Centimeter

Inches

THE DEVELOPMENT OF A VERSATILE FIELD PROGRAM FOR MEASURING TRITIUM IN "REAL-TIME"

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ABSTRACT

Robust sample handling and liquid scintillation counting procedures have been developed to routinely monitor tritium in the field relative to the 20,000 pCi/L drinking water standard. This procedure allows tritium to be monitored hourly during 24 hour drilling operations at depths in the saturated zone potentially contaminated by sub-surface nuclear weapons testing at the Nevada Test Site. Using retrofitted shock hardened and vibration damped counters and strict analytical protocols, tritium may be measured rapidly in the field under hostile conditions. Concentration standards and "dead" tritium backgrounds are prepared weekly in a central laboratory and delivered to remote monitoring locations where they are recounted daily as a check on counter efficiency and calibration. Portable counters are located in trailers and powered off a battery pack and line filter fed by mobile generator. Samples are typically ground-waters mixed with drilling fluids returned after circulation through a drill string. Fluids are aerated and "de-foamed", filtered, mixed with scintillation cocktail and subsequently dark-adapted before counting. Besides meeting regulatory requirements, "real-time" monitoring affords drilling and field personnel maximum protection against potential exposure to radionuclides; for routine operations, tritium activities may not exceed a 10,000 pCi/L threshold.

INTRODUCTION

Tritium is being utilized as an early indicator of potential migration of radionuclides in groundwater as part of a CERCLA driven remedial investigation/feasibility study designed to characterize the subsurface impact of nuclear weapons testing at the Nevada Test Site (NTS), Nye County, Nevada. Known as a conservative tracer of contaminant transport under saturated and partially saturated conditions, tritium is a good choice because it is completely soluble in and in equilibrium with groundwater.

A field based tritium monitoring program has been implemented to measure tritium activity in fluids circulated through the drill string during the drilling and development of monitoring wells. Under the auspices of the U. S. Department of Energy, Nevada Operations Office Underground Test Area Remedial Investigation/Feasibility Study (UGTA RI/FS), monitoring wells are being constructed close to NTS nuclear test centers. The monitoring program insures that radioactivity is not being transported to potential receptors by groundwater.

TRITIUM AND GROUNDWATER

Tritium has a half-life of 12.43 years. It is produced naturally in the atmosphere by energetic reactions of cosmic rays with nitrogen by the following reaction:

$$^{14}N + {}^{1}n = {}^{12}C + {}^{3}H$$

As an isotope of hydrogen, tritium can readily incorporate itself in the water molecule as HTO. Typically, 99.9% of the tritium occurs as molecular HTO. Natural production of tritium in rainwater is approximately 10 tritium units (TU) or 3.2 pCi/L. Tritium levels in rainwater rose in excess of 103 TU between 1952 and 1969 during the period of the atmospheric testing of thermonuclear weapons before returning to modern values between 10-20 TU in central continental areas and approximately 5 TU in precipitation near the California Coast (Drever, 1988).

The State of Nevada drinking water standard of 20,000 pCi/L is equivalent to 6250 TU. Tritium levels in drinking water wells vary from 0.1 to 19.8 TU. By contrast, field tritium scans of groundwaters collected from "hot" wells near sites of subsurface detonations range from 144 to 15,000,000 TU (Davisson, et al., 1994).

While tritium is a conservative radionuclide under saturated conditions, under partially saturated or unsaturated conditions, tritium may be less easily mobile. Tritium introduced in the vadose zone is derived from water condensed from the explosion cavity as well as from tritium promptly injected along fractures during the explosion (Nimz and Thompson, 1992). As such, the distribution of tritium is variable and strongly influenced by the phenomenology of the detonation. The extreme thickness of the unsaturated zone and the NTS (nearly 600 m), aridity (<15cm/year annual precipitation) and high daily maximum temperatures (13 to 40^{o}C) together support the conclusion that downward water movement through the unsaturated zone is negligible and will not significantly remobilize tritium (Smith, 1994). While vapor phase transport in the unsaturated zone may redistribute tritium, Tyler et al. (1992) suggests the effects are minimal.

Due to its unique occurrence and abundance at NTS, nearly conservative behavior under saturated conditions and ready detection, tritium serves as an excellent proxy for groundwaters potentially impacted by post-nuclear test contamination. Stand-alone tritium monitoring of groundwater under saturated condition is necessary and sufficient for the prompt detection of soluble radionuclides.

FIELD BASED TRITIUM MONITORING

Hourly monitoring is required for all down-hole operations producing drilling fluids or detergent effluent. In February 1992, an initial field tritium monitoring program was instituted using several existing liquid scintillation counting (LSC) instruments. Because each existing instrument was different during this initial phase, instrument specific counting/monitoring protocols were established. Counting facilities were set up inside two - 30 foot wheeled modular trailers.

High operating cost associated with the NTS rotary drilling program is a driver for reducing stand-down time for both the crew and rig. The second modular trailer facility is used alternately with the first to expedite monitoring activities at multiple drill sites.

Radiological technicians are trained for continuous 24 hour, three shift operations. The technicians are trained to collect representative drilling fluid samples, filter and prepare a 5 ml sample for counting. The sample is prepared by adding 10 ml Ultima Gold Scintillation Cocktail, dark adapted the mixture for 5 minutes in the machine, and counted for a 15 minutes interval. Any anomalous measurements are recounted.

Due to the extremely hostile deployment during the first nine months of operation, difficulties were encountered. Line fluctuations from erratic power generation caused background interferences. Transport over rough roads damaged delicate instrumentation. Additives to the drilling fluid interfered with the tritium background determination. Wind, rain, snow, mud and dust contributed to siting and logistical problems in remote areas. Finally, operator/technician turn-over necessitated on-going training sessions.

By December 1992, the first of three Packard 1600TR liquid scintillation counters (LSC) were installed in the field. One LSC was assigned to each field facility and the third used as a backup for the two counters assigned to field facilities. To enhance the durability for frequent relocation, each counter was 'hardened' by strapping the lead housing around the detector securely to the counter frame. In addition, shock mounts were installed between the counter and the cart support in each of the four corners. To avoid chemiluminescence interference due to the emulsifiers in the drilling fluid, correction software and quench correction hardware and software options were also installed.

Air-synthetic foam detergent is circulated as a drilling fluid for drilling in the vadose zone. Defoaming agents are required to prepare the sample for filtration when air-foam is used. A low pressure filter press is used to accelerate the filtration of high particulate drilling fluid samples.

An instrument generator replaced a common generator responsible for creating power spikes capable of damaging the photomultipliers of the LSC. A line conditioner is used as a final filter in advance of the LSC.

A new set of calibration standards and a "dead water" background samples are delivered to the drill site weekly to insure LSC instrument calibration. The set includes an efficiency standard and three reference standards. The efficiency standard is used to calculate the machine efficiency. The activity of the reference standards are 10,000, 20,000 and 40,000 pCi/L. The standards are traceable to the National Institute of Standards and Technology (NIST). The set is counted by every shift operator and logged on the respective shift reporting log. This daily record is FAXed to the Field Over-Sight Supervisor for review. However, any set of three measurements trending to 10,000 pCi/L is reported immediately to the on-site lead drilling geologist. An extrapolated trend exceeding tritium concentration in excess of 20,000 pCi/L is reported to the Department of Energy. If the observed tritium activity exceeds 100,000 pCi/L, drilling operation are suspended. (Note: The action levels for this operation were revised as of 3/28/93).

CONCLUSION

Field tritium monitoring provides analysis in "real-time" during drilling and well testing under extremely hostile conditions. An effective monitoring program assumes "dead" (background radioactive) local formation water is the make-up water for all drilling fluids. Under unsaturated or partially saturated conditions, stand-alone tritium monitoring is less diagnostic as a indicator of potential contamination. At NTS water movement through the vadose zone is minimal; vapor transport is similarly negligible. Radionuclides that are injected or have volatile gaseous precursors may be heterogeneously redistributed in the vadose zone and missed by a stand-alone tritium monitoring program. In the unsaturated zone, tritium monitoring should be supplemented with an allied monitoring program incorporating gamma spectroscopy analysis of ^{137}Cs; ^{137}Xe(T1/2=3.82 minutes) is the volatile gaseous precursor of ^{137}Cs and will be distributed in the vapor phase before decay to the longer lived fission product (Smith, 1994).

A companion program is charged with developing field instrumentation for detection of radionuclides in the vadose zone. An LSC capable of alpha-beta discrimination shows promise. Another proof-of-principle study is an evaluation of a 30% efficient Canberra germanium detector for monitoring gamma emitting radionuclides in the field. We ultimately hope to discriminate between anthropogenic radionuclides from weapons testing and natural radioactivity introduced as ^{40}K, ^{232}Th, and ^{238}U by bentonite drilling fluids used for completions within incompetent geology.

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