Introduction

Although primarily known for its role in developing the first nuclear weapons, research at Los Alamos National Laboratory encompasses many scientific disciplines including the study of porous media. Very broadly, the twelve thousand employees of Los Alamos National Laboratory focus on nuclear deterrence, nuclear stockpile stewardship, protecting the nation against nuclear threats, understanding emerging threats and cybersecurity, and energy security solutions. The Earth and Environmental Sciences Division at LANL performs much of our research regarding porous materials and the flowing fluids passing through them. We combine our capabilities in field and laboratory measurements with analysis and modeling on high-performance computers to model and predict subsurface movement, reveal climate change drivers and ecosystem impacts, characterize geological greenhouse gas sequestration, improve geothermal and unconventional oil/gas extraction, predict wildland and urban fires, and provide advanced seismic imaging. Our division is broken up into three groups: Computational Earth Science, Earth Systems Observations, and Geophysics. The Computational Earth Science group develops and applies subsurface flow and transport modeling in porous and fractured media, from pore-scale lattice Boltzmann to regional watershed scales with applications to programs involving subsurface water, subsurface contamination of aquifers, CO_2 capture storage, and utilization, energy development in hydrothermal and fossil energy, environmental cleanup, and waste-storage. The Earth Systems Observations group works broadly across the traditional fields of geology, ecology, and atmospheric sciences, with an emphasis on experimental and observational sciences, often coupled to the development and improvement of predictive modeling. The Geophysics group has extensive expertise in seismic and acoustic monitoring, numerical modeling of tectonic processes across a broad spectrum of scales from the near-field (explosive modeling) to the far-field (reservoir- and plate-scale stress fields). In addition, we study the nonlinear properties of Earth materials, model the mechanics of rock fracture, and use seismology to evaluate the interaction of porous rocks and fluids.

Below are a selected number of studies performed by researchers at LANL and their collaborators.

Hydrates

Seafloor methane venting has been observed in many regions of the world oceans, making estimates of global carbon budget difficult. We still do not fully understand the fundamental mechanisms by which methane gas migrates through the deep marine sediments. A key mechanism that affects venting is the formation of methane hydrate, an ice-like solid that forms from a methane–water mixture under pressure and temperature conditions typical of deep marine settings. Here, we study the mechanics of gas percolation under hydrate-forming using experiments at *in situ* temperatures and pressures integrated with pore-scale computational modeling at MIT. We uncovered a phenomenon, which we call crustal fingering, that helps explain how, counterintuitively, hydrate formation may facilitate instead of prevent methane gas migration through deep ocean sediments.

Reference: Fu, X., Jimenez-Martinez, J., Nguyen, T. P., Carey, J. W., Viswanathan, H., Cueto-Felgueroso, L., & Juanes, R. (2020). Crustal fingering facilitates free-gas methane migration through the hydrate stability zone. *Proceedings of the National Academy of Sciences*, *117*(50), 31660-31664.

Unconventional Hydrocarbon Extraction - Shale Gas

Unconventional shale reservoirs currently produce more than 60% of US natural gas, a number predicted to rise to 75% by 2050. Despite this, gas extraction efficiencies from unconventional reservoirs are limited to around 20%, due in large part to limited recovery from the shale matrix during the late stage of well life. In this study, we integrated molecular simulation with high-pressure small-angle neutron scattering (SANS) to examine methane behavior in shale nanopores during pressure cycling. Pressure management is a cheap but effective strategy that operators can employ to improve recovery. However, they currently only have adhoc knowledge on optimizing production using this method. Due to the high penetrating ability of uncharged neutrons, SANS is uniquely capable of characterizing nanopores and hosted fluid behavior *in situ* within a pressure cell. The findings indicate that while high pressures are beneficial for methane recovery from larger pores, dense gas is trapped in smaller, common shale nanopores. For the first time, we present experimental evidence that this deformation exists and proposed a methane-releasing pressure range that significantly impacts methane recovery. These insights help optimize strategies to boost natural gas production as well as better understand fluid mechanics in nanoporous shale.

C. W. Neil, M. Mehana, R.P. Hjelm, M.E. Hawley, E.B. Watkins, Y. Mao, H. Viswanathan, Q. Kang, and H. Xu, Reduced methane recovery at high pressure due to methane trapping in shale nanopores, *Nature Communications Earth & Environment, 2020.*

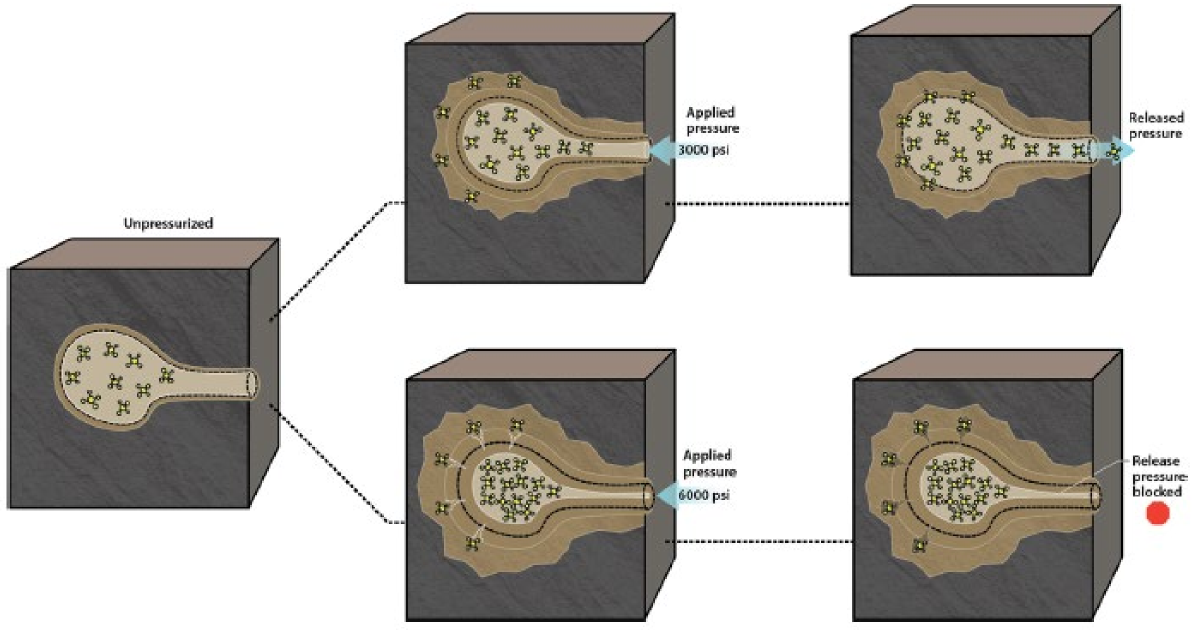

Figure 1:Methane trapping proposed mechanism. Proposed mechanism for dense methane trapping in nanopores within the kerogen matrix. At higher At higher pressures (6000 psi), irreversible deformation of the kerogen matrix results in methane retention in pores even after pressure drawdown.

Spent Nuclear Fuel Disposal – Salt Formation

Can high-level nuclear waste be safely stored in natural salt formations? The answer is unknown, but not for long. Los Alamos researchers in the Earth and Environmental Sciences (EES) Division along with collaborators from Sandia National Laboratories and Lawrence Berkeley National Laboratory designed the **Brine Availability Test in Salt** (BATS) project to better understand the impacts of high-level waste long-term storage in salt formations.

To simulate high-level waste, a specially designed radiative heater was sent into drilled boreholes at the Waste Isolation Pilot Plant (WIPP)—a natural salt formation currently used to permanently disposes of transuranic (TRU) waste that is the byproduct of the nation's nuclear defense program.

High-level waste, primarily from spent nuclear fuel (SNF) generated by nuclear power reactors producing copious amounts of energy, remains a challenge to permanently dispose. In particular, the heat this spent fuel waste generates can complicate long-term (hundreds to thousands of years)salt disposal because of the coupled behavior of thermal–hydrological–mechanical–chemical processes in salt.

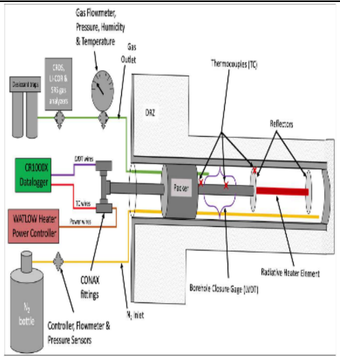

Figure 2: A schematic of a heater design used in the BATS experiments

The BATS project is divided into phases. The overall goal of BATS is to bolster the technical basis for disposal of heat-generating waste in salt. The first phase began in June 2018 and ran through May 2019. The results earned BATS phase 1s the cover feature spot in *Vadose Zone Journal*. Although this was the first experiment in the BATS testing plan, these scientists had already set the research foundations through previous studies. One such study established the multiphase porous flow simulator needed to accurately examine coupled thermal–hydrological–chemical behavior.

“For phase 1s, experimental testing included placing a resistive heater, a 260-W radiative heater, and a 750-W radiative heater within previously drilled horizontal boreholes at WIPP while monitoring temperature and water inflow," said Phil Stauffer of Computational Earth Science (EES-16). "Due to the extremely low permeability of salt, these systems take many years to reach steady state when perturbed by mining activities."

Time is truly the critical factor in the BATS project. To ensure high-level waste can be safely stored for very long periods of time, the researchers will model and experimentally determine how the heat from this waste will play into the salt environment. For example, the researchers noted damaged rock zones around a borehole significantly affects water inflow and thermal pressurization. Increased pressure around highly radioactive waste could lead to undesirable consequences.

Follow-on tests are being designed using information gathered from BATS phase 1s. One such follow-on test started running in January 2020. This test uses a heater design that was proven to be robust in phase 1s and is providing detailed data from 28 newly drilled boreholes.

"A primary goal of LANL's salt repository research is to reduce uncertainty associated with brine availability to repository excavations in salt formations, as part of a larger effort to ensure that we can defend a long-term safety case for disposal of high-level waste in salt," said LANL staff member Phil Stauffer. "By showing the public that we have a firm understanding of how water moves in salt formations, we will be able to more clearly explain how a salt-based geological waste repository will behave over hundreds to thousands of years."

References:

Guiltinan, K. Kuhlman, J. Rutqvist, M. Hu, H. Boukhalfa, M. Mills, S. Otto, D. Weaver, B. Dozier, P. Stauffer; "Temperature response and brine availability to heated boreholes in bedded salt," *Vadose Zone Journal*. April 2020; https://doi.org/10.1002/vzj2.20019.

Johnson; S. Otto, D. Weaver, B. Dozier, T. Miller, A. Jordan, N. Hayes-Rich, P. Stauffer; "Heat-Generating Nuclear Waste in Salt: Field Testing and Simulation," *Vadose Zone Journal*. February, 2019; https://doi.org/10.2136/vzj2018.08.0160.

Johnson, G. A. Zyvoloski, P. Stauffer, "Impact of a porosity dependent capillary function on simulations of porous flow," *Transport in Porous Media*. March 2019; https://doi.org/10.1007/s11242-018-1188-x.

Discrete Fracture Network Modeling

Predicting the passage time of solutes transported through sparse fracture networks is a common and critical challenge in many subsurface applications including the detection of nuclear tests, hydrocarbon extraction, aquifer storage and management, environmental restoration of contaminated fractured media, nuclear waste storage, and geological CO_2 sequestration. Using, dfnWorks, LANL's R&D100 winning, high-fidelity discrete fracture network simulator, Jeffrey Hyman (EES-16) led an international team of researchers in collaboration with a cross-divisional cohort of LANL scientists to identify key geophysical attributes that control the long-term behavior of solutes transported in subsurface fractured media. The results were published in three recent articles.

In the first article, published in Physical Review Letters*,* purely advective transport was simulated through high-fidelity three-dimensional fracture networks (Figure 1a) and analysis of the spatial memory of the particle transport showed that individual particle motion can be accurately modelled stochastically using a time domain random walk conditioned on the steady-state velocity data. The approach identifies advective tortuosity, the velocity point distribution and the average fracture link length as the key physical quantities for the prediction of first passage times in fractured media. Using this approach, a new theory for the evolution of first passage times to converge towards a power-law tailed first passage time distribution was derived and showed outstanding agreement with the high-fidelity simulation results.

Reference: Hyman, Jeffrey D., Marco Dentz, Aric Hagberg, and Peter K. Kang. "Emergence of stable laws for first passage times in three-dimensional random fracture networks." *Physical review letters* 123, no. 24 (2019): 248501.

In the second article, published in Geophysical Research Letters, the effects of solutes exchanging between flowing regions (fractures) and non-flowing regions (matrix) via molecular diffusion, are also considered. A long-standing question in this area of research is the relative impact of matrix diffusion on power-law scaling in solute transport breakthrough curve tails, which are observed in field and laboratory experiments and predicted by the theory published in the article described above. While classical theory requires that matrix diffusion produces a decay rate of time to the -3/2 power, deviations are commonly observed. This article addresses the question through the development of a new theory that elucidates how interactions between two important physical processes (advection and matrix diffusion) can produce either the classical -3/2 decay rate or alternative decay rates based on two dimensionless parameters. A comparison of the theoretical predictions against numerical simulations (figure 1 - inset) shows that the two are in excellent agreement.

Reference:
Hyman, J. D., Rajaram, H., Srinivasan, S., Makedonska, N., Karra, S., Viswanathan, H., & Srinivasan, G. (2019). Matrix diffusion in fractured media: New insights into power law scaling of breakthrough curves. *Geophysical Research Letters*, *46*(23), 13785-13795.

In the third article, published in Transport in Porous Media, the interplay between network structure and multiphase flow effects was systematically explored using an ensemble of semi-generic fracture networks. Advancing our understanding of how water is displaced by CO_2 in fractured media is a critical step towards geologic sequestration of CO_2 in the subsurface. Figure 2 shows six snapshots from one simulation and highlights how the complicated fracture network structure and the properties of the multiphase flow combine to form intricate and complex fluid flow behavior. This study presents the first calculations of multi-phase flow in a three-dimensional discrete fracture network. The research was enabled by combining dfnWorks with the multi-physics/multiphase porous flow simulator, FEHM, which has been developed for over 30 years LANL. The study shows that single phase flow properties can provide a lower bound on more complicated multiphase flow properties in these complex networks.

Reference: Hyman, J. D., Jimenez-Martinez, J., Gable, C. W., Stauffer, P. H., & Pawar, R. J. (2020). Characterizing the Impact of Fractured Caprock Heterogeneity on Supercritical CO_2 Injection. *Transport in Porous Media*, *131*(3), 935-955.

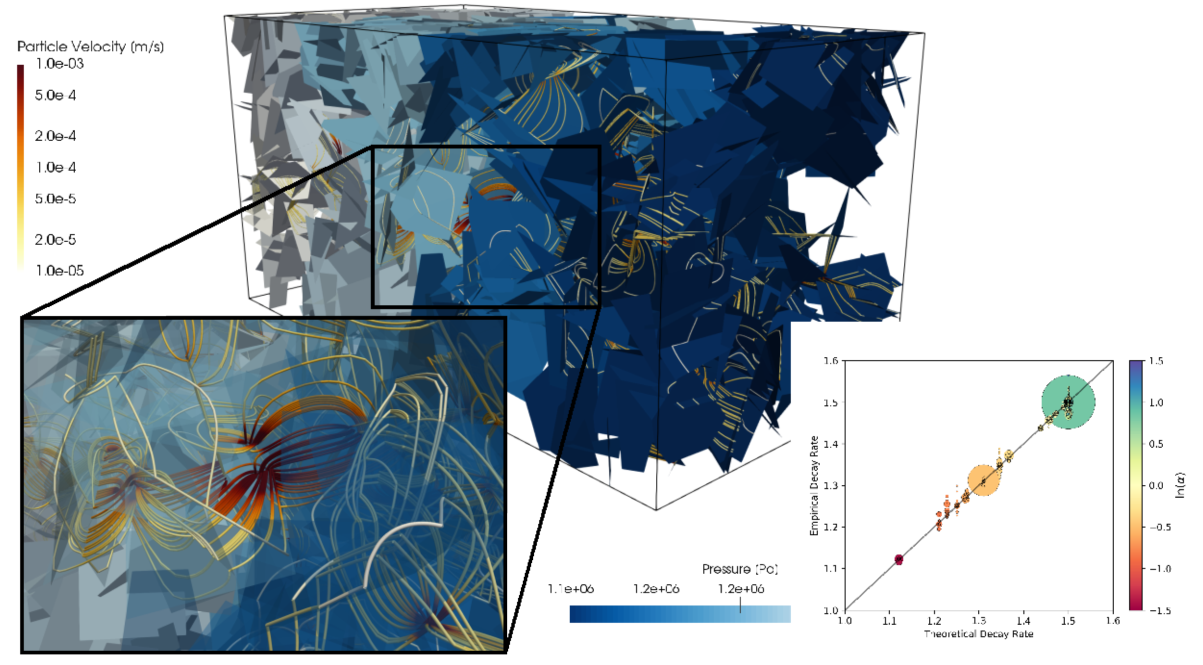

Figure 3: Particle pathlines (colored according to their velocities) within a fracture network (fractures are colored by pressure). The complex network structure along with individual fracture geometry results in a spatially variable velocity field within individual fractures that is sampled by the ensemble of particles. The inset in the lower right corner shows the agreement between the theoretical predictions and high-fidelity simulation results.

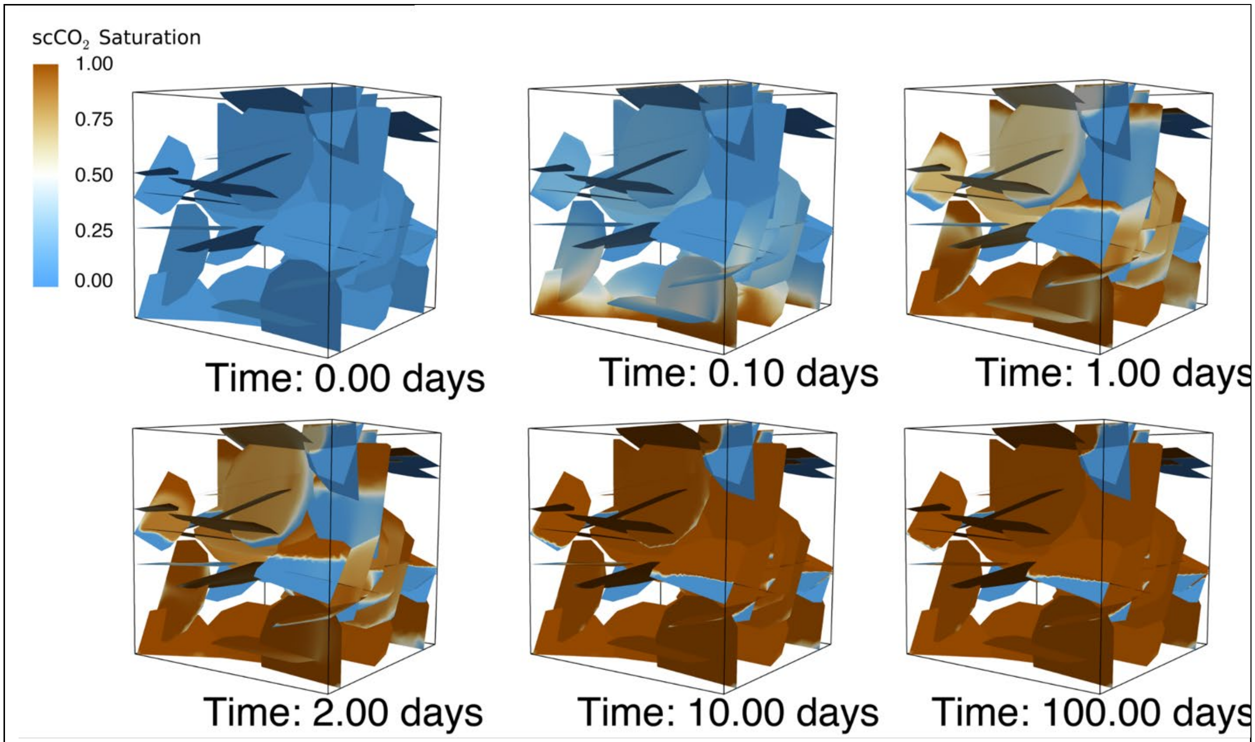

Figure 4: Snapshots of scCO$_2$ displacing water in a DFN with constant permeability. scCO$_2$ is injected at a constant rate into the domain through fractures connected to the bottom. The scCO scCO$_2$ rises through the network displacing water. At this point, the majority of the domain is fully saturated with scCO$_2$ but there are some regions (hanging fractures) that remain fully saturated with water.

Historical data leads to better modeling for radioactive gas seepage from underground nuclear tests

Using historical U.S. nuclear test program data from 16 underground nuclear explosions with similar geology and test setup, LANL researchers analyzed discriminating factors that heavily impact whether an underground nuclear explosion will result in radioactive gas seepage.

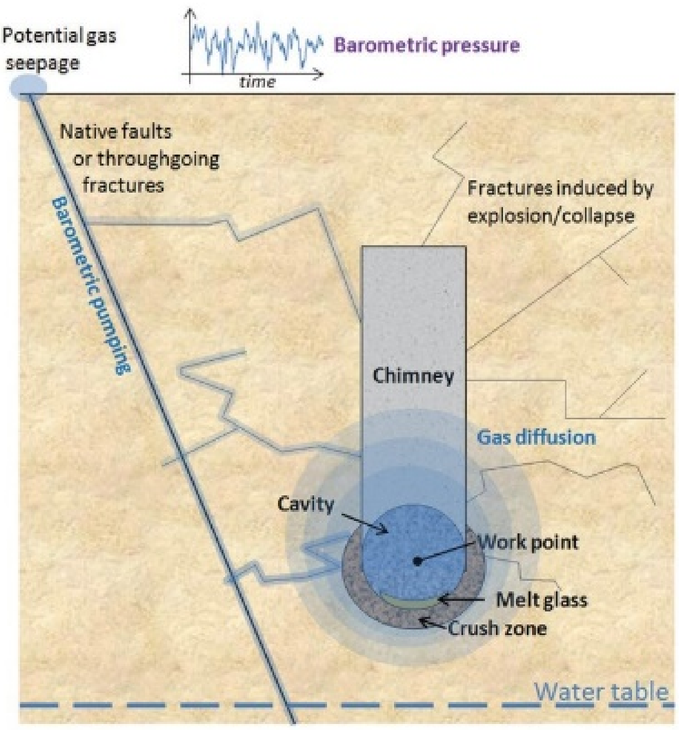

Figure 5: Conceptualization of subsurface damage and gas migration pathways to the ground surface by barometric pumping after an underground nuclear explosion (from Jordan et al., 2014).

Verification of test ban treaties

Detecting, confirming, and understanding underground nuclear explosions is of vital importance to national security. This new research will aid in the verification of nuclear test ban treaties.

Detecting radioactive gases seeping into the atmosphere can provide "smoking gun" evidence of an underground nuclear explosion. These explosions generate a suite of radioactive gases, including noble gases that do not interact easily with other materials and some of which decay slowly enough to persist until transported to the ground surface over days to months, a process known as late-time seepage. Detection of particular noble gases with specific isotopic ratios seeping into the atmosphere from a suspected test site can positively identify the occurrence of a nuclear explosion when little other evidence is available.

Factors that predict late-time seepage

The rate at which radioactive gases seep from underground nuclear explosions to the ground surface depends on the geology, atmospheric conditions, and the nuclear device and its yield. The researchers used subsurface gas transport models that account for these factors to understand how radioactive gases move from the depth of the nuclear explosion to the ground surface.

The researchers selected 16 historical underground nuclear explosions that occurred at the Nevada National Security Site (NNSS): five with late-time releases and 11 without. Potential factors that could impact late-time gas seepage include: (1) post-explosion surface damage, (2) proximity to faults, (3) barometric pressure trends before and after the test date, (4) gas seepage soil properties (permeability and air-filled porosity), and (5) geologic stratigraphy. The importance of these factors was evaluated by the researchers using the historical data along with numerical and analytical modeling.

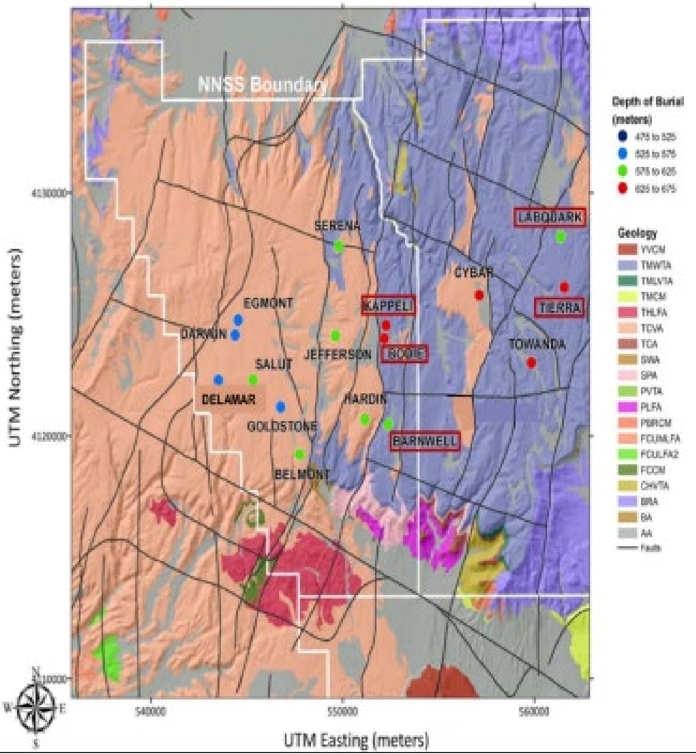

Figure 6: Map of the surficial hydrostratigraphy of Pahute Mesa (NSTec, 2014) with underground nuclear explosion locations. Black lines indicate faults and white lines indicate NNSS boundaries and operational areas (Area 20 on left Area 19 on right). Red boxes indicate where late-time seeps were observed.

A key finding of the research is the amount of air space available in the rock (air-filled porosity) is inversely correlated with whether an underground nuclear explosion would result in late-time seepage. Less air-filled porosity increases the potential for late-time seepage. Another key finding is the time of year when the underground nuclear explosion occurs is important. This is because barometric pressure trends change seasonally, with larger variations during the fall and winter. Therefore, explosions conducted in the fall and winter at the NNSS (when barometric gas transport efficiency is higher) are more likely to have late-time seeps.

Surprisingly, neither the severity of post-explosion surface damage nor proximity to existing natural faults with surface expressions are strong indicators of late-time seeps. Numerical and analytical models are able to discriminate between underground nuclear explosions resulting in late-time seeps and those without. However, the numerical models did not accurately predict the exact timing of the releases. Analytical modeling indicated that knowledge of rock fracture widths created by the explosions (information not available at all 16 explosions analyzed) would improve the identification of underground nuclear explosions resulting in late-time seeps.

References:

Bourret, S. M., Kwicklis, E. M., Harp, D. R., Ortiz, J. P., Stauffer, P. H. 2020. “Beyond Barnwell: Applying lessons learned from the Barnwell site to other historic underground nuclear tests at Pahute Mesa to understand radioactive gas-seepage observations.” *J. Environ. Radioact.* 222C. https://doi.org/10.1016/j.jenvrad.2020.106297

Harp, D. R., Bourret, S. M., Stauffer, P. H., and Kwicklis, E. M. (2020).“Discriminating underground nuclear explosions leading to late-time radionuclide gas seeps.” *Geophysical Research Letters*, 47, e2019GL086654. https://doi.org/10.1029/2019GL086654