# 3 EMISSION/EFFLUENT SOURCES AND PATHWAYS

## 3.1 INTRODUCTION

BNL estimates potential exposures from radioactive and chemical substances that could be received by humans, terrestrial and aquatic plants, and flora and fauna through various pathways. To calculate the exposures, the character of the pollutants emitted (e.g., identity, amount, rate of release, chemical form, etc.) and how the pollutants are subsequently absorbed, retained, and passed along by the various possible exposure pathways, must be researched. Sources of radioactive and chemical emissions and effluents from Laboratory facilities are described below. A general description of the primary exposure pathways to members of the public and environment is also provided.

## 3.2 PATHWAYS

Chemicals and radionuclides released into the environment can move through the biosphere by a number of routes and can eventually lead to exposure of humans, animals, and vegetation. These routes can be direct, as in the inhalation of contaminated air or ingestion of contaminated drinking water; or indirect, involving many complex levels of the food chain and different transport mechanisms. Exposure is defined as the interaction of an organism with a physical or chemical agent of interest. An exposure pathway is identified based on:

- An examination of the type, location, and source (contaminated soil, raw effluent, etc.) of contaminants
- Principal release mechanisms
- Probable environmental fate and transport (including persistence, partitioning, and intermediate transfer) of contaminants of interest
- Location and activities of potentially exposed populations

Mechanisms that influence the fate and transport of chemical and radiological contaminants through the environment and also influence the amount of exposure a person might receive at various receptor locations are listed below. While processes that move contaminants through the atmosphere and hydrosphere tend to reduce their concentrations, many pathway components or processes that move contaminants through the food chain to humans can cause bioaccumulation. Once a radionuclide or chemical is released into the environment, it may be:

- *Transported* (e.g., migrate downstream in solution or on suspended sediment, travel through the atmosphere, or be carried off site in contaminated wildlife)
- *Physically or chemically transformed* (e.g., deposition, precipitation, volatilization, photolysis, oxidation, reduction, hydrolysis, or radionuclide decay)
- *Biologically transformed* (e.g., biodegradation)
- *Accumulated in the receiving media* (e.g., strongly absorbed in the soil column, stored in organism tissues)

The atmosphere and surface water are the primary pathways for movement of radioactive materials and chemicals from the Laboratory site to the public. Figure 3-1 illustrates the potential routes and exposure pathways to humans. The significance of each pathway is determined by comparing

measurements and calculations that estimate the amount of radioactive material or chemical transported along each pathway with the concentrations or potential doses to environmental and public health protection standards or guides. Pathways are also evaluated based on prior studies and observations of radionuclide and chemical movement through the environment and food chains. Calculations based on effluent and emission data show the expected concentrations beyond the BNL site to be low for all Laboratory-produced radionuclides and most chemicals. Frequently, concentrations are below the level that can be accurately detected by monitoring technology. To ensure that radiological and chemical analyses of samples are sufficiently sensitive, minimum detection limits of key radionuclides and chemicals have been established at levels well below applicable health standards.

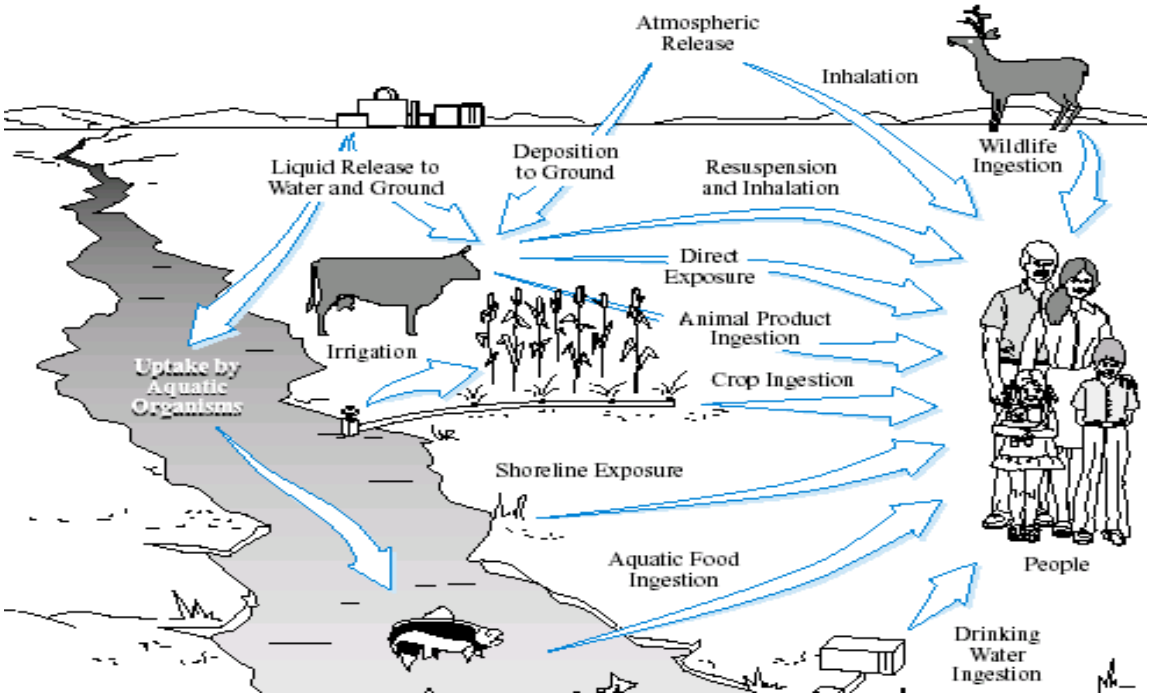



Figure 3-1. Primary Exposure Pathways to Humans.

## 3.3 SOURCES

### 3.3.1 Airborne Emissions – Radioactive

Federal air quality laws and DOE regulations that govern the release of airborne radioactive material include 40 CFR 61 Subpart H: National Emission Standards for Hazardous Air Pollutants—part of the Clean Air Act, and DOE Order 458.1 (2011), *Radiation Protection of the Public and Environment*. Facilities with emissions that have the potential to deliver a radiation dose greater than 0.1 millirem per year to a member of the public must be continuously monitored for emissions. Facilities capable of delivering radiation doses below that limit require periodic, confirmatory monitoring. BNL has one facility that is continuously monitored, the Brookhaven Linac Isotope Producer (BLIP). Periodic monitoring is conducted at one active facility, the Target Processing Laboratory (TPL), and one inactive facility, the High Flux Beam Reactor (HFBR). Figure 3-2 indicates the location of each of these monitored facilities.

Figure 3-2. Potential Air Emission Release Points Subject to Monitoring.

Over the years, there have been significant decreases in the types and quantities of radiological emissions from BNL operations that are described further in this chapter. The most significant sources of radionuclide emissions are from the BLIP and the TPL. The BLIP typically contributes the largest fraction (99 percent or more) of the total annual effective dose equivalent to the maximally exposed individual residing at the BNL site boundary. The primary radionuclide releases from Laboratory operations are carbon-11 (C-11), oxygen-15 (O-15), and tritium (H-3). Table 3-1 presents the airborne radionuclide releases from monitored facilities in calendar year 2012 (most current data).

Table 4-1. Airborne Radionuclide Releases from Monitored Facilities.

| Facility | Nuclide | Half-Life | Ci Released |
|---|---|---|---|
| BGRR | Tritium | 12.3 years | 3.46E-01 |

| | | | |
|---|---|---|---|
| HFBR | Tritium | 12.3 years | 4.06E-01 |
| BLIP | Carbon-11 | 20.4 minutes | 1.91E+03 |
| | Oxygen-15 | 122 seconds | 3.88E+03 |
| | Tritium | 12.3 years | 9.98E-04 |
| TPL - Bldg. 801 | Arsenic-74 | 17.8 days | 3.11E-10 |
| | Bromine-77 | 57.0 hours | 3.69E-08 |
| | Germanium-68 | 270.8 days | 3.99E-08 |
| | Selenium-75 | 119.8 days | 8.94E-09 |
| Total | | | 5.79E+03 |

Notes:

Ci = 3.7E+10 Bq

BGRR = Brookhaven Graphite Research Reactor

BLIP = Brookhaven Linac Isotope Producer
HFBR = High Flux Beam Reactor (operations were terminated in November 1999)

| Facility | Nuclide | Half-Life | Ci Released |
|---|---|---|---|
| BGRR | Tritium | 12.3 years | 1.40E-01 |
| HFBR | Tritium | 12.3 years | 8.06E-01 |
| BLIP | Carbon-11 | 20.4 minutes | 1.60E+03 |
| | Oxygen-15 | 122 seconds | 3.31E+03 |
| | Tritium | 12.3 years | 2.42E-04 |
| TPL - Bldg. 801 | Arsenic-74 | 17.8 days | 2.45E-10 |
| | Bromine-77 | 57.0 hours | 2.59E-08 |
| | Germanium-68 | 270.8 days | 5.93E-08 |
| | Selenium-75 | 119.8 days | 6.75E-09 |
| Total | | | 4.90E+03 |

Notes:
Ci = 3.7E+10 Bq
BGRR = Brookhaven Graphite Research Reactor
BLIP = Brookhaven Linac Isotope Producer
HFBR = High Flux Beam Reactor
TPL = Target Processing Laboratory

TPL = Target Processing Laboratory

Other facilities that have the potential for radiological emissions are associated with accelerator operations, such as the Alternating Gradient Synchrotron (AGS) Booster, the 200-MeV Linac Accelerator, and associated experimental facilities. Emissions from these facilities are extremely low and are insignificant contributors to off-site dose. The other potential source of airborne radionuclide emissions is laboratory hoods, where work with dispersible radionuclides is performed. Small quantities of radioactive materials are typically used in these hoods, usually on the order of micro to millicurie quantities. Compliance with National Emission Standards for Hazardous Air Pollut-

ants (NESHAPs) regulations for these sources is demonstrated annually through the use of an inventory system, as allowed under Appendix D of the NESHAPs regulations. Environmental surveillance air monitoring conducted at the site boundaries also provides verification that off-normal emissions from these sources have not occurred.

New facilities or planned activities that will generate environmental releases of airborne radionuclides are reviewed for NESHAPs compliance. The review documents the details of the operation generating the release, the source term involved, proposed effluent control equipment, and the calculated dose impact from the proposed release. The evaluation is also used to assess the need for possible modifications to the environmental monitoring program.

The following sections briefly describe the primary sources of radioactive air emissions from BNL operations.

Table 3-1. Airborne Radionuclide Releases from Monitored Facilities.

#### *3.3.1.1 Brookhaven Medical Research Reactor*

The BMRR was permanently shut down in December 2000 due to a reduction of research funding. During operation, the BMRR was fueled with enriched uranium, moderated and cooled by “light” (ordinary) water, and was operated intermittently at power levels up to 3 megawatts (MW), thermal. Air from the interior of the containment building was used to cool the neutron reflector surrounding the core of the reactor vessel. As air was drawn through the reflector, it was exposed to a neutron field, resulting in activation of the argon fraction of the air. This produced argon-41 (Ar-41), an inert, radioactive gas (half-life: 1.8 hours). After passage through the reflector, the air was routed through a roughing filter and a high-efficiency particulate air (HEPA) filter to remove any particulate matter. Charcoal filters were also used to remove radioiodine produced during the fission process. Following filtration, the air was exhausted to the atmosphere through a 150-foot stack adjacent to the reactor containment building and continuously monitored for Ar-41 emissions.

After the BMRR discontinued operation, continuous Ar-41 monitoring was reduced to semi-annual monitoring to confirm that radionuclide concentrations remained below detection limits. In January 2003, the remaining fuel was removed from the BMRR reactor vessel, eliminating the last significant source for radionuclide emissions. The sole remaining BMRR emission source was evaporation of the cooling water, which contained the radioactive isotope H-3 (half-life: 12.3 years), produced by neutron activation when the BMRR operated.

In January 2005, the Environmental Protection Agency (EPA) approved BNL’s petition to discontinue emissions monitoring at the BMRR. As a result, sample collection was discontinued in 2006, and all removable radioactive materials were shipped to an off site disposal facility. Currently, the BMRR remains in a “cold” shutdown mode as a radiological facility and periodic inspections are conducted.

#### *3.3.1.2 High Flux Beam Reactor*

When the HFBR operated, “heavy” water was used as a neutron moderator and fuel coolant. Heavy water is water composed of a nonradioactive isotope of hydrogen known as deuterium. When exposed to neutron fields generated inside a reactor vessel, deuterium becomes activated and produces radioactive tritium. As a result of the transfer of fuel elements from the reactor, the spent fuel storage pool contained tritiated heavy water (HTO) from the HFBR system. In 1997, a plume of tritiated groundwater was traced back to a leak in the pool. Consequently, the HFBR was put in standby mode, the pool was pumped out, and the HTO from the pool was properly disposed of as

radioactive waste. The pool was then repaired and double lined in accordance with Suffolk County Article 12 regulations (SCDHS 1993) and remained empty while the facility was in a standby mode. The HFBR continued in standby mode until November 1999, when DOE declared that it was to be permanently shut down. Residual tritium in water in the reactor vessel and piping systems continued to diffuse into the building's air through valve seals and other system penetrations, though emission rates continued to be much lower than during years of operation.

From 2002 through 2008, emissions from the HFBR facility were monitored via air sampling of the building at a frequency of one week per month. In 2009, the frequency of monitoring was increased to bi-weekly to better account for changes in tritium emissions during planned decontamination and dismantlement activities. In 2010, the HFBR was disconnected from its 100-meter stack and a new HFBR exhaust system was installed in 2011.

Consistent with the HFBR Long-Term Surveillance and Maintenance Manual, prior to scheduled surveillance and maintenance activities, air samples are now collected from outside the HFBR confinement using a permanently installed sample port. The samples are collected by bubbling air through a container of water using a fritted sampling device to ensure better collection efficiency. Samples are analyzed in-house for tritium to ensure air quality within the building is acceptable to permit staff entry. Samples are also collected from the fixed sampling system for mold analysis. Additionally, samples are collected one week per month from the HFBR exhaust system using a standard desiccant sampling system for tritium analysis. Desiccant samples are analyzed by an off-site contract laboratory.

#### *3.3.1.3 Brookhaven Linac Isotope Producer*

Protons from the BNL Linear Accelerator (Linac) are sent via an underground beam tunnel to the BLIP, where they strike various metal targets to produce new radionuclides for medical diagnostics. The activated metal targets are transferred to the TPL in Building 801 for separation and shipment to various radiopharmaceutical research laboratories. During irradiation, the targets become hot and are cooled by a continuously recirculating water system. The cooling water also becomes activated during the process, producing secondary radionuclides. The most significant of these radionuclides are oxygen-15 (O-15, half-life: 122 seconds) and carbon-11 (C-11, half-life: 20.48 minutes). These isotopes are released as gaseous, airborne emissions through the facility's 33-foot stack. Emissions of these radionuclides are dependent on the current and energy of the proton beam used to manufacture the radioisotopes.

In 2012 (most current information available), BLIP operated over a period of 30 weeks, during which 1,595 Ci of C-11 and 3,305 Ci of O-15 were released. Tritium produced from activation of the target cooling water was also released, but in a much smaller quantity, 2.42 E-4 Ci. The combined emissions of C-11 and O-15 were 15 percent less than the combined emissions of these isotopes in 2011.

#### *3.3.1.4 Former Evaporator Facility*

Liquid waste generated on site that contained residual radioactivity was accumulated at BNL's Waste Concentration Facility (WCF). At this facility, the water was treated to remove suspended solids and was then transferred to the former Evaporator Facility, where it was converted to steam and released as an airborne emission. The BNL Evaporator Facility was constructed primarily to reduce the amount of tritiated water released to the Peconic River through the Laboratory's Sewage Treatment Plant (STP).

The Evaporator Facility began processing wastewater in 1995 and ceased operations in 2001. Wastes are now processed through solidification and disposed at an approved off-site facility. Under BNL's Environmental Restoration Project, the facility was demolished in 2011.

#### *3.3.1.5 Target Processing Laboratory*

Metal targets that have been irradiated at BNL's BLIP facility are transported to the Laboratory's TPL where isotopes are chemically extracted for radiopharmaceutical production. In the past, airborne radionuclides released during the extraction process were drawn through multistage HEPA and charcoal filters and then vented to the HFBR stack through an underground system of duct work. In 2009, it was decided to decommission and demolish the stack and three new stacks were built. The types of radionuclides released depend on the isotopes chemically extracted from the irradiated metal targets, which can change from year to year. Annual radionuclide quantities released from this facility are very small, typically in the µCi to mCi range. In 2012, the total release from the TPL was 0.0944 µCi.

#### *3.3.1.6 Linear Accelerator (Linac)*

The Linac produces beams of polarized protons of energies up to 200 Me V for use at both the Collider-Accelerator Department (CAD) and BLIP. Due to the composition of the beam and the energies involved, production of airborne radionuclides through air activation and/or spallation interactions is possible. The most significant production point of airborne radionuclides inside the tunnel occurs where the beam crosses an air gap as it enters the BLIP vacuum system. These radioactive products are available for atmospheric release via the tunnel ventilation exhaust stack, located adjacent to the BLIP building. However, since the exhaust fan has been turned off and the nuclides are permitted to decay within the tunnel, the potential for emissions is small.

#### *3.3.1.7 Alternating Gradient Synchrotron Cooling Tower 2*

Magnets used to steer particle beams from the Laboratory's AGS experience significant heating and are cooled via a recirculating, non-contact water loop. Under certain conditions (such as high-energy proton operations), low concentrations of radioactive elements may be produced in the cooling water when it circulates in the vicinity of the beam line. Radioisotopes that exist as gases may be liberated from the water when exposed to air during circulation in the outdoor cooling tower, resulting in an airborne emission. The radionuclides that are likely to be released via this mechanism include oxygen-14 ($t_{1/2}$ = 1.2 minutes), O-15 ($t_{1/2}$ = 2.1 minutes), nitrogen-13 ($t_{1/2}$ = 10 minutes), and C-11 ($t_{1/2}$ = 20 minutes). Tritium is also present and may be emitted from the tower as water vapor in microcurie quantities per year. Modeling using EPA CAP88-PC software indicates that the typical annual dose to the maximally exposed individual from this source is approximately 0.00002 mrem. Note that this cooling tower is only an airborne radionuclide emission source during those times when the AGS C-line is in use. Cooling Tower 2 does not service any other beam lines.

#### *3.3.1.8 Additional Minor Sources*

Several research departments at BNL use designated fume hoods for work that involves small quantities of radioactive materials (in the µCi to mCi range). The work typically involves labeling chemical compounds and transferring material between containers using pipettes. Due to the use of HEPA filters and activated charcoal filters, the nature of the work conducted, and the small quantities involved, these operations have a very low potential for atmospheric releases of any significant quantities of radioactive materials. Compliance with NESHAPs Subpart H is demon-

strated through the use of an inventory system that allows an upper estimate of potential releases to be calculated. Facilities that demonstrate compliance in this way include Buildings 463, 490, 490A, 510, 535, 555, 725, 801, and 830, where research is conducted in the fields of biology, medicine, high energy physics, chemistry, photon science, advanced technology, and environmental sciences.

### 3.3.2 Airborne Emissions-Nonradioactive

Various state and federal regulations governing nonradioactive airborne releases require facilities to conduct periodic or continuous emissions monitoring in order to demonstrate compliance with emission limits. BNL's Central Steam Facility (CSF) is the only facility that requires monitoring for nonradiological emissions. The Laboratory has several other emission sources subject to state and/or federal regulatory requirements that do not require emission monitoring.

The CSF supplies steam for heating and cooling to all major facilities on site through an underground steam distribution and condensate grid. The combustion units at the CSF emit oxides of nitrogen, sulfur dioxide, oxides of carbon, and particulate matter. Continuous emissions monitors are used on two boilers to measure $NO_x$, and particulate (i.e., opacity). Data are reported quarterly to EPA and the New York State Department of Environmental Conservation (NYSDEC).

### 3.3.3 Liquid Effluents

BNL's State Pollutant Discharge Elimination System (SPDES) permit provides the basis for regulating wastewater effluents at the Laboratory. The SPDES permit establishes release concentration limits and dictates monitoring requirements. BNL's SPDES permit was issued in 1995, and last renewed in 2009. In 2007, NYSDEC initiated a review of BNL's SPDES permit because more than 10 years had passed since its last renewal. The Laboratory completed an application and submitted it to NYSDEC in 2007. In 2008, NYSDEC requested additional information regarding the use and discharge to the STP and other outfalls of corrosion-control chemicals used in cooling water systems. The permit was significantly modified in June 2009 and renewed, effective March 1, 2010.

The modified permit proposed significant reductions in the concentration of six metals (copper, iron, lead, mercury, nickel, and zinc) discharged from BNL's STP to the Peconic River. Studies were completed to assess the sources of these metals and to evaluate the feasibility of achieving new discharge limits. In order to achieve compliance with the required discharge limits at the STP, a decision was made to modify the STP from a surface water discharge to a groundwater discharge system. The modified treatment process is scheduled to be completed by September 2014, and a modified SPDES permit application to reflect this change was prepared and submitted to NYSDEC for review in May 2012. A Notice of Complete Application prepared by NYSDEC along with a copy of BNL's updated SPDES Permit was received in November 2013.

#### *3.3.3.1 BNL Sewage Treatment Plant (Outfall 001)*

Sanitary and process wastewaters generated by Laboratory operations are conveyed to the BNL STP for subsequent treatment prior to discharge to the Peconic River. In 1997, the STP underwent significant construction modifications and was upgraded from a primary plant (i.e., separation of settleable solids and floatables) to a tertiary treatment system (i.e., biological reduction of organic matter and reduction of nitrogen). This treatment process became fully functional in 1998. In 2000, additional changes to the plant and sewage system were initiated. These changes included replacing or lining the sewage collection system and replacing the former anaerobic digester with an aerobic digester. These modifications were completed in early 2002. In 2006, the primary clarifier was re-

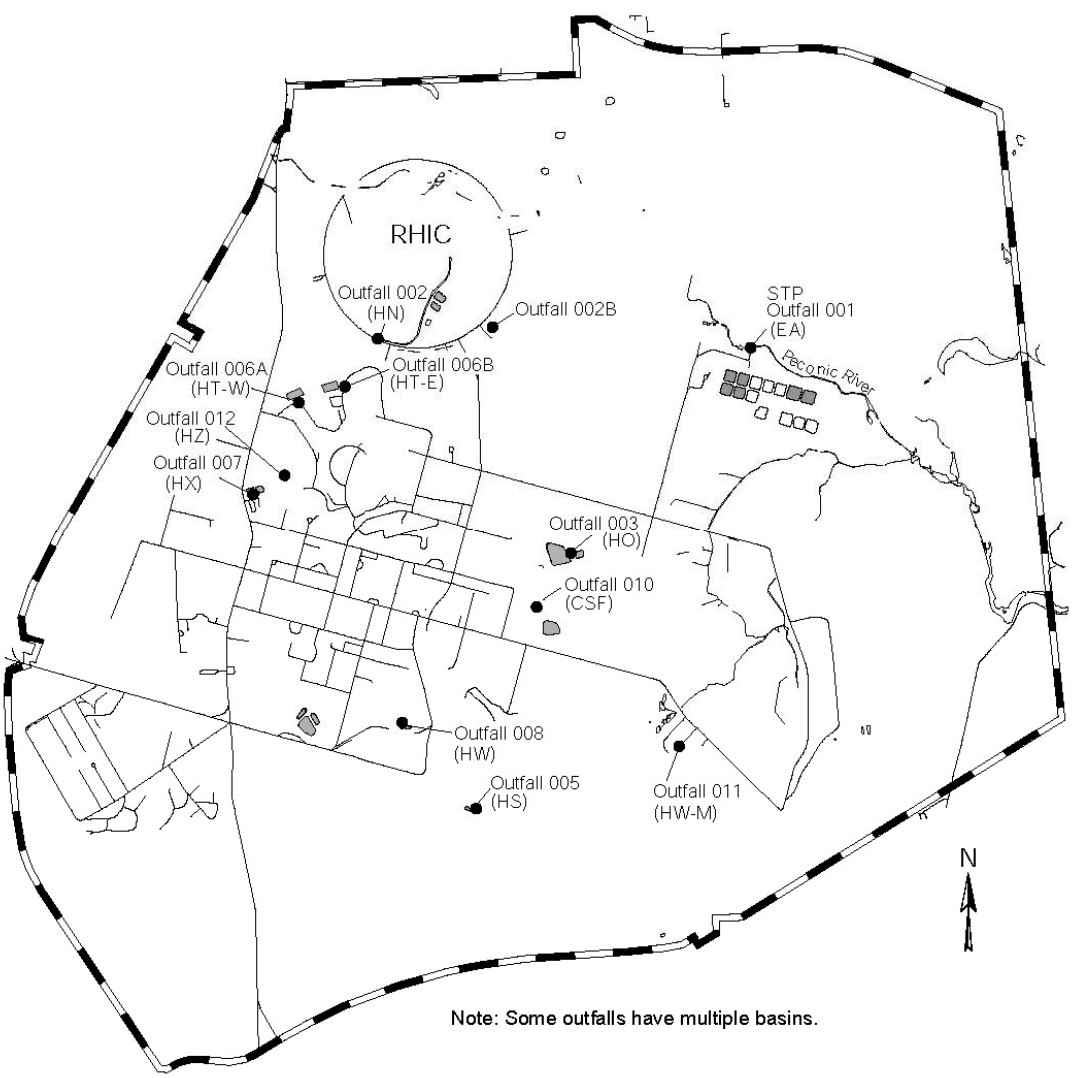
moved from the treatment sequence to permit the entry of all waste products into the aeration process. This change was necessary to enhance nitrogen removal by providing more nutrients for the biological organisms to be used during the denitrification step. The STP has a design capacity of 3.0 million gallons per day and receives sanitary and certain process wastewaters from Laboratory facilities for treatment before discharge into the Peconic River.

#### *3.3.3.2 BNL Recharge Basins and Stormwater (Outfalls 002–008, 010-012)*

Recharge basins are used for the discharge of "clean" wastewater streams, including once-through cooling water, stormwater runoff, and cooling tower blowdown. Figure 3-3, on the following page, depicts the locations of BNL's recharge basins and stormwater outfalls.

- Recharge Basins HN, HT-W, and HT-E receive once-through cooling water discharges generated at the Collidar Accelerator Department (CAD) and the Relativistic Heavy Ion Collider (RHIC), as well as cooling water tower blowdown and stormwater runoff.
- Recharge Basin HS receives predominantly stormwater runoff, once-through cooling water from Building 555 (Chemistry Department), and minimal cooling tower blowdown from the National Synchrotron Light Source (NSLS) and NSLS-II.
- Basin HX receives Water Treatment Plant filter backwash water. Recharge Basin HO receives cooling water and cooling tower discharges from CAD and formerly from the HFBR, as well as stormwater runoff. Discharges from the CAD consist of once-through domestic water used to cool the main magnet heat exchanger located in Building 911.

In addition, several other recharge areas are used exclusively for discharging stormwater runoff.




Note: Some outfalls have multiple basins.

## 3.4 ENVIRONMENTAL RESTORATION MONITORING

In 1980, the U.S. Congress enacted the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA, also known as Superfund) to ensure that sites with historical contamination were cleaned up and to hold the responsible party liable for the cleanup. CERCLA established the National Priorities List (NPL). The NPL is a list of sites nationwide where cleanup of past contamination is required. In November 1989, BNL, as one of a number of sites on Long Island, was listed on the NPL. Much of the contamination at the Laboratory is due to past practices for handling, storing, and disposing of chemical and radiological materials, as well as to accidental spills.

The purpose of BNL's Environmental Restoration (ER) program, established in 1991, was to begin characterizing and removing sources of contamination on site, as well as treat groundwater and soils that were contaminated from past practices. Initially, historical facility records and environmental sampling were used to determine where contamination might be present on site (i.e., Areas of Concern), and these areas were geographically grouped into Operable Units (OUs). Since 1991, cleanup efforts have included remediating contaminated soils and river sediments, capping landfill areas, installing groundwater treatment systems, and decommissioning and decontaminating former waste handling and reactor facilities. In addition, public water service was provided to homeowners immediately south and east of the Laboratory to replace the use of private potable supply wells.

In 2006, the program was divided into two programs: the Environmental Restoration Project (ERP), to complete the decontamination and decommissioning (D&D) of the HFBR and BGRR reactor facilities and ancillary structures, and the Groundwater Protection Group, to carry out long-term surveillance and maintenance (S&M) activities associated with the groundwater and surface projects remediation programs. After completion of the D&D activities in 2012, the ERP was disbanded and the Groundwater Protection Group was also tasked with the long-term S&M activities for the former HFBR and BGRR reactor facilities.

Many remediation techniques can result in temporary increases in contaminant effluents or emissions, and therefore require monitoring to minimize potential impacts. The BNL Environmental Monitoring Program supports restoration activities by selecting monitoring locations and determining what media samples will best assist ERP in evaluating the impact of restoration activities.

## REFERENCES

40 CFR 61, Subpart H. 1999. U.S. Environmental Protection Agency. *"National Emissions Standards for Hazardous Air Pollutants, Subpart H, National Emission Standards for Emissions of Radionuclides other than Radon from Department of Energy Facilities."* U.S. Code of Federal Regulations .

Areas of Concern at BNL, Upton, New York. A Reference Handbook. Brookhaven National Laboratory, Upton, NY. June 1998.

Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) of 1980, as amended, Public Law 96-510,94 Stat. 2767,42 USC 9601 et seq.

State Pollutant Discharge Elimination System (SPDES) Permit No. NY 000 5835. Issued by the New York State Department of Environmental Conservation, 2009.