

**Soil Activation and Groundwater Contamination at
Brookhaven National Laboratory, Upton, New York - 8080**

Douglas E. Paquette, PG
Brookhaven National Laboratory
Building 120, Upton, NY 11973

Chek Beng Ng, PE
New York State Department of Environmental Conservation
625 Broadway, Albany, NY 12233

Gail Penny
Federal Project Director
United States Department of Energy – Brookhaven Site Office
P.O. Box 5000, Upton, NY 11973

ABSTRACT

In November 1999, tritium (H-3) was detected in the groundwater near one of Brookhaven National Laboratory's (BNL) accelerator experiments at concentrations above the 20,000 pico curie per liter (pCi/L) Maximum Contaminant Level (MCL). Sodium-22 (Na-22) was also detected in the groundwater, but at concentrations well below the 400 pCi/L MCL. An investigation into the source of the contamination revealed that the tritium and sodium-22 originated from activated soil shielding located adjacent to the g-2 target building where approximately five percent of the beam was inadvertently striking one of the beam-line magnets. Rainwater was able to infiltrate the activated soils and carry the tritium and sodium-22 into the groundwater. The highest tritium level detected in groundwater during the 1999 investigation was nearly 1.8 million pCi/L. To prevent additional rainwater infiltration into the activated soil shielding, a concrete cap was constructed over the soil shielding in December 1999. Other corrective actions included refocusing the beam and improved beam loss monitoring to reduce additional soil activation, stormwater management improvements, and additional groundwater monitoring.

From 2001 through 2004, three high concentration zones (or slugs) of tritium were observed passing through the groundwater monitoring well network immediately downgradient of the source area, with a maximum observed concentration of 3.4 million pCi/L. Some of the tritium that was previously leached from the activated soil was trapped in the vadose (unsaturated) zone soils directly above the water table after then cap was installed. A portion of this residual tritium was later mobilized into the groundwater during periods of high groundwater table elevations, which can occur following heavy seasonal rainfall. Monitoring results for the past two years indicate that the amount of tritium being released from the vadose zone is decreasing, with tritium concentrations consistently below 100,000 pCi/L. The tritium plume is currently 550 meters long, and is located entirely in the central portion of the BNL site. The plume has not impacted any of the Laboratory's drinking water supply wells. Contaminant transport modeling suggests that the tritium plume will attenuate entirely in the central portion of the BNL site by years 2010-2015.

INTRODUCTION

Brookhaven National Laboratory (BNL) is owned by the U.S. Department of Energy (DOE), and conducts research in physical, biomedical and environmental sciences and energy technologies. BNL is located in Upton, Suffolk County, New York, near the geographic center of Long Island. The U.S. Army occupied the site, formerly known as “Camp Upton,” during World Wars I and II. Between the wars, the Civilian Conservation Corp maintained the site. The site was transferred to the Atomic Energy Commission in 1947, to the Energy Research and Development Administration in 1975, and to DOE in 1977.

The BNL site is situated over three highly permeable water-bearing units that are designated by the U.S. Environmental Protection Agency (EPA) as a “sole source aquifer” system [1]. BNL was added to New York State’s Inactive Hazardous Waste Sites List in 1980 and to the Federal National Priorities List in 1989. BNL has a comprehensive and ongoing environmental cleanup program conducted under a tri-party Federal Facilities Compliance Agreement, also known as the Interagency Agreement (IAG). This IAG integrates the requirements of CERCLA, the corrective action requirements of the Resource Conservation and Recovery Act, DOE cleanup authorities under the Atomic Energy Act, and any corresponding New York State regulations. The signatories of this agreement are U.S. DOE, U.S. EPA, and the New York State Department of Environmental Conservation (NYSDEC).

ACCELERATOR FACILITY

The BNL Alternating Gradient Synchrotron (AGS) facility is used to conduct accelerated particle beam experiments. The facility is located in the central area of the BNL site, approximately 2.5 km north (upgradient) of the BNL southern property boundary. The g-2 experiment was on an independent beam line originating from the AGS, and operated from April 1997 through April 2001.

Radionuclides can be produced in some of the soil shielding used along accelerator beam lines by the interaction of secondary particles (primarily neutrons) created when proton beams strike beam-line targets and magnets. The high-energy particles emerging from a target are eventually attenuated in the surrounding shielding. This attenuation process results in the creation of radioactive atoms many feet into the soil shielding. Radioactivity results from a process whereby the high-energy particle slows down by striking an atom and breaking the atom into pieces, some of which are unstable. This is also referred to as spallation. Typical long-lived radioactive atoms created in soil and concrete shielding by the activation process are tritium (H-3), with a half-life of 12.3 years, and sodium-22 (Na-22), with a half-life of 2.7 years. The infiltration of rainwater through the activated soil can transport tritium and sodium-22 to the groundwater [2]. Whereas tritium is highly mobile and can be found in groundwater at concentrations greater than the 20,000 pico curie per liter (pCi/L) Maximum Contaminant Level (MCL), sodium-22 is only detected close to the source area, and usually at concentrations that are much less than the 400 pCi/L MCL. Other radionuclides detected in the soil are short-lived, with half-lives of less than one month, and are usually less mobile. To protect groundwater resources, impermeable caps constructed of concrete or geotextile fabric are installed in these beam loss areas to prevent rainwater infiltration.

During the design of the g-2 experiment, beam losses were expected to produce activated soil below the target building and the nearby beam stop. The building structure and the underlying concrete pad protect the activated soil below the target building, and an impermeable cap was

constructed over the g-2 beam stop to protect the soil shielding from rainfall infiltration. Groundwater monitoring wells were installed to verify the effectiveness of these controls.

INITIAL DETECTION OF TRITIUM IN GROUNDWATER

In November 1999, tritium was detected in the groundwater near the g-2 experiment at concentrations above the 20,000 pCi/L MCL. Following the discovery, an investigation into the source of the contamination revealed that the tritium originated from activated soil shielding adjacent to the g-2 experiment's VQ-12 magnet [3]. It was determined that approximately five percent of the beam was inadvertently striking the VQ-12 magnet, which is located inside the g-2 target building. Soil samples collected through the tunnel wall adjacent to the VQ-12 magnet confirmed the extent of soil activation [3,4]. The previously installed concrete base pad and beam stop cap did not protect this new soil activation area. Figure 1 is a simplified cross-section view of the beam line and associated activated soil shielding. Activation of the soils surrounding the VQ-12 magnet area may have begun during the g-2's first major experiment run period in June and July 1997, and the release of tritium and sodium-22 may have begun soon after that time. The contaminated leachate that was generated would likely have penetrated the concrete base-pad near VQ-12 through small penetrations that remained after a 1 m by 61 m strip was removed from the concrete pad to allow for the installation of a sheet pile wall during beam line construction. Records indicate that the concrete base-pad was resealed on the sheet-pile side that faces the target building, but not on the side facing the soil sheilding. The cut in the base pad and the sheet pile wall provided a conduit for the rainwater leachate to penetrate quickly into the ground below the VQ-12 area. The highest tritium level detected in groundwater during the 1999 investigation was approximately 1.8 million pCi/L. Sodium-22 was also detected in the groundwater, but at concentrations below the 400 pCi/L MCL.

To prevent additional rainwater infiltration into the activated soil, a concrete cap was constructed over the VQ-12 area in December 1999 (Figure 2). Other corrective actions included refocusing the beam and improved beam loss monitoring to reduce additional soil activation, stormwater management improvements, and additional groundwater monitoring. The g-2 experiment concluded its operations in April 2001, and the facility is being maintained for possible future use.

SOURCE AREA AND PLUME SURVEILLANCE

Since the discovery of the g-2 tritium plume in November 1999, BNL has been monitoring the source area surveillance wells on a quarterly basis. Monitoring of the plume is accomplished using a combination of permanent and temporary wells. Monitoring data indicate that the high levels of tritium have entered the groundwater as a series of slug releases (Figure 3). This resulted in the creation of four small-scale zones of high tritium concentrations, or hot spots, within the tritium plume where concentrations exceed 100,000 pCi/L. The extent of the g-2 tritium plume during the fourth quarter of 2005 is depicted on Figure 4. The plume is approximately 550 m long, and positioned entirely within the central portion of the BNL site. Because tritium is an integral part of some of the water molecules, it migrates in the aquifer at the same rate as groundwater, which is approximately 0.2 m/day or 72 m/yr.

POST-CAP RELEASE OF TRITIUM TO GROUNDWATER

Prior to cap installation over the VQ-12 source area in December 1999, tritium was leached from the activated soil shielding into the vadose zone soil above the water table and then into the groundwater. When tritium first enters the groundwater system, it generally forms a thin plume

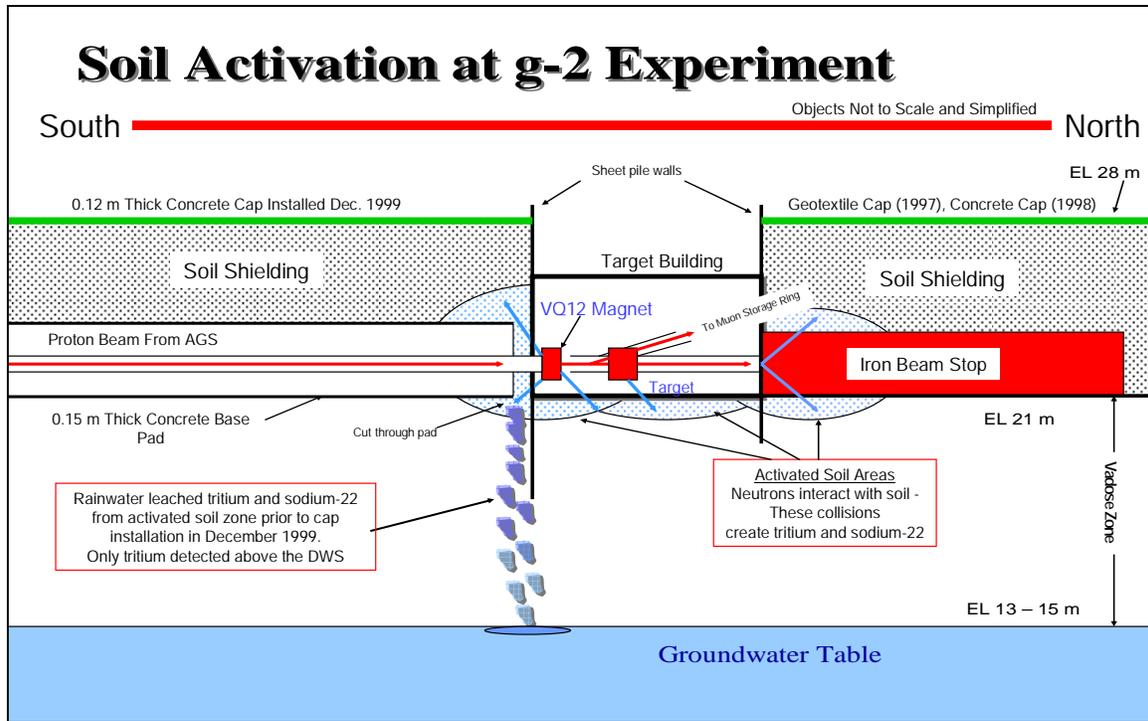


Fig. 1. Simplified Cross-Section View through the g-2 Source Area.



Concrete Cap

Fig. 2. Concrete Cap at the g-2 Source Area.

at the top of g-2 Target Building in the VQ-12 source area, the vad Concrete Base Pad is 6 m to 8 m thick, and the time of travel for tritiated water to reach the water table was probably less than one month. However, once the cap was installed and water infiltration ended, the driving force for vertical migration was reduced, and some of the tritium was “trapped” near the capillary fringe above the water table. With low vadose zone seepage rates, the residual tritium would be slowly released to the aquifer with natural fluctuations in the water table elevation, and would produce a “tailing” effect in the release of tritium to groundwater. A similar pattern for the slow

release of residual tritium from the vadose zone has been observed at the BNL High Flux Beam Reactor (HFBR) and the Brookhaven Linac Isotope Producer.

Water levels in the central BNL area in mid-2000, mid-2001, and mid-2003 were near the highest observed in almost 50 years of record for the BNL site, to a level of approximately 15 m above mean sea level (see water table trend shown on Figure 4). Approximately one year after each of these periods of high water table elevations, elevated tritium concentrations were observed in the first set of downgradient surveillance wells. Over time, the amount of tritium remaining in the vadose zone near the water table is expected to decrease by this flushing mechanism and by natural radioactive decay. Although the water table increased to nearly 15 m above mean sea level during three periods since 2004, tritium levels in all but one set of quarterly samples from source area surveillance wells have been less than 100,000 pCi/L, with concentration of only 39,400 pCi/L being detected in July 2007. These reductions in tritium concentrations suggest that the amount of residual tritium available in the deep vadose zone is decreasing.

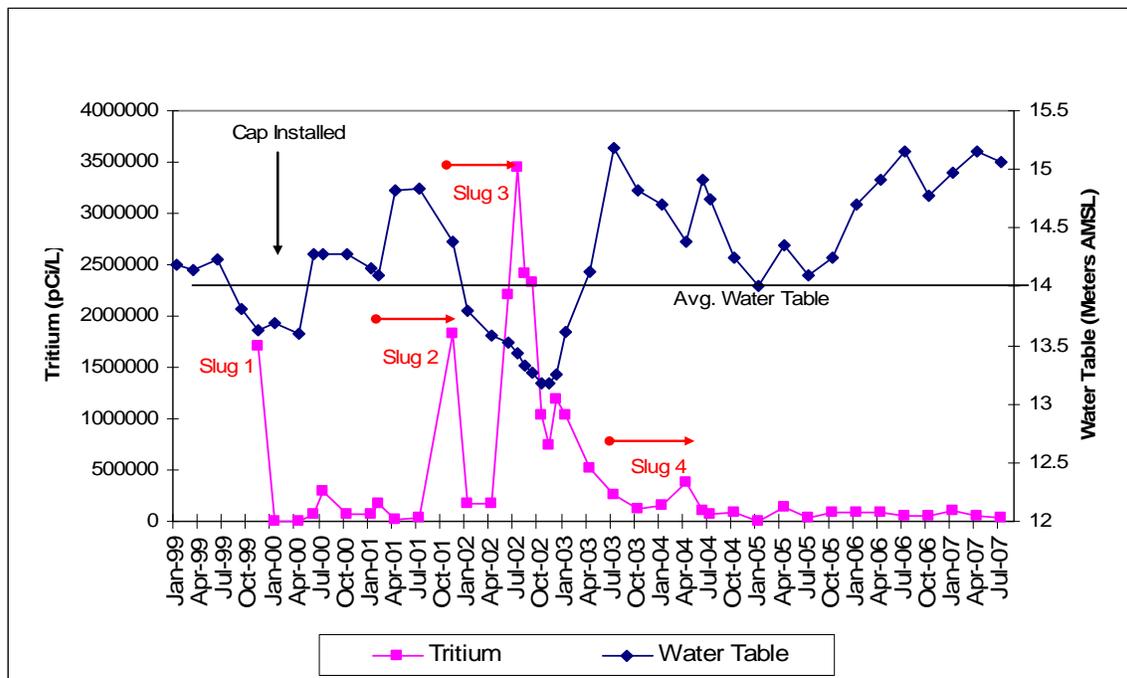


Fig. 3. Maximum tritium concentrations observed in groundwater downgradient of the VQ-12 source area from 1999 through 2007. Red arrows represent approximately one year of travel time from the source area to the first set of downgradient monitoring wells.

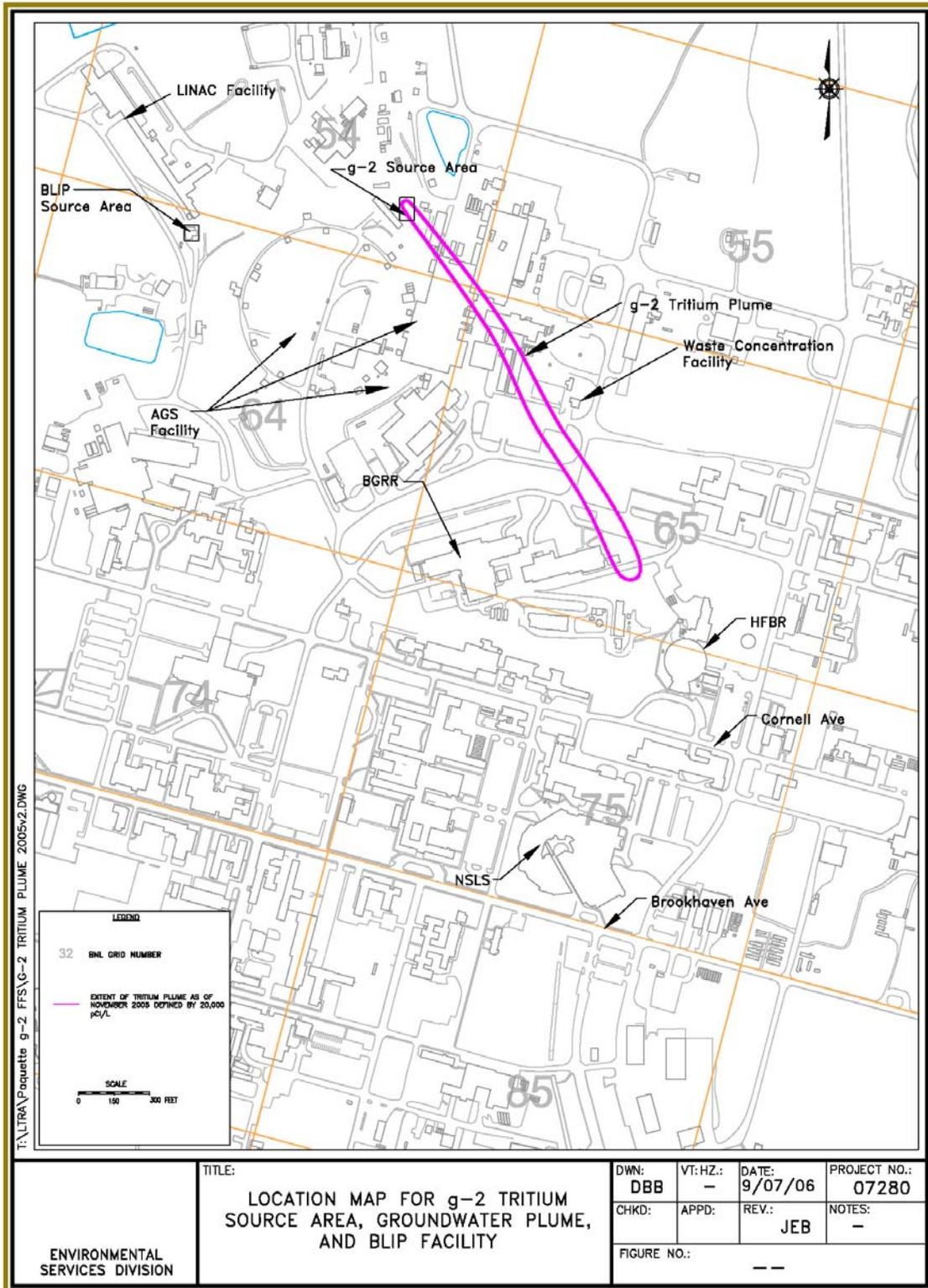


Figure 4. Location Map for g-2 Tritium Source Area and position of the g-2 tritium plume in late 2005.

CONTAMINANT TRANSPORT MODELING

A new sub-model of the calibrated BNL Regional Groundwater Flow Model [6] was used as a tool to predict the movement of the plume and reductions in tritium concentrations due to natural radioactive decay and dispersion in the aquifer. The same model was used to successfully predict the fate of the HFBR tritium plume. Based on the model results, tritium concentrations in the plume are expected to decrease to less than the 20,000 pCi/L MCL near BNL's Cornell Avenue (Figure 6) between years 2010 and 2015, assuming that there are no additional significant tritium releases from the source area [4]. Cornell Avenue is located in the central area of the BNL site, and is over 1.6 km away from the property boundary line. The g-2 plume will not impact any public or private drinking water supply wells.

REMEDIAL ACTIONS

Following the concurrence from the NYSDEC, a Record of Decision (ROD) was signed by the U.S. DOE and U.S. EPA in early 2007 [7]. This ROD requires continued routine inspection and maintenance of the impermeable cap, groundwater monitoring of the source area to verify the continued effectiveness of the storm water controls, and monitoring the tritium plume until it attenuates to less than the 20,000 pCi/L MCL. Monitoring of the source area will continue for as long as the activated soils remain a threat to groundwater quality. Contingency actions have been developed that require the evaluation of unexpected future releases from the source area or if the tritium plume does not attenuate as predicted by the groundwater model.

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