at LLNL for 7 hours using a slowly rotating tumbler (active-leach method)
- Resulted in leachates with high Cl concentrations and low $^{36}Cl/Cl$ values of 40 to 275 $\times 10^{-15}$
  - Reported to NWTRB, Spring 2000
  - Method considered too aggressive in extracting rock Cl
- Led to experiments on leaching method
  - Passive leaching extracts most labile Cl after several hours
  - Results are relatively insensitive to small differences in particle size and leach times
- Final protocol: passive leaching of 1-2 kg of rock for 1 hr
- Shorter leach times have greater chance of identifying youngest, most labile Cl components

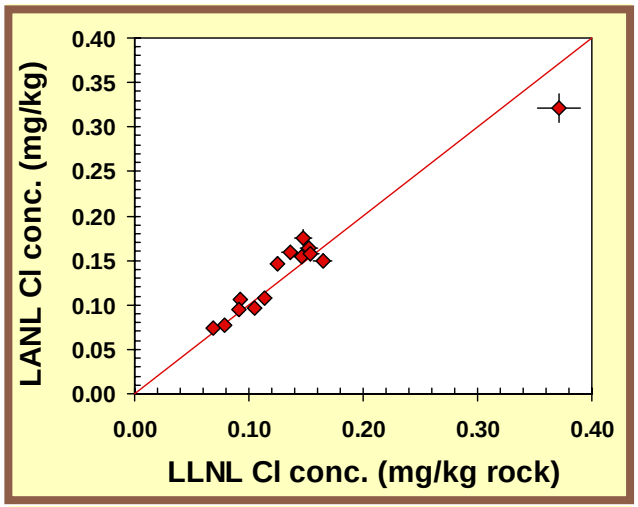
Results of Passive Leaching

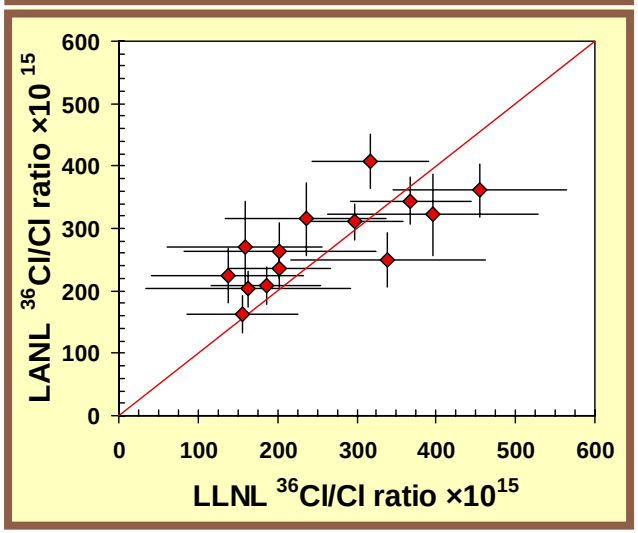
- **Samples crushed at Sample Management Facility (SMF) (DOE, Nevada Test Site (NTS) Area 25), leached at USGS, and analyzed at LLNL:**
  - **34 Sundance fault core**
  - **6 Niche #1 drill core**
- **Cl conc. and $^{36}Cl/Cl$ lower than original LANL results**



Measurement	Validation-Study Results	Original LANL Results
Cl conc. (mg/kg rock) [Mean value]	0.037 – 0.372 [0.141]	0.20 – 2.3 [0.72]
$^{36}Cl/Cl \times 10^{15}$ [Mean value]	137 – 717 [337]	363 – 4,105 [1,437]

Comparison of U.S. Geological Survey-Lawrence Livermore National Laboratory and Los Alamos National Laboratory Splits

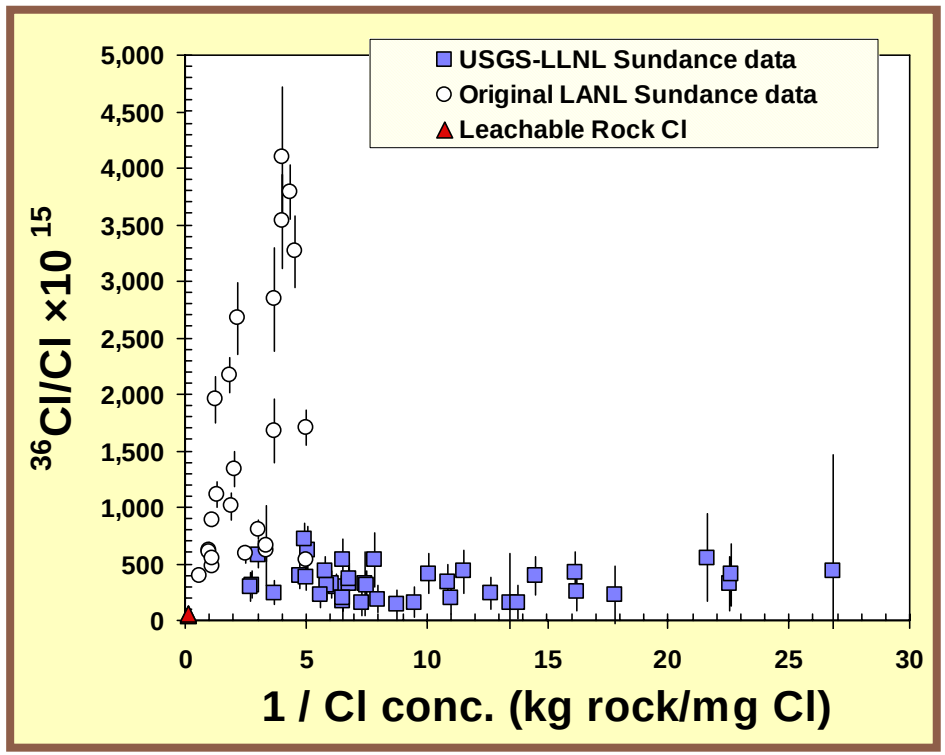




- **A subset of the samples leached at the USGS were split and processed at both LLNL and LANL**
- **Results are comparable for both Cl concentrations and ^{36}Cl/Cl ratios**
- **Indicate that inter-laboratory differences are not caused by:**
  - **Spiking**
  - **Target preparation**
  - **Accelerator mass spectrometry**

Comparison of U.S. Geological Survey-Lawrence Livermore National Laboratory and Original Los Alamos National Laboratory Data

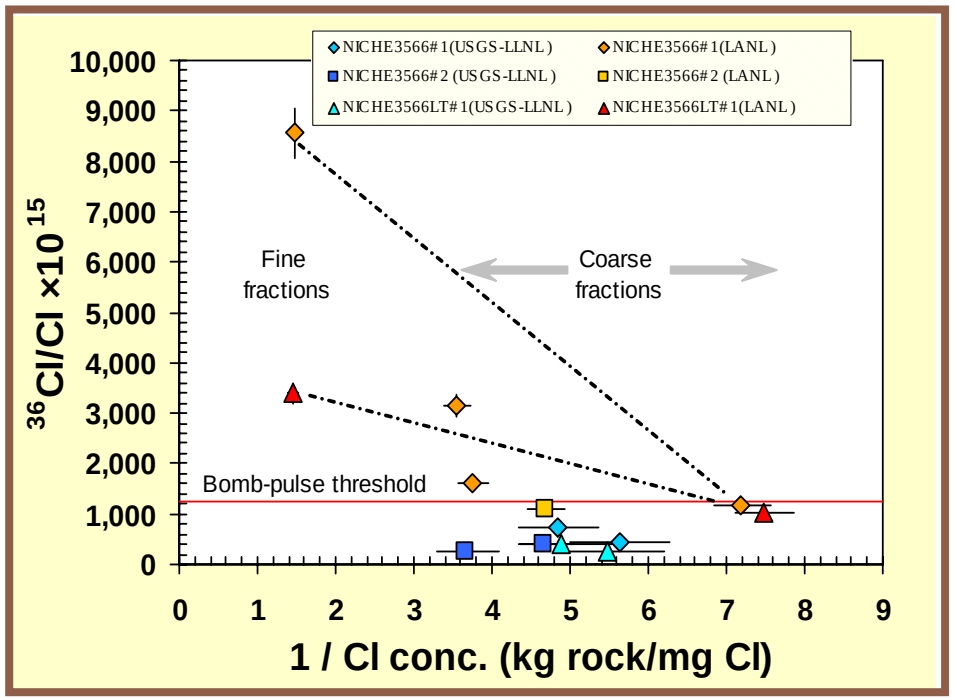
- **USGS-LLNL data form a horizontal trend**
  - **No correlation between Cl conc. and $^{36}Cl/Cl$ ratio**
  - **Low-conc. leachates are highly susceptible to contamination**
  - **Uniform $^{36}Cl/Cl$ values show no evidence for mixing Cl sources***
- **Original LANL data show highest $^{36}Cl/Cl$ in samples with lowest Cl concentration**
  - **"Bomb-pulse" trend is consistent with mixing between sources with low and high ^{36}Cl**



** Mixing of two components results in straight lines*

Comparison of U.S. Geological Survey-Lawrence Livermore National Laboratory and Los Alamos National Laboratory Data for Niche #1

- **LANL results for 7 analyses:**
  - **^{36}Cl/Cl between 1,016 ×10^{-15} and 8,558 ×10^{-15}; 4 of 7 "bomb-pulse"**
  - **Fine fractions (<6.3 mm) had highest Cl conc. and ^{36}Cl/Cl values**
- **USGS-LLNL results for 6 analyses:**
  - **^{36}Cl/Cl from 226 to 717 ×10^{-15}**
  - **Statistically identical to validation-study core**



- **Results from these two data sets indicate that differences in ^{36}Cl/Cl results cannot be explained by differences in sampling approaches**

Other Isotope Tracers

- **Tritium (^{3}H) concentrations measured in pore water extracted from drill core**
  - **Data indicate a cutoff value for post-bomb percolation of ~2 TU**
  - **^{3}H from validation-study core are below post-bomb cutoff**
  - **Elevated ^{3}H in some ESF south ramp and Enhanced Characterization of the Repository Block (ECRB) Cross Drift core**
  - **"Bomb" ^{3}H and $^{36}Cl/Cl$ generally are not spatially coincident**
  - **^{3}H data in ECRB Cross Drift samples requires additional work**
- **$^{234}U/^{238}U$ ratios measured in bulk-rock samples from Sundance fault zone and ECRB Cross Drift**
- **$^{87}Sr/^{86}Sr$ ratios measured in leachates of Niche #1 core**
  - **No statistical differences with pore water from other areas**
  - **Values indicate likelihood that pore water had substantial residence time in PTn**

Summary: Main Findings

- **USGS-LLNL ^{36}Cl data from validation-study boreholes across the Sundance fault zone do not show bomb-pulse signals despite shorter leach times and lower Cl concentrations**
- **^{36}Cl/Cl ratios for samples leached at USGS and processed separately at LLNL and LANL agree within analytical error**
- **USGS-LLNL ^{36}Cl/Cl analyses of 6 Niche #1 core samples are indistinguishable from validation-study core results**
- **LANL ^{36}Cl/Cl analyses of 7 Niche #1 core samples yield "bomb-pulse" values comparable to earlier LANL results**
- **^{3}H data may indicate areas of rapid percolation, but are generally not coincident with LANL ^{36}Cl/Cl results**

Summary: Remaining Issues

- **Validation Study did not yield conclusive result regarding the presence of "bomb-pulse" $^{36}Cl/Cl$**
  - **USGS-LLNL unable to reproduce original LANL $^{36}Cl/Cl$ results under independent laboratory conditions**
  - **New LANL results continue to identify elevated $^{36}Cl/Cl$ ratios**
- **Interpretations remain controversial**
- **What can be excluded as a cause for large $^{36}Cl/Cl$ discrepancies?**
  - **Differences in sampling strategies (Niche #1 data)**
  - **Differences between mechanical versus hand crushing at USGS**
  - **Differences in passive-leaching protocols including small variations in grain size and leach times**
  - **Target preparation and Accelerator Mass Spectrometor (AMS) analysis**

Summary: Remaining Issues
(Continued)

- **What cannot be excluded?**
  - **Possibility of contamination with low $^{36}Cl/Cl$ source in USGS-LLNL environment so that "bomb-pulse" values are masked**
  - **However,**
    - **No correlation between Cl conc. and $^{36}Cl/Cl$ in validation-study core argues against a separate contaminant**
    - **No systematic differences in $^{36}Cl/Cl$ ratios for samples crushed at either Golden, SMF, or USGS labs**
    - **No evidence of anomalously low $^{36}Cl/Cl$ source in either silicon crushing blanks or leaching blanks measured at USGS-LLNL**
    - **Validation-study $^{36}Cl/Cl$ ratios are generally consistent with ESF south ramp samples where no "bomb-pulse" values were reported**

Summary: Remaining Issues

(Continued)

- **What cannot be excluded? (Continued)**
  - **Possibility of contamination with high $^{36}Cl/Cl$ source in NTS or LANL environment resulting in "bomb-pulse" values**
    - **Early studies identified the possibility of ^{36}Cl-contaminated equipment used in field (Fabryka-Martin and Liu, 1995; Fabryka-Martin, Turin, et al., 1996)**
    - **Original LANL data had high $^{36}Cl/Cl$ ratios in low Cl conc. samples (most susceptible to ^{36}Cl addition)**
    - **No crushing blanks were measured at LANL**
    - **^{36}Cl contamination was recognized in LANL laboratory environments (Fabryka-Martin, Wolfsberg, et al., 1996; Fabryka-Martin et al., 1997)**
    - **Small, but systematic, elevation of ^{36}Cl in LANL blanks and regression intercepts**
    - **Small, but statistically significant, differences in $^{36}Cl/Cl$ measured in LANL south ramp and systematic ESF samples versus USGS-LLNL data**

Summary: Recommendations

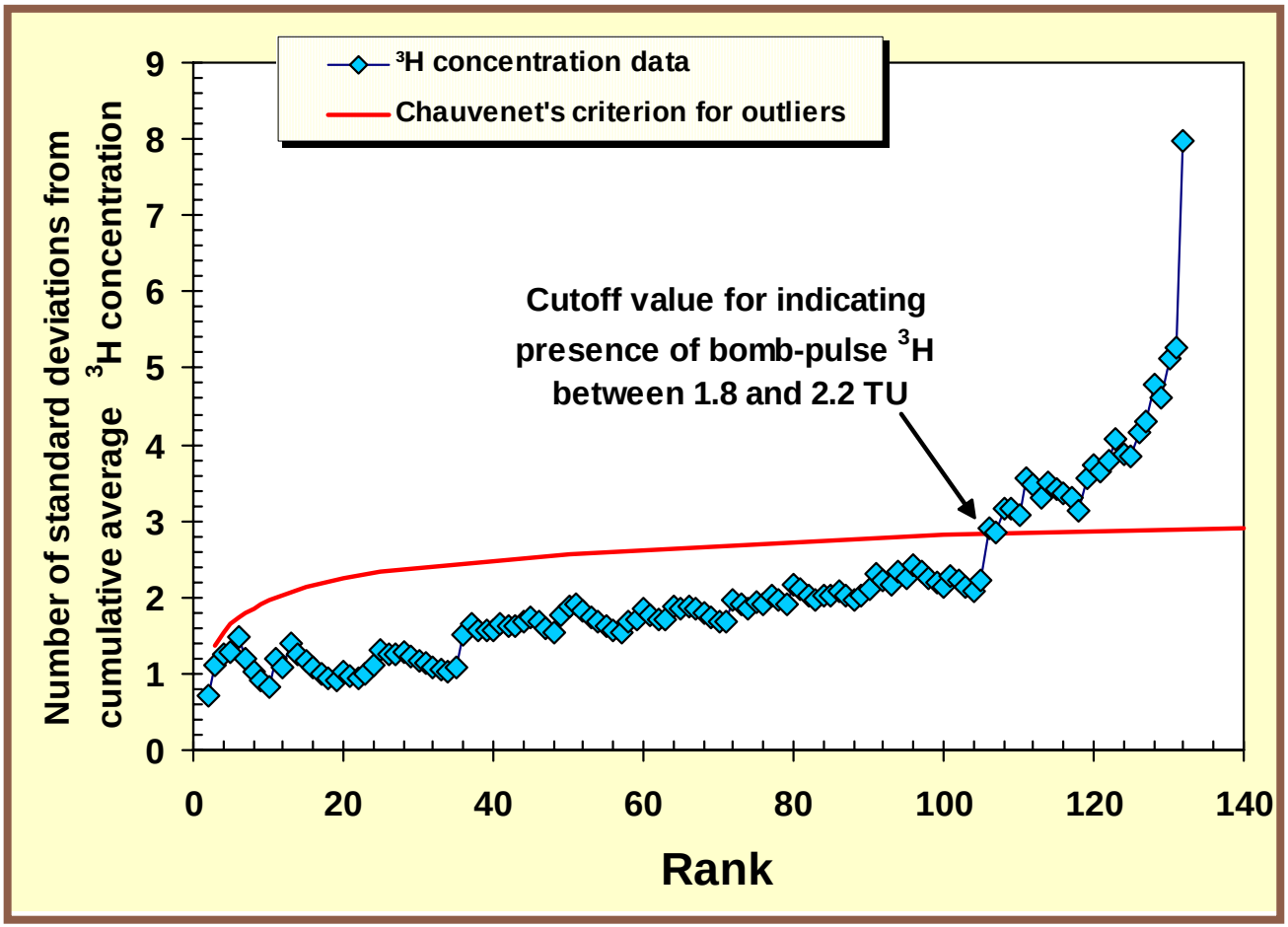
- **Detailed evaluation of sample handling and processing**
- **Rigorous evaluation of crushing and environmental blanks**
- **Additional $^{36}Cl/Cl$ determinations on existing validation-study core**
- **Additional $^{36}Cl/Cl$ determinations on samples previously crushed at LANL**
- **Verification of young water in high-^{3}H samples using $^{36}Cl/Cl$ on leachates of vacuum-distilled core**
- **Independent validation study by third party on new samples**

Supplementary Material

Tritium (^3H) in Validation-Study Samples

- **Pore water was extracted from drill core by vacuum distillation and measured for ^3H at University of Miami**
- **Tritium sources**
  - **Pre-bomb pore water and in-situ production: <1 TU**
  - **Post-bomb atmosphere: 10's to 1000's TU**
  - **Modern atmosphere: ~6 TU**
- **Although analytical method can distinguish background ^3H concentrations of ~0.5 TU, these low levels may not be appropriate for pore water extracted from unsaturated zone rock**
- **Statistical analysis using Chauvenet's criterion indicates a cutoff of ~2 TU before data can confidently be interpreted as indicating post-bomb percolation**

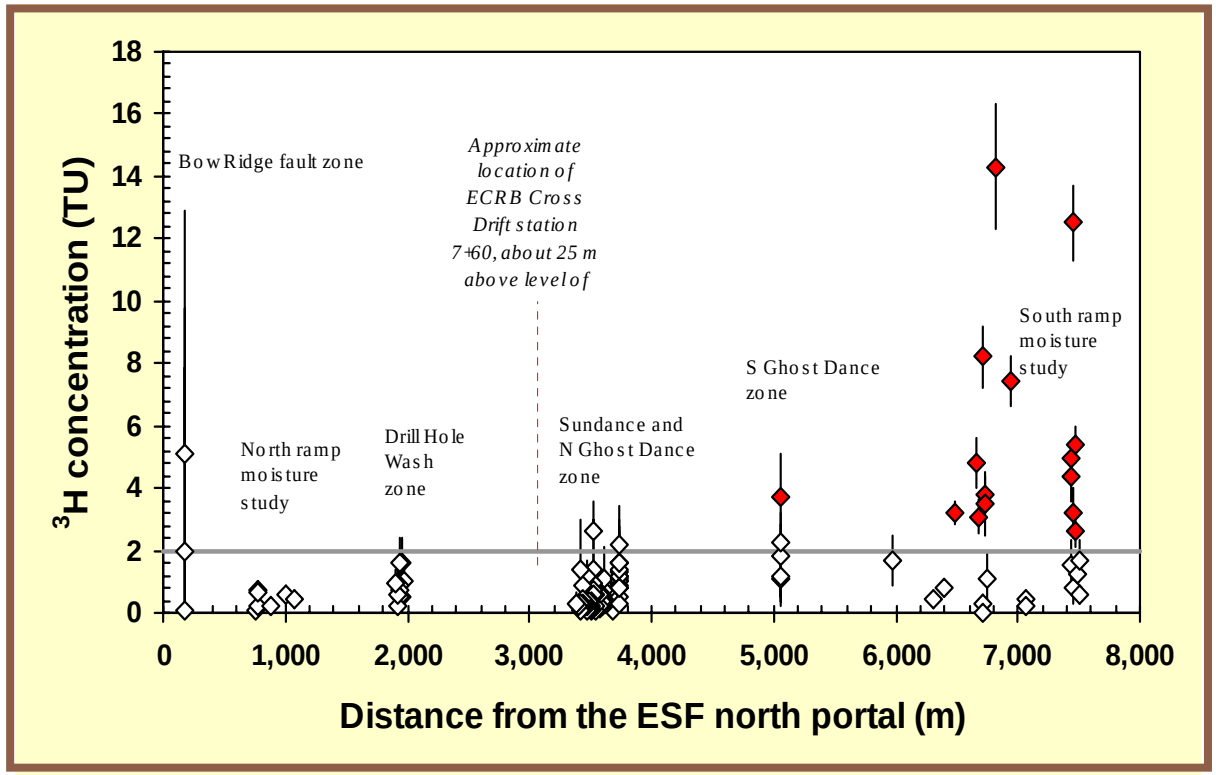
Post-bomb Cutoff Value for Tritium in Pore Water Extracted from Core



Statistical analysis of ^{3}H data using Chauvenet's criterion for identifying outliers

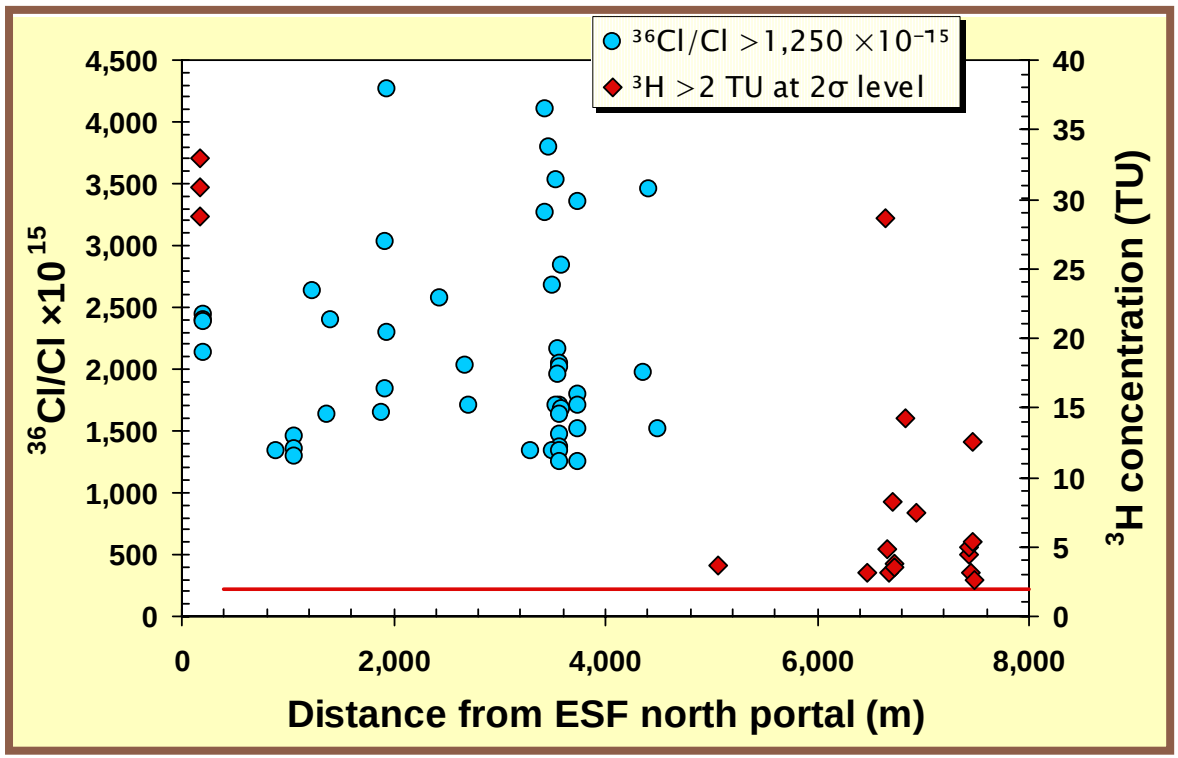
Tritium (^{3}H) in Validation-Study Samples

- **ESF north ramp, Drill Hole Wash, and Sundance samples have ^{3}H concentrations less than or within error of the 2 TU cutoff value**
- **ESF south ramp samples have ^{3}H conc. from <0.1 to 14.3 TU**



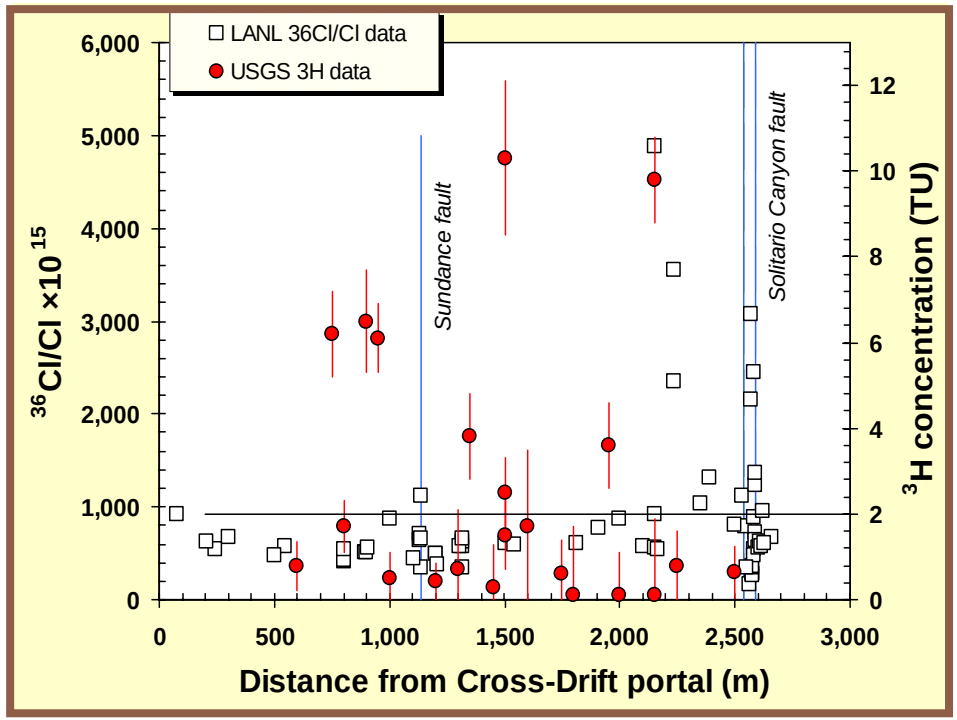
Tritium (^3H) in Validation-Study Samples

- **Distribution of ^3H and ^{36}Cl data indicating rapid percolation are inconsistent**
  - **^3H concentrations imply that rapid percolation is most common in the southern ESF whereas $^{36}Cl/Cl$ ratios imply that rapid percolation is restricted to northern ESF**



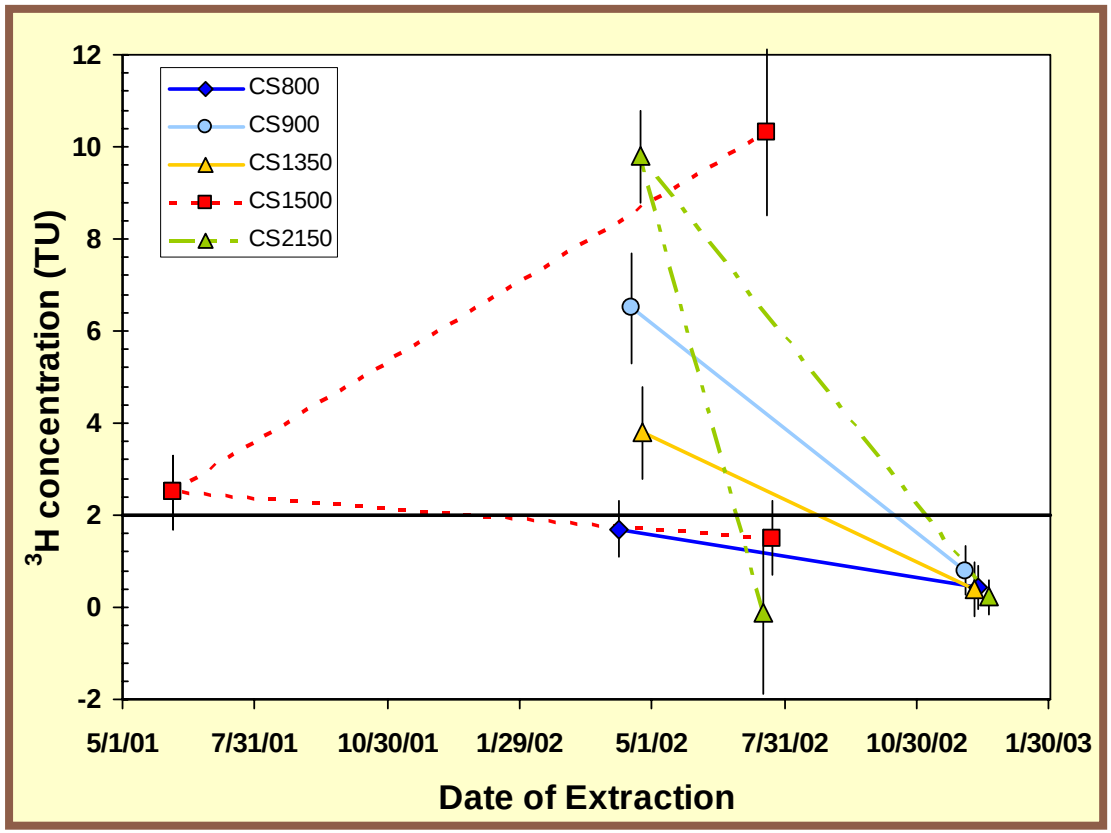
Tritium (3H) in Enhanced Characterization of the Repository Block Cross-Drift Samples

- **^{3}H concentrations range from <0.1 to 10.3 TU**
  - **High values are not clearly associated with major PTn-cutting faults**
  - **Elevated values are difficult to reproduce analytically, although true duplicates have not been tested**
- **^{3}H results generally do not support the distribution of ^{36}Cl/Cl in Cross Drift with one possible exception around station 21+50**



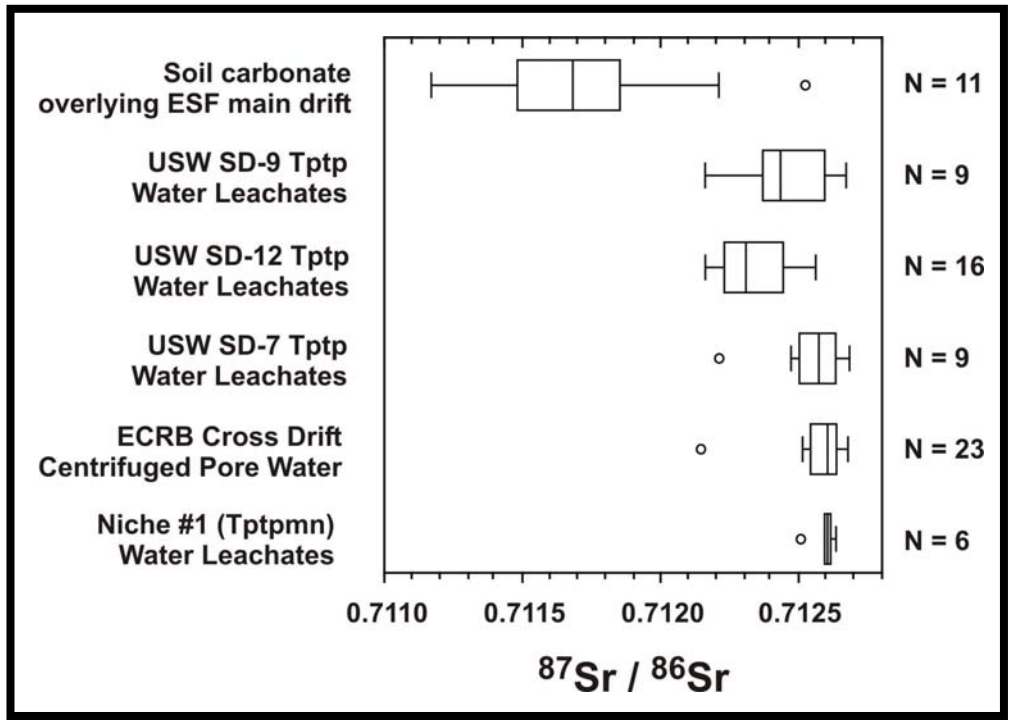
Tritium (^{3}H) "Duplication" Efforts

- **Attempts to reproduce ^{3}H analyses in ECRB Cross Drift core**
  - **Multiple analyses of core from different intervals in 5 boreholes**



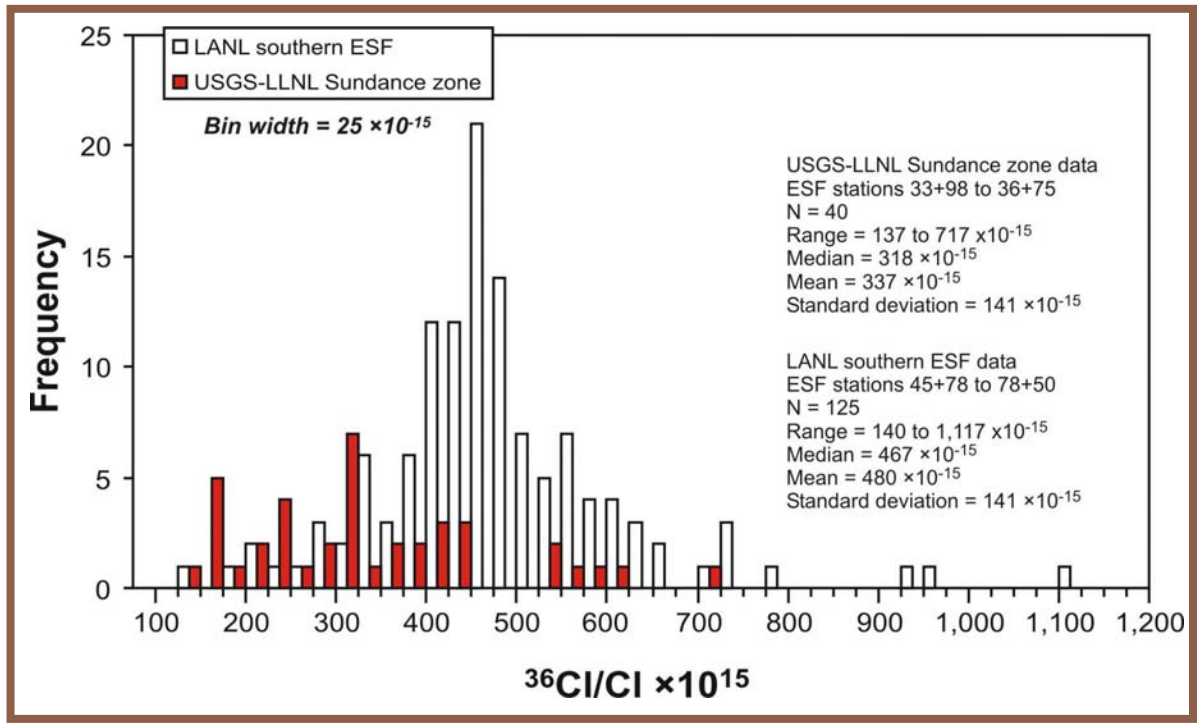
Sr Isotopic Data from Niche #1 Core

- **Most infiltration over ESF main drift will have $^{87}Sr/^{86}Sr$ ratios between ~0.7111 and 0.7122**
- **Water/rock interaction in the PTn results in pore water with elevated $^{87}Sr/^{86}Sr$ values (~0.7122 and 0.7127)**
- **Niche #1 leachates have $^{87}Sr/^{86}Sr$ ratios similar to values elsewhere in the Topopah Spring tuff**
- **Results are consistent with slow percolation through PTn matrix rather than rapid percolation through faulted pathways**



Comparison of U.S. Geological Survey-Lawrence Livermore National Laboratory Sundance and Los Alamos National Laboratory Exploratory Studies Facility South Ramp Data

- **USGS-LLNL data have:**
  - **Much lower Cl concentrations**
  - **Overlapping $^{36}Cl/Cl$ ranges**
  - **Statistically distinct means of 337 $\times 10^{-15}$ and 480 $\times 10^{-15}$**



Comparison of U.S. Geological Survey-Lawrence Livermore National Laboratory Sundance and Los Alamos National Laboratory Exploratory Studies Facility South Ramp Data

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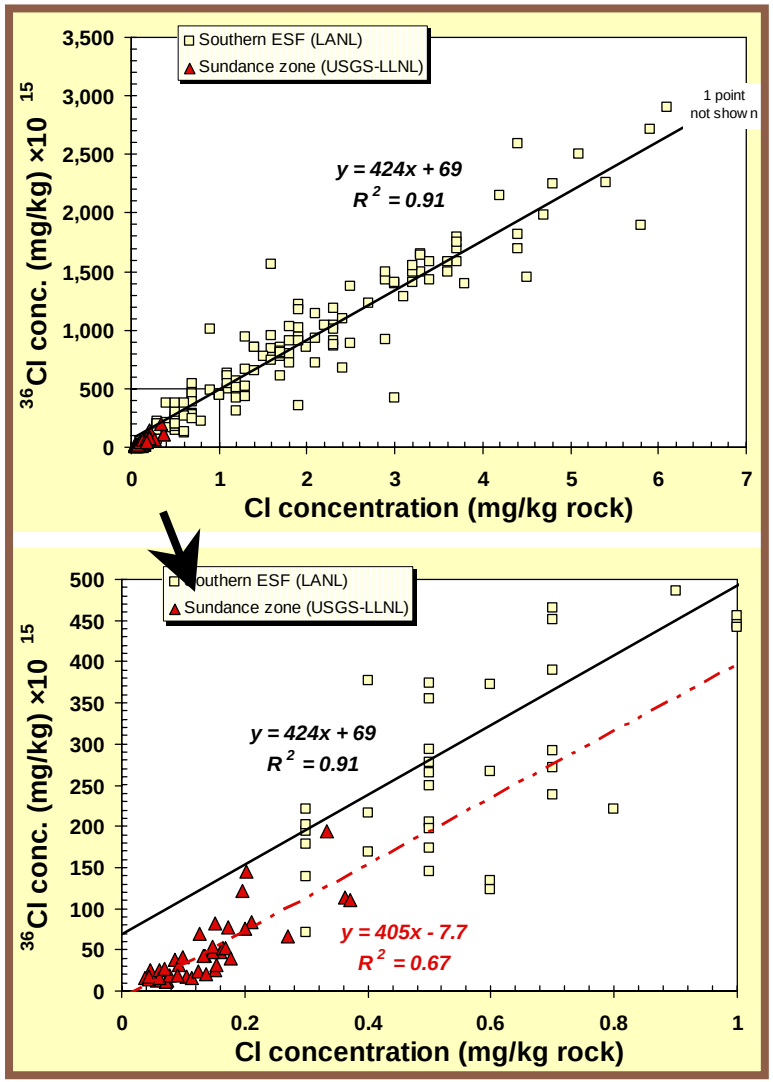

- **Differences are less evident using linear regression of total Cl versus ^{36}Cl concentrations**
  - **Regression slopes represent mean ^{36}Cl/Cl**
  - **Intercept not constrained to pass through 0**

Data set	N	^{36}Cl/Cl slope	^{36}Cl intercept (mg/kg rock)
USGS-LLNL	40	405 ±46	$-7.7 \pm 7.6 \times 10^{-15}$
LANL	125	424 ±12	$69 \pm 31 \times 10^{-15}$

Uncertainties are ±1 standard error

- **Non-zero, positive intercept implies the presence of initial ^{36}Cl when no stable Cl is present**

Comparison of U. S. Geological Survey-Lawrence Livermore National Laboratory Sundance and Los Alamos National Laboratory Exploratory Studies Facility Systematic Samples

- **Means for $^{36}Cl/Cl$ ratios in LANL systematic samples are distinct from USGS-LLNL Sundance samples at high degrees of probability**
  - **Implies that discrepancies are related to analytical rather than sampling problems**