



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Linking Legacies of the Cold War to Arrival of Anthropogenic Radionuclides in the Oceans through the 20th Century

T. F. Hamilton

December 12, 2003

Marine Radioactivity

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Chapter 2

Linking Legacies of the Cold War to Arrival of Anthropogenic Radionuclides in the Oceans through the 20th Century

**T. F. Hamilton, Contributing Author
Environmental Science Division**

**This article was submitted to
Elsevier Applied Science Publication**

August 2003

Approved for public release; further dissemination unlimited

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendations, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

This report has been reproduced
directly from the best available copy.

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information
P.O. Box 62, Oak Ridge, TN 37831
Prices available from (423) 576-8401
<http://apollo.osti.gov/bridge/>

Available to the public from the
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Rd., Springfield, VA 22161
<http://www.ntis.gov/>

OR

Lawrence Livermore National Laboratory
IBIS Print & Digital Media
<http://www-r.llnl.gov/ibis/print/print>
Livermore, 94550 CA, U.S.A.

Contents

Introduction	1
Discovery of Elements Beyond Uranium.....	6
Spent Nuclear Fuel Reprocessing and Nuclear Weapons Production	10
Nuclear Weapons Testing.....	19
Modes of Production of Radioactive Debris	20
Atmospheric Nuclear Weapons Testing.....	23
Dispersion and Deposition of Radioactive Debris.....	25
Sources of Anthropogenic Radioanuclides in the Oceans.....	30
Ocean Dumping of Nuclear Waste	36
Other Sources of Artificial Radioactivity in Regional Seas.....	39
Sources of Anthropogenic Radionuclides in the Arctic	42
The Ocean Water Column	49
Acknowledgments.....	53
References	53

List of Tables

Table 1. Current inventory of radioactive releases to the environment in the Western Siberian Basin and adjoining territories of the Russian Federation.....	9
Table 2. Atmospheric nuclear tests (1954–1980) by number, country, site, year, and fission yield (Data Source: UNSCEAR, 2000).....	24
Table 3. Production and global release of key radionuclides in atmospheric nuclear tests.	31
Table 4. Oceanic inventory of fission products and transuranium elements originating from globally dispersed debris, and local and regional deposits from atmospheric nuclear weapons test.....	34

List of Figures

Figure 1. Sites where atmospheric nuclear detonations have occurred.	2
Figure 2. Map of weapons production reactors and reprocessing facilities, nuclear test sites, and waste disposal sites within the western Siberian Basin and adjoining territories of the Russian Federation	12
Figure 3. Detonation of a high-energy nuclear device showing formatio of a fireball.....	20
Figure 4. Yield curves for fission of ^{235}U and ^{239}Pu	21
Figure 5. Annual partitioning yields from nuclear detonations and apportionment of debris in the atmosphere. Partitioning from equatorial sites such as Christmas Island and high-altitude	

tests on Johnson Island were assumed equally divided between the Northern and Southern Hemispheres. For tests conducted at temperate sites (30°–60°) releases were essentially averaged between the equatorial and polar atmospheres depending on the month of the year the nuclear test was conducted (Data source: UNSCEAR, 2000).	27
Figure 6. Schematic diagram of transfer mechanisms between atmospheric compartments (to right) and the measured latitudinal fallout deposit of ⁹⁰ Sr (through 1990, to left) (Monetti, 1996).....	29
Figure 7. Estimated global production, and the cumulative and annual deposition of ⁹⁰ Sr calculated from individual fission yields of tests and atmospheric model predictions (after UNSCEAR, 2000) compared with the measured fallout deposit of ⁹⁰ Sr through 1990 (Monetti, 1996).	30
Figure 8. Local and intermediate fallout following the detonation of a 15 Mt thermonuclear test “Bravo” on Bikini Atoll on 1 March of 1954 in the northern Marshall Islands.	32
Figure 9. Inventory of radioactive waste disposals at sea.	37
Figures 10a and 10b. Effluent discharges from the Sellafield reprocessing facility in the United Kingdom (1952–1992) to the Irish Sea: A. Beta/Gamma Emitters; B. Alpha Emitters (Data source: Gray et al., 1995).....	41
Figure 11. Activity concentration of ¹³⁷ Cs in surface waters of the Northeast Atlantic, Black Sea, and Arctic Ocean (1985–1995) in relation to reprocessing facilities (Data source: Dr. Kathy Crane, Office of Naval Research, U.S.A.).....	44
Figure 12. Activity concentrations of ²³⁹⁺²⁴⁰ Pu in surface waters of the Northeast Atlantic, Black Sea, and Arctic Ocean (1985–1995) in relation to reprocessing facilities (Data source: Dr. Kathy Crane, Office of Naval Research, U.S.A.)	45
Figure 13. Depth distribution of ⁹⁰ Sr, ¹³⁷ Cs, and ²³⁹⁺²⁴⁰ Pu in the ocean, Tuamotu Archipelago, French Polynesia (Data source: Hamilton et al., 1996).....	50
Figure 14. Temporal pattern in the activity concentration of ¹³⁷ Cs in French Polynesian surface waters (modified after Hamilton et al., 1996).....	51
Figure 15. Surface ocean response to global dispersion and deposition of ¹⁴ C in the ocean: a subtropical South Pacific high-resolution coral record from Rarotonga (21°, 159°W) (Data source: Guilderson, personal communication).....	52

LIST OF ACRONYMS

AMAP	Arctic Monitoring and Assessment Program
AEC	Atomic Energy Commission
CRESP	Coordinated Research and Environmental Surveillance Programme
DOE	Department of Energy
DU	Depleted Uranium
EURT	East Urals Radioactive Trace
ERDA	Energy Research and Development Administration
EARP	Enhanced Actinide Removal Plant
GEOSECS	Geochemical Ocean Section
HLW	High-Level Waste
HEU	Highly Enriched Uranium
IASAP	International Arctic Sea Assessment Project
IAEA	International Atomic Energy Agency
LDC	London Dumping Convention
LEU	Low Enriched Uranium
MED	Manhattan Engineer District or “Manhattan Project”
MPA	MAYAK Production Association
MRI	Meteorological Research Institute
NAS	National Academy of Sciences
NDRC	National Defense Research Committee
NTS	Nevada Test Site
NEA	Nuclear Energy Agency
OSRD	Office of Scientific Research and Development
PNE	Peaceful Nuclear Explosions
RCU	Radiocarbon Unit
REDOX	Phosphate Precipitation, Reduction, Oxidation
PUREX	Plutonium Uranium Extraction
TRU	Transuranic
TU	Tritium Units
TR	Tritium Ratio
SIXEP	Site Ion Exchange Effluent Plant
OECD	United Nations Organization for Economic Co-operation and Development
WIPP	Waste Isolation Pilot Plant

Introduction

The nuclear age has presented social, moral, political, and environmental challenges that have impacted on the security and prosperity of mankind and his planet. Significant quantities of man-made radionuclides have been released into the environment as a result of atmospheric weapons testing, nuclear weapons production activities, and nuclear power fuel-cycle operations. Other releases have resulted from accidents involving nuclear materials, intentional disposals, and the general use of radioactive materials in medicine, industry, research, and space exploration. Societal fear of radiation, growing concerns about disposal of nuclear waste, and public opposition to nuclear power have virtually ended the development of the nuclear energy industry as a viable alternative to the burning of fossil fuels. Yet, the uncontaminated natural environment, including the marine environment, is inherently radioactive.

Knowledge about specific sources and exposure pathways of environmental radioactivity provide a scientific basis for estimating health risks and developing appropriate safety standards to regulate releases and minimize exposures. For most individuals, exposure to ionizing radiation from natural sources far exceeds that delivered by the man-made sources in the environment. The same can be said of the global marine ecosystem. The oceans cover an area of about 3.6×10^8 km², or about three-quarters of the surface area of the earth, and constitute an enormous natural reservoir of radioactivity. Approximately 12,600 Bq m³ of naturally occurring radioactivity in seawater is due largely to the

presence of long-lived radionuclides of primordial origin: ⁴⁰K, ⁸⁷Rb, and radionuclides arising from the decay chains of ²³⁸U, ²³⁵U, and ²³²Th. The total activity of the natural marine radiation environment, including the upper few meters of deep-sea sediment, exceeds 5×10^7 PBq⁽¹⁾. It is also widely acknowledged that naturally occurring ²¹⁰Po and ²¹⁰Pb provide the major dose to marine organisms and, in turn, the dietary intake of ²¹⁰Po is a major contributor to the natural background dose for those population groups consuming large quantities of fish and shellfish.

Studies of the natural radiation environment provide us with a useful perspective for assessing the impacts of intentional or accidental releases of radionuclides into the environment. In addition to answering key questions about the total amount and potential dose contribution from man-made radionuclides entering the environment, researchers are able to predict the fate and transport of individual radionuclides from knowing the transport properties of naturally occurring stable and radioactive analogs.

The global dispersion and deposition of debris from atmospheric nuclear weapons is by far the largest source of artificial radioactivity released into the global environment. The location of declared sites where atmospheric nuclear detonations have taken place is shown in Figure 1. The levels and distributions of fallout radionuclides in the oceans are reasonably well described, although new information and data about

⁽¹⁾ 1 PBq = 1×10^{15} Bq

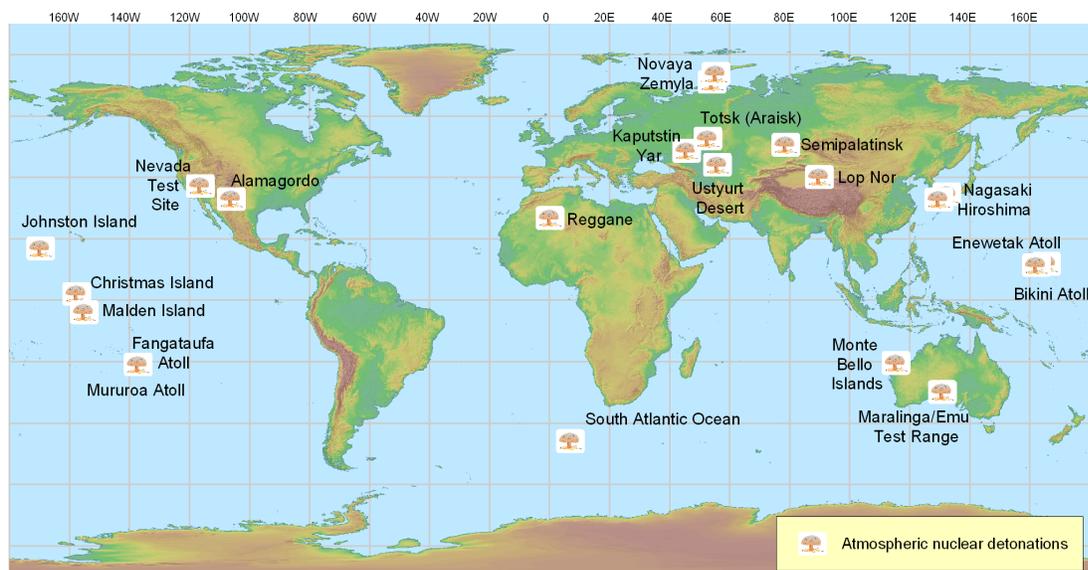


Figure 1. Sites where atmospheric nuclear detonations have occurred.

accidental and deliberate disposals at sea have raised concerns about the environmental consequence of such actions on a local and regional scale. Public concern about the potential sources, levels, and effects of radionuclide releases into the environment continue to grow as new information becomes available. Moreover, the guidance on radiation protection and standards associated with potential, low-level exposures compared to those delivered by the natural radiation environment are cause for scientific debate and controversy. The fact that a substantial quantity of artificial radioactivity has entered the environment over the past five decades without obvious detriment to the environment, or significantly increasing the measured collective dose to the world's population, is contrary to public perception.

International organizations such as the United Nations Committee on the Effects of Atomic Radiation continue to evaluate

global radiation sources and the effects of radiation exposure as a scientific basis for estimating radiation risk (UNSCEAR, 2000). However, the behavior and transport of radionuclides through the biosphere may be influenced by the type and nature of the source, the release conditions, radionuclide speciation, aging effects and/or other environmental factors. There is also a need to develop a better understanding of the impacts of radioactive contamination on biota to test the assumptions that existing radiation protection standards and controls for humans provide adequate protection for all living organisms. Consequently, efforts are ongoing to develop a better understanding of the behavior, fate and transport of man-made radionuclides in the environment, especially in relation to source-specific assessments and localized releases. For example, new data on the total number and yields of individual atmospheric nuclear weapons tests have only recently

become available. This information has been used to assess the partitioning of radioactive debris in the atmosphere, and to reevaluate deposition patterns and doses from individual fallout radionuclides. New information and data on nuclear waste disposal and handling operations on land and sea have also served to help quantify the relative risks and hazards posed by the legacy of waste management and weapons production activities. Database compilations are also being shared to promote the development of more accurate assessment tools.

The story behind the arrival of artificial radioactivity into the environment during the 20th century is an odyssey linked by technological development and sociopolitical change. From the turn of the century until the 1940s, the first measurements of radioactivity in the natural environment provided an early insight into radiation effects on humans. Published radiation protection standards for safe handling of xrays and radium already existed in the United States by World War II (NCRP, 1938; NCRP, 1936) although these regulations were never strictly enforced. Research on radiation biology, cancer treatments, and accidents involving ionizing radiation have demonstrated the need to control radiation exposure and radionuclide releases into the environment. Moreover, research on environmental radioactivity and radiation biology accelerated interest in studies on the potential health and ecological consequences of other pollutants in the environment. Today, studies on environmental radioactivity form an integral part of interdisciplinary research on climate change and carbon sequestration; atmospheric sciences; ocean dynamics and particle transport; archeology; and studies

on the origin, fate, and transport of nonradioactive pollutants in the oceans.

During the period between 1945 and 1963, environmental radioactive contamination studies were concerned primarily with the behavior of radioactive fallout from atmospheric nuclear weapons tests. The principal nuclear powers agreed to a test-ban treaty on atmospheric nuclear weapons testing in 1963, after which the annual deposition of radionuclides produced in atmospheric nuclear testing rapidly diminished. Secrecy surrounding nuclear weapons production processes and overriding public concerns about the rapid buildup of nuclear weapons arsenals tended to hide the emerging problems in nuclear waste management and environmental contamination. The production and stockpiling of nuclear materials and weapons required an extensive reprocessing effort that generated large volumes of radioactive waste. In the aftermath of the Cold War, the nuclear weapons states have begun the process of identifying legacy waste generated from nuclear weapons production activities, providing environmental restoration, and nuclear materials and facilities stabilization.

Public anxiety about nuclear technology has historically been linked to the consequences of nuclear weapons testing and the threat of nuclear war. The nuclear weapons testing era of the 1960s was followed by a period of great optimism about the potential peaceful uses of atomic energy. The civilian nuclear power industry had established a good safety record since its inception in 1956, and nuclear energy production was growing at an average annual rate of over 20% per year (1970–1986). Nuclear power production peaked in the United States during the late-1970s

because the industry was plagued by economic problems, lower demands for energy, and public and political controversy about reactor safety and radioactive emissions. Public opposition to the nuclear power industry was already on the rise in the aftermath of the 1979 reactor accident at Three Mile Island in Pennsylvania and came to the forefront of international attention following the catastrophic accident at the Chernobyl nuclear power plant in the Ukraine during May 1986. Chernobyl fallout was dispersed around the globe with measurable deposition occurring throughout the Northern Hemisphere. Worldwide public attention on environmental radioactivity shifted from the effects of nuclear weapons testing and the Cold War to the safety of nuclear power plants. Waste handling and disposal practices of the past were no longer tolerated by the public or regulatory agencies. A conservative policy in waste management added to the nuclear legacy with large quantities of nuclear waste—from both military and peaceful uses—being held in interim storage. Within the United States, growing concerns about nuclear energy including stockpiling of nuclear waste and associated environmental issues, caused parts of the weapons production complex to close and orders for new nuclear power plants to be canceled.

The Cold War ended with the United States and the Russian Federation agreeing on arms reductions but dismantling nuclear weapons, closing surplus production facilities, stabilizing waste, and environmental cleanup have only added to the nuclear waste stockpile. By the mid-1990s, the United States Department of Energy (DOE) managed over 3.6×10^7 m³ of solid and liquid waste containing about 37,400 PBq of radioactivity (DOE/EM,

1997). Most of the high-level waste from chemical processing of spent nuclear fuel and irradiated target assemblies is being held in interim storage at the four sites where it was originally generated: Hanford, the Idaho National Engineering Laboratory, the Savannah River Site, and the West Valley Demonstration Project (DOE/EM, 1997). The waste stockpile has been categorized into high-level waste (~35,500 PBq) composed of transuranic elements and fission products in concentrations that require permanent isolation, transuranic (TRU) waste (~140 PBq) resulting almost exclusively from weapons production processes, and low-level waste (~1850 PBq) characterized as all other radioactive waste not classified as high-level waste, TRU waste, spent fuel, or natural uranium and thorium byproduct material. Prior to 1970, portions of all waste categories were disposed of as effluents from nuclear facilities in shallow burial trenches, by sea burial, or by deep underground injection. These practices have since been discontinued. TRU waste is segregated and placed in retrievable storage above- or below-ground, typically in metal drums on soil-covered storage pads. More than 300,000 barrels of such waste are either buried or in temporary storage throughout the United States. The barrels await permanent disposal at the Waste Isolation Pilot Plant (WIPP), a planned geologic repository near Carlsbad, New Mexico. The DOE is also responsible for managing materials that are either not required for immediate use or no longer meet the current mission of the Department (DOE/EM, 1997). Materials in inventory include natural uranium; highly enriched uranium (HEU); low enriched uranium (LEU); depleted uranium (DU), plutonium, and other nuclear

materials used for nuclear research and weapons production; spent nuclear fuel; and lithium and lithium compounds used in the manufacture of nuclear weapons and production of tritium. About half the total of 8.2×10^8 kg of legacy materials held in inventory up until the mid-1990s has resulted from nuclear weapons production activities. Depleted uranium comprises 71% of the mass of radioactive materials in inventory, the majority of which is maintained at the three gaseous diffusion plants in Paducah, Kentucky; Portsmouth, Ohio; and Oak Ridge, Tennessee (DOE/EM, 1997). As of 1996, the DOE had identified approximately 5000 of its 20,000 facilities as surplus and accumulated approximately 1.9×10^9 m³ of contaminated environmental media as waste from site cleanup and facilities stabilization programs.

The United States has begun the process of addressing the environmental legacy of over five decades of nuclear weapons production. The end of the arms race appears to have become more problematic for the Russia Federation. Large quantities of nuclear waste built up from nuclear military operations of the Former Soviet Union have already been abandoned. Military shipyards in the Russian Arctic region are littered with idle nuclear-powered submarines and containers of spent nuclear fuel extracted during refueling operations. New information and data have recently confirmed that the Former Soviet Union dumped high-level nuclear waste, including reactor assemblies, some with spent nuclear fuel in the Russian Arctic Seas (1964–1992) (IAEA, 1999). Concerns about the potential impacts of nuclear waste dumping in the Arctic led to the development of the Arctic Monitoring and Assessment Program (AMAP) and renewed efforts by the

International Atomic Energy Agency (IAEA) to gather, compile, and evaluate information and data on source terms and radionuclide inventories. Scientific expeditions to the region during the 1990s indicate that elevated levels of radioactive contamination are confined to the immediate vicinity of waste containers and dumped objects. General levels of radioactive contamination in regional waters are said to be lower than those observed in the 1970s. These findings suggest that previous waste disposal operations at sea have had a negligible radiological impact on the environment. Nonetheless, the continued practice of sea disposal of radioactive waste has been widely condemned by the international community.

This chapter is intended to summarize the sources and occurrences of man-made radioactivity in the oceans, as well as provide a technical basis for addressing public misunderstandings about the risks posed by environmental radioactivity. The main contribution of dispersed radioactivity entering the environment has come from testing of atmospheric nuclear weapons from 1945 to 1980. The measured total global deposit of long-lived fission products such as ⁹⁰Sr and ¹³⁷Cs is in agreement with the estimated fission yields and partitioning of radioactive debris in the atmosphere. Other unrestrained sources of radioactive contamination of the marine environment include direct discharges of radioactive effluents into rivers and coastal seas from reprocessing and fuel cycle operations, and runoff or leakage from other land-based sources of contamination. Human activities involving nuclear materials will always be subject to accidents and accidental releases of radioactivity. However, the greatest challenge in controlling the man-made

radiation environment (i.e., limiting exposures to both humans and biota) and, to some extent, in maintaining international peace and nuclear security is seen as the management of legacy waste and nuclear materials in inventory. Today, the risk of illicit trafficking, terrorism, safety and facility vulnerabilities, and the temptation to use substandard waste containment or disposal options continue to grow with the increase in the global nuclear-waste burden. New challenges are also arising from the

growing accumulation of stored plutonium from both civilian and military operations. By the end of 1997, the plutonium in storage included 170 t of separated plutonium from civilian reprocessing operations, and another 100 t of excess plutonium from dismantled warheads no longer required for defense purposes (Oi, 1998). Moreover, the cumulative amount of plutonium in spent fuel from power reactors worldwide is predicted to exceed 1700 t by 2010.

Discovery of Elements Beyond Uranium

The early production history of man-made elements beyond uranium (element 92), and the discovery of their nuclear properties, is one of the most fascinating periods in science. Following the discovery of an uncharged primary particle—the neutron—by Sir Charles Chadwick in 1932, experimenters continued the study of the atom's nucleus by bombarding ordinary elements with neutrons, producing a range of radioactive isotopes one or two atomic numbers removed from that of the target nucleus. In 1934, Enrico Fermi and his coworkers at the University of Italy in Rome investigated the irradiation of uranium with slow neutrons and found that they produced radioactive species whose chemistry did not fit the properties of existing elements 90, 91, or 92. Fermi reasoned that he must have produced new elements 93 or 94. It was not until December of 1938 that two German scientists, Otto Hahn and Fritz Strassman, conclusively identified that one of the products of neutron capture by uranium was actually an isotope of a known element, element 56 (barium). They communicated their findings in the scientific journal *Nature*, and privately to former co-worker Lise

Meitner, who had escaped from Nazi Germany. Meitner met up with her nephew Otto R. Frisch in Copenhagen and together they worked out a detailed explanation of what Hahn and Strassman had observed: the breakup of the nucleus of a heavy atom into two lighter isotopes or fission products with a force unparalleled on an atomic scale. Enrico Fermi, working at Columbia University, was joined by Neils Bohr, a Danish physicist at Princeton University, and Ernest O. Lawrence at the University of California, Berkeley, to formulate a strong contingent of nuclear physicists and chemists working in the United States on nuclear fission. Before the theory behind nuclear fission could be published, two separate groups of experimenters at Columbia University, headed by Fermi and Leo Szilard, demonstrated that two or more neutrons were released in the fissioning of a uranium nucleus, and postulated that under the right conditions a chain reaction was possible. The military significance of producing a chain reaction was realized immediately. Scientists in the United States agreed to withhold information related to the military use of atomic energy from publication but F. Jolot

and co-workers in Paris reached the same conclusion and published their findings (Compton, 1956). By the time of the German invasion of Poland on September 1, 1939, more than 100 papers related to nuclear fission had appeared in the scientific literature along with evidence that uranium fission occurred by neutron capture on ^{235}U and not ^{238}U , the most abundant isotope of uranium. With this knowledge, nuclear physicists speculated that a chain reaction could potentially be sustained by using ^{235}U separated from natural uranium or by devising a technique to slow the passage of neutrons to enable their more selective absorption on ^{235}U .

The flow of information on nuclear physics research diminished with the outbreak of World War II. In the summer of 1939, Hungarian-born refugee physicists Leo Szilard, Eugene Wigner, and Edward Teller convinced Albert Einstein of the need to alert President Roosevelt about the potential dangers of Nazi Germany developing and using atomic weapons in a world conquest. Einstein outlined details of recent advances and the imposing dangers of harnessing atomic energy in a personal letter to the President. President Roosevelt immediately decided to establish a joint Army–Navy uranium committee to give government financial assistance to those engaged in uranium fission research. The work of the uranium committee got off to a slow start reporting that the military applications of atomic energy...must be regarded only as possibilities (Compton, 1956). At the same time, British and German scientists received substantial support aimed directly at atomic bomb production and were making significant advances in developing techniques for separation of fissionable ^{235}U . As events in Europe intensified, it was those scientists working outside of government at American

universities with private funding that helped lay the true foundation for advancing the early wartime effort in nuclear fission. Links between American universities, the National Academy of Sciences (NAS), and military services were further strengthened by the formation of a National Defense Research Committee (NDRC) in June of 1940 to oversee the work of the Government's Uranium Committee as well as other government projects. By the time the United States entered into World War II there was a broad representation of American science devoted to national service.

The first artificially produced elements beyond uranium (elements 93 and 94) were discovered at the University of California at Berkeley. Plutonium was synthesized by bombarding uranium with charged particles (deuterons) to produce ^{238}Np , which decayed by beta emission to ^{238}Pu . The isotope ^{239}Pu was isolated in the spring of 1941 and found to undergo slow neutron-induced fission (Kennedy et al., 1946). The discovery of an artificially produced fissile element brought new life to the military objective of nuclear energy. In January of 1942, President Roosevelt gave approval for the development of the atomic bomb. The exploratory stage came under the direction of what was known as the S-1 Committee organized under the Office of Scientific Research and Development (OSRD) and headed by Dr. Vannevar Bush at the Carnegie Institute in Washington. Research on various methods of ^{235}U separation and plutonium production proceeded on parallel fronts. Tracer experiments on the chemical and nuclear properties of plutonium continued through the early 1940s using cyclotron production facilities at the University of California's "Old Radiation Laboratory" at Berkeley and the wartime Metallurgical Laboratory at the University of Chicago. The

atomic bomb development program was re-assigned to a new district within the U.S. Army Corps of Engineers in June 1942 in what was known as the Manhattan Engineer District (MED) or “Manhattan Project” under the direction of General Leslie Groves. A detailed history of the Manhattan project and the events leading up to the development of the first atomic bomb can be found elsewhere (Gosling, 1994; Rhodes, 1986; Jones, 1985; Hewlett and Anderson Jr., 1962). Under the Atomic Energy Act of 1946, management of the United States nuclear programs was reassigned to the Atomic Energy Commission (AEC, 1946–1975) and, over the next 20 years, consolidated into a large, government-owned research and development complex. The AEC was abolished in 1975. Management of the United States nuclear weapons complex was transferred to the Energy Research and Development Administration (ERDA), and, later, to the United States Department of Energy (DOE) (1977–present).

In general, waste management practices used in the United States over the past two decades have come under increasing scrutiny. The United States has largely suspended nuclear weapons production activities since the early 1980s and begun to reduce the size of the weapons complex under a stockpile stewardship program. Growing public awareness and stringent agency controls related to the treatment, processing, storage, transport, and disposal of radioactive waste have increased the burden of nuclear waste management and environmental stewardship. Negligent nuclear waste management

practices used by the Former Soviet Union are also presenting serious technical, political, and economic problems within the Russian Federation. However, the collapse of the Soviet Union and end of the Cold War have ushered in a new era of cooperation between the United States and the Russian Federation in linking the legacies of nuclear weapons production processes and their environmental consequences. Considerable information has become available on the nuclear waste management and extent of environmental contamination within the Former Soviet Union (Bradley, 1997; OTA, 1995).

Responsibility for the Russian nuclear weapons complex and associated waste management operations falls largely under the Ministry of Atomic Energy (Minatom). This organization is said to be arguably the owner of the world's largest nuclear waste stockpile (Bradley, 1997). Estimates of releases to the environment through the end of 1996 exceed 6.3×10^4 PBq compared with less than 100 PBq in the United States (Table 1). Approximately 97% of the radioactive waste entering the environment has been disposed of by underground injection or discharged into surface waters. A major problem facing the Russian nuclear complex today is the lack of adequate facilities for the safe handling, treatment, and storage of nuclear waste. Large quantities of untreated waste and spent nuclear fuel assemblies are being placed in interim storage on sea and land. These circumstances are seen as dramatically increasing the risk of radiation accidents and uncontrolled releases to the environment.

Table 1. Current inventory of radioactive releases to the environment in the Western Siberian Basin and adjoining territories of the Russian Federation.

Site	Measured or estimated releases to the environment (PBq, 1×10^{15} Bq)
Production Reactor and Reprocessing Waste	
Tomsk-7 waste injection	37,000
Krasnoyarsk-26 waste injection	16,700
Mayak reservoirs, lakes, Techa River	4500
Tomsk-7 reservoirs	4800
Krasnoyarsk-26 reservoirs	>0.7
Mayak production reactor coolant water discharges	4.9
Tomsk-7 production reactor coolant water discharges	1.4
Krasnoyarsk-26 production reactors coolant water discharges	3.9
Mayak-1957 high-level waste storage tank explosion	1.6
Mayak-1967 air borne releases from Lake Karachai	0.02
<i>Subtotal</i>	63,000
Other Radioactive Releases to the Environment	
Chernobyl Accident	58
Uranium mill tailings	220
Dimitrovgard waste injection	3.3
To Irtysh River from weapons testing	3.3
Other nuclear power plant operations	1.3
<i>Subtotal</i>	290
<i>Grand Total</i>	63,290 PBq

Spent Nuclear Fuel Reprocessing and Nuclear Weapons Production

Enrico Fermi and his associates at the University of Chicago brought the first nuclear reactor (the Chicago Pile, CP-1) into operation on December 2, 1942. Reactor technology developed quickly during War World II and less than a year later a 3.8 thermal megawatt (MW_t) research reactor (Clinton Pile, X-10) began operation at a site in eastern Tennessee, now called Oak Ridge. The X-10 was used to test reactor operations and plutonium separation technologies, and by February of 1944 was producing several grams of plutonium per month. A test pile (the Hanford 305 Test Pile) constructed at Hanford, near Richland, Washington also served as a platform to develop and research materials for use in full-scale reactors. Three full-scale, single-pass plutonium production reactors were initially constructed at Hanford during 1944 to produce plutonium for the Manhattan Project. The Atomic Energy Commission (AEC) built eleven additional production reactors between 1948 and 1955, including five single-pass reactors and a new generation N Reactor at Hanford, and five heavy water moderated reactors (R, P, L, K and C) at the Savannah River Site near Aiken, South Carolina. By the early 1960s, demand for weapons-grade plutonium was being adequately met by the Savannah River Site reactors and the original single-pass Hanford reactors were closed down (1964–1971). The N reactor at Hanford was shut down in 1987. The P, L, K, and C reactors at the Savannah River Site were converted to use highly enriched uranium (HEU) fuel in 1968, and continued to produce tritium and other

radioisotopes for nuclear weapons programs into the late 1980s.

Low-level and transuranic waste management operations at nuclear weapons production and fabrication plants in the United States have historically included land and sea disposals, ground injection, and temporary storage in tanks, seepage basins, and ponds. The single-pass Hanford reactors used water from the Columbia River for cooling: the water passed through the reactors into retention ponds and, after a few hours, was released back into the river. The effluent stream contained induced radioactivity from neutron activation of naturally dissolved minerals, water treatment chemicals, and other entrained corrosive products. Uranium fuel element failures within the reactor added fission products and other fuel products to the effluent stream. Transport of Hanford radioactivity down the Columbia River to the Pacific Ocean possibly represented the first significant occurrence of contamination of the marine environment by man-made radioactivity. Plutonium production at the Savannah River Site differed by the fact the reactors used a closed-loop cooling system so that discharges under routine operations were considerably lower than at Hanford. However, significant quantities of radioactivity escaped the Savannah reactors from nonroutine occurrences such as reactor purges, heat exchanger leaks, and fuel element failures. The reactor effluents included activation, fission and fuel element products, as well as significant quantities of tritium. During the first year of operation of the Savannah reactors, coolant water and disassembly basin

effluents were released directly into local stream and creeks. Two artificial lakes, known as the PAR Pond and L Lake, later served as coolant water reservoirs to allow the reactor effluent to cool before final discharge. The lake sediments are now contaminated with ^{137}Cs and transuranic elements built up from historical discharges (DOE/EM, 1997). Other low-level liquid wastes generated from fuel storage and disassembly, cleaning and decontaminating reactor equipment, use of toxic water treatment chemicals such as hexavalent chromium, and oils and other fuel products were disposed of in Hanford-style cribs, ponds, or seepage basins. The seepage basins at the Savannah River Site were closed and backfilled during the early 1960s, and then replaced with seepage basins with containment. Until 1991, an estimated $1.3 \times 10^9 \text{ m}^3$ of waste water containing about 52 PBq of radioactivity was discharged into the ground at the Hanford Site alone (DOE/EM, 1997).

The bulk of the DOE high-level waste was generated when plutonium and/or uranium was chemically separated from spent nuclear fuel either by bismuth phosphate precipitation, reduction oxidation (REDOX), or plutonium uranium extraction (PUREX). Chemical process waste included cladding waste produced by removal of coatings from irradiated fuel assemblies. Miscellaneous low-level and transuranic wastes streams also were generated from plutonium concentration and finishing processes, waste volume-reduction operations, uranium solidification, laboratory analysis, spill cleanup, and other operations. High-level radioactive wastes are stored at four main sites in the United States including Hanford, the Savannah River Site, Idaho National Engineering Laboratory, and the West Valley Demonstration Project. Significant quantities of transuranic waste are

also stored or buried at the Rocky Flats Environmental Technology Site in Colorado and at the Los Alamos National Laboratory in New Mexico. Most high-level radioactive and hazardous wastes generated from chemical processing at Hanford are stored in underground tanks. These tanks contain alkaline liquid, salt cake, and sludge with an average activity concentration around 30 TBq m^{-3} . Spills and leaks associated with the tank operations and other waste management practices have released large quantities of radioactivity into the environment causing a flow of contaminated ground water toward the Columbia River. As of 1995, the Hanford Site managed $240,000 \text{ m}^3$ of high-level radioactive waste containing about 11,800 PBq. About 40% of the high-level waste stored at Hanford is contained in highly radioactive capsules. The capsules contained cesium and strontium salts segregated from high-level waste generated from the REDOX and PUREX plants. This was done to allow additional low-level waste to be discharged to the environment and to conserve available tank space. The Savannah River Site manages about $130,000 \text{ m}^3$ of high-level waste containing in excess of 19,500 PBq. The primary radionuclides in storage at both sites include ^{137}Cs , ^{90}Sr , ^{90}Y , $^{137\text{m}}\text{Ba}$, and ^{241}Pu . In 1996, defense waste reprocessing plants at Savannah River Site and West Valley Demonstration Project began producing vitrified forms of high-level waste. During the period between 1954 and 1988, hazardous and low-level radioactive wastes from chemical reprocessing at the Savannah River Site were reportedly discharged into seepage basins, and after evaporation, some waste discharged into local streams (DOE/EM, 1997). Generation of high-level waste has decreased significantly since the early 1990s when the DOE stopped

reprocessing of spent nuclear fuel. All high-level waste produced in acceptable forms for long-term storage will eventually be disposed of in a geologic repository.

The first plutonium production reactors in the Former Soviet Union began operation in June of 1948 at the MAYAK Production Association (MPA). Mayak is located about 70 km north of Cheryabinsk adjacent to the city of Ozersk (formerly Chelyabinsk-65) on the border of the West Siberian Basin

(Figure 2). The Mayak facility occupies an area of about 200 km² and borders an extensive system of rivers, lakes, and marshes. The flood plain connects to the Techa River system providing an important transport vector for passage of contaminated surface waters and runoff down the Ob River into the Kara Sea. Today, the MPA continues to operate two production reactors to produce radionuclides for military and civilian use, an

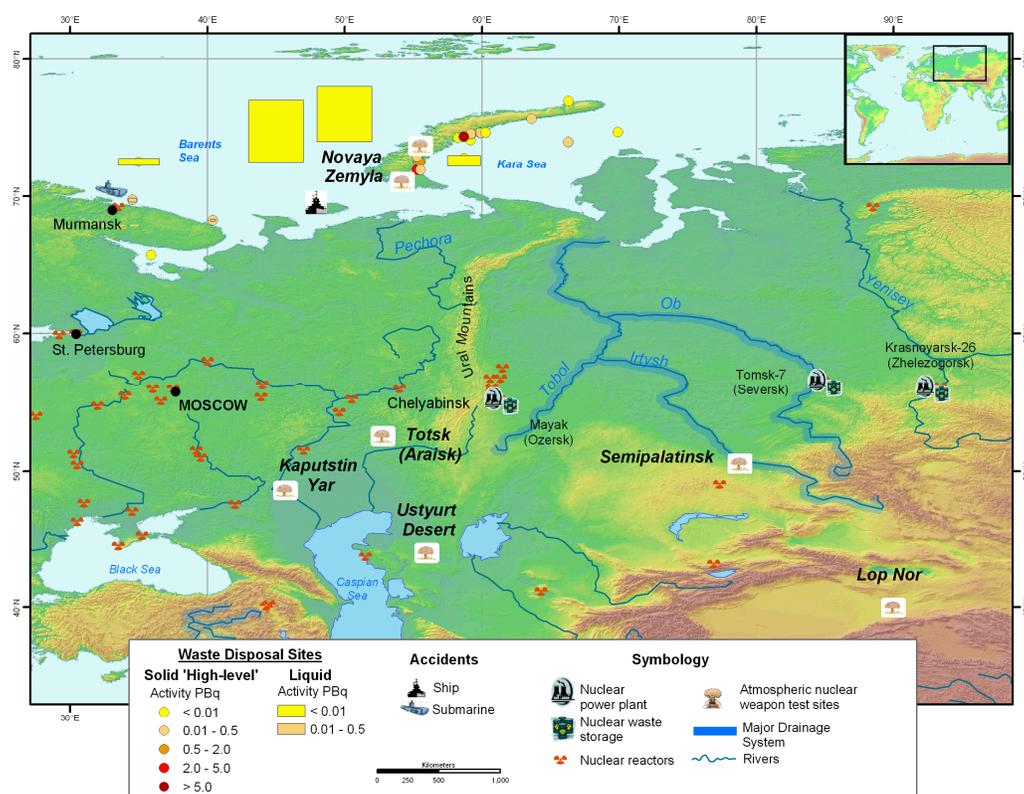


Figure 2. Map of weapons production reactors and reprocessing facilities, nuclear test sites, and waste disposal sites within the western Siberian Basin and adjoining territories of the Russian Federation

isotope production plant for commercial sale of isotopes and radiation sources, scientific research and manufacturing laboratories, as well as waste management and storage facilities. Other major Russian plutonium

production, reprocessing, and waste storage facilities include Krasnoyarsk-26 and Tomsk-7, all three sites being located within the Siberian Basin (Figure 2). Krasnoyarsk-26, now known as

Zheleznogorsk or the “Iron City,” is situated underground about 50 km north of Krasnoyarsk on the eastern bank of the Yenisey River. Tomsk-7 and the associated city of Seversk are situated on the Tom River about 25 km from the city of Tomsk.

The Former Soviet Union operated five graphite-modified, single-pass, water-cooled reactors at Mayak (1948–1990), two reactors at Krasnoyarsk-26 (1958–1992) and a single reactor at Tomsk-7 (1955–1990). Although no official data are available, cooling water passing through these “Hanford style” reactors was most likely contaminated with neutron-activation products formed from naturally occurring minerals and chemical additives in the coolant water or from entrainment of corrosion products from fuel cladding materials. Nonroutine releases of fission products, and fuel element failures and leaks, were also common in this type of reactor design. The reactor coolant water was released directly into nearby reservoirs, lakes, and rivers. Five additional graphite-moderated, production reactors with closed circuit cooling were brought into operation during the early 1960s: four of these reactors were located at Tomsk-7 and the other reactor at Krasnoyarsk-26. The original eight single-pass production reactors and two of the Tomsk-7 closed-circuit reactors were shut down between 1987 and 1992. Russia has continued to reprocess spent nuclear fuel from two remaining plutonium production reactors at Tomsk-7 and the single remaining reactor at Krasnoyarsk-26. Much of this material has been placed in storage for potential use in future energy and/or weapons production programs. The estimated total inventory of radionuclide releases from the Mayak production reactors to Lake Kyzyl-Tash on the Upper Techa River is estimated to be around 4.9 PBq (Table 1; Bradley, 1997). Associated releases from production reactors

at the Krasnoyarsk-26 and Tomsk-7 sites are 3.9 and 1.4 PBq to the Yenisey and Tom River, respectively (Table 1).

Beginning in 1948, the MPA commissioned their first radiochemical plant for separation of plutonium from spent nuclear fuel and a plant for conversion of plutonium to high purity metallic components for atomic bomb production. High-level chemical process waste was initially routed to storage tanks but tank storage was soon overwhelmed. A decontamination process was introduced to conserve tank space and allow a portion of the waste stream to be diverted into the Techa River. The technological waste-management process failed with large quantities of radioactivity being released into the Techa River (Degteva et al., 2000; Vorobiova et al., 1999). About 76 million m³ of liquid radioactive waste containing an estimated 100 PBq of radioactivity was reportedly discharged directly into the Techa River between 1949 and 1956. Ninety-five percent of the release occurred between March 1950 and November 1951 (Vorobiova et al., 1999; Degteva et al., 1994). The bulk of the radioactivity consisted of isotopes of ruthenium (¹⁰³Ru, ¹⁰⁶Ru) and the rare-earth elements along with an estimated 12 PBq of ⁹⁰Sr and 13 PBq of ¹³⁷Cs (Cochran et al., 1993). Approval was obtained to divert the bulk of the chemical process waste into Lake Karachai (reservoir 9) with lesser amounts (~4-7 TBq d⁻¹) entering the Techa River. Over the next 10–12 years a cascade of natural lakes, dams, and by-pass canals were constructed for the management of low-level and intermediate-level wastes in an effort to contain radioactive contamination to the Upper Techa River catchment and reduce radiation exposures to residents living downstream. The reservoirs acted as sedimentation ponds for adsorbed

radionuclides but the discharges were still sufficient to cause severe contamination of the entire floodplain. Systematic measurements of radioactive contamination of the Techa River began in 1951, including monitoring of river water, bottom sediments, flood-plain soils, vegetation, fish, milk and other foodstuffs, and external gamma-exposure rates. The practice of discharging chemical reprocessing waste into Lake Karachai was terminated towards the end of 1956. Construction on a second reprocessing plant (Complex BB) began in 1954 but did not commence operations until 1959. Complex BB was designed with improved radiation protection controls to reduce worker exposure and quickly reached its production goals enabling authorities to close down the original plant (Bradley, 1997). Reprocessing of defense reactor fuel was discontinued at Mayak in 1987. The Mayak production reactors remained in operation until 1990 with spent nuclear fuel shipped to Tomsk-7 for reprocessing. A total of 123,000 to 136,000 t of defense reactor fuel was reprocessed at Mayak between 1949 and 1987 (Bradley, 1997). Modernization and expansion of the site continued through the 1970s. Beginning in 1972, the original reprocessing plant (Complex B) was upgraded to receive, store, and reprocess spent fuel from different types of reactors. An additional 3400 t of spent nuclear fuel has since been processed in this facility, known as the RT-1 plant. The fuel has come from Russian-designed VVER-440, BN-350, and BN-550 power reactors; breeder reactors; research reactors; and nuclear-powered submarines and icebreakers (Bradley, 1997). The total present-day inventory of liquid radioactive waste released into the environment at Mayak (1949–1995), consisting mostly of ^{137}Cs and ^{90}Sr , is estimated at 4500 PBq (Table 1). By comparison, the total combined inventory of

radioactivity released into the near-surface environment at the Hanford (Washington) and Savannah River (Georgia) sites in the United States is around 60 PBq (Bradley et al., 1996).

Other secondary sources of radioactive contamination of the Upper Techa have included the explosion of a waste storage tank at Mayak in 1957 (the Kyshtym accident) and wind-borne releases from Lake Karachi. The Kyshtym explosion was caused by the failure of a cooling system allowing highly explosive nitrate salts inside a high-level waste storage tank to overheat (Nikipelov and Drozhko, 1990). An estimated 74 PBq of radioactivity was injected into the atmosphere and dispersed by wind to form the East Urals Radioactive Trace (EURT) (Botov, 1992). Deposition densities along the contamination track ranged from 5 PBq km^{-2} near the source to about 4 GBq at up to ~ 100 km distance (Nikipelov et al., 1990). At the time of the accident, the tank contained about 740 PBq of high-level liquid waste consisting mostly of short-lived fission products including ^{144}Ce (^{144}Pr), ^{95}Zr (^{95}Nb), ^{90}Sr (^{90}Y), and lesser amounts of ^{137}Cs (Drozhko et al. 1989). The present-day residual environmental inventory attributed to the Kyshtym accident is dominated by ^{90}Sr (^{90}Y) and is estimated to be ~ 1.6 PBq (Table 1).

Wind-borne releases from Lake Karachai were first detected in 1967 when a combination of meteorological conditions led to the resuspension of contaminated dust and silt from the exposed shoreline of the lake (Bol'shakov et al., 1991; Botov, 1992). Radioactive contamination was dispersed by wind up to distances of 50–75 km from the MPA site. Deposition from wind-blown sources of contamination have added about 15–370 GBq km^{-2} of radioactivity to regional soils, with some hot spots containing activity levels up to 1850 GBq km^{-2} (Cochran et al.,

1995). An estimated 22 TBq of radioactivity was associated with this contamination event (Table 1). Countermeasures introduced during the late 1960s to help reduce the spread of secondary contamination from Lake Karachai included covering the exposed shoreline with sand and improving lake embankments. Large quantities of soil and rock as well as hollow cement blocks have since been added to reduce the water holding capacity of the lake and stabilize muddy bottom deposits. Today, effluent discharges from waste management operations are still discharged into Lake Karachai to help maintain the water level and prevent further wind erosion of the shoreline.

As will be shown, the 4900 PBq of radioactivity released to surface waters of Lake Karachai and the Upper Techa River represent only a small fraction of the total inventory released to the environment at Tomsk-7 and Krasnoyarsk-26 by deep underground injection (Table 1). Discharges of radioactive liquid effluents from the Mayak facility have decreased from a maximum rate of ~0.16 PBq per day during the early 1950s (Degteva et al., 1994) to about 10 PBq per year during the 1990s (Christensen et al., 1995). As a result, the average annual concentrations of ^{90}Sr and ^{137}Cs in Techa River water collected near the village of Muslyumovo, located about 80 km downstream from the MPA site, have reportedly decreased from 1900 and 1.5 kBq L^{-1} , in 1951, to 0.15 and 0.015 kBq L^{-1} , in 1988, respectively (Jachmenev, 1995).

The construction of reservoir dams and canals to divert the Techa River from flowing into the reservoirs has helped regulate the outflow of water, silt, and associated secondary contamination into the Lower Techa River system. The closure of original defense reactor reprocessing plants, waste minimization efforts, increased availability of

tank storage, and use of improved reprocessing and waste management technologies have all helped reduce radioactive waste and associated discharges into the environment. Nonetheless, high levels of residual radioactive contamination within the Mayak region have made it necessary to establish a health protection zone covering an area of approximately 350 km^2 (Aarkrog et al., 2000). Both agricultural practices and permanent residences are forbidden within the region. A larger area has been designated as an observation zone where agriculture practices are permitted with environmental surveillance.

As of 1994, high-level waste derived from chemical reprocessing of spent nuclear fuel at Mayak has been vitrified into solid forms (Bradley, 1997), and steps have been taken to improve technologies for handling intermediate- and low-level liquid waste. Some releases to the environment are expected from storage (disposal) of legacy liquid and solid wastes in underground trenches and concrete repositories, but the inputs are probably insignificant when compared with previously declared releases to the environment. Through 1990, approximately 30,000 PBq of liquid and solid radioactive waste had been accumulated on the MPA site (Aarkrog et al., 2000)—about 75% of the radioactive waste inventory was classified as high-level waste (HLW). About one third of the HLW on site was vitrified into solid forms while the remainder was stored as nitric acid liquors or suspensions in stainless tanks or concrete tanks lined with stainless steel. By 1996, the MPA was generating about 16–20,000 m^3 of liquid intermediate-level waste (ILW) per year with a total radioactivity content of about 30 PBq. ILW and liquid low-level waste (LLW) from the facilities sewage treatment plant, and coolant water from the

reactor and waste management operations, continue to be discharged into the industrial reservoirs and lakes.

Radioactive contamination of the Techa River basin system remains a significant human health and ecological concern. Past activities at Mayak have severely contaminated the Techa River and associated floodplains that feed successively into the Islet, Tobol, and Ob rivers out to the Kara Sea: surface water runoff, overflow from the Mayak reservoirs and canals, and drainage from contaminated marshes near the former village of Assanov continue to provide a source-term for secondary contamination of the Techa River. Moreover, Russian scientists believe that infiltration of contaminated groundwater will ultimately become a major source of secondary contamination of rivers flowing into the Arctic Ocean. The reasoning is twofold. The main source of water supplying the Techa River during the summer months is groundwater seepage (Vorobiova et al., 1999). Raising the height of the Mayak dams to control the overflow of contaminated water into the Techa River has been offset by an increase in the rate of groundwater recharge. Secondly, the concentration of radioactivity in seepage water is increasing because the Mayak reservoirs contain highly mineralized water from discharges of chemical reprocessing waste; the natural sorption capacity of bottom deposits is practically exhausted leading to more rapid mobilization and migration of radioactive contaminants. The front of a southward-migrating mound of contaminated groundwater formed under Lake Karachai has already advanced 25 kms over the past 40 years and is approaching the Mishelyak River (Bradley et al., 1996).

Discharges of radioactive waste into Lake Karachai on the MPA site have captured

international attention but, still higher quantities of spent-fuel reprocessing waste was discharged into reservoirs and open pits at the Tomsk-7 nuclear materials production and reprocessing site. Recent estimates indicate that the Tomsk-7 reservoirs contain about 4,800 PBq of radioactivity (Moscow Interfax, 1994). Large quantities of liquid radioactive waste have regularly been discharged into Romashka and Tom Rivers including more than 17.5 PBq of radioactivity from a single-pass production reactor that operated from 1955 through 1990. The present inventory of radioactivity released from reactor operations (about 1.4 PBq) is dominated by activation products ^{63}Ni , ^{55}Fe , and ^{60}Co , formed from corrosion of aluminum fuel cladding and process tubes (Bradley and Jenquin, 1995). The reservoirs and basins are now mostly covered over with sand, concrete, or asphalt. Other waste management practices employed at Tomsk-7 have included storage of solid and liquid radioactive waste in tanks and special concrete repositories (buildings or underground tanks), but the predominant form of waste disposal is by far deep-well injection (Table 1). Well injection technology has been in use at Tomsk-7 since the early 1960s, and the practice continues today. Radioactive waste injected into the ground at Tomsk-7 accounts for nearly 90% of the total waste released or about 37,000 PBq. The waste is reportedly injected into sandy Cretaceous strata at depths of up to 450 m under very high pressure (Bradley, 1997). Russian authorities believe that radioactive waste injected into underground formations at Tomsk-7 (and Krasoyarsk-26) will remain isolated from the surrounding environment for the next 500 to 1,000 years allowing sufficient time for the bulk of the radioactive material to decay. Other scientists are concerned about contamination of the local water supply,

located at depths of only 20 m below ground, and possible long-term migration of radioactive contamination into the Tom and Ob River.

Other sources of radioactive contamination at Tomsk-7 include atmospheric emissions (e.g., ^{85}Kr , ^3H , ^{131}I , and alpha-emitting radionuclides) and incidents involving reactor or waste management operations. The most serious radiation accident occurred on April 6, 1993 when a waste storage tank exploded. The tank contained nearly 9 t of uranium in the form of partially processed uranyl nitrate solution, about 310 g of plutonium, tributyl phosphate (TBP), and paraffin (Nuclear Fuel, 1993; Nuclear Waste News, 1993). The blast released about 0.03 PBq of radioactivity composed mainly of short-lived beta- and gamma-emitting radionuclides such as ^{95}Nb , ^{106}Ru , and ^{95}Zr . Fortunately, wet snow began to fall soon after the blast limiting dispersion of radioactive aerosols to nearby forests and uninhabited regions up to a distance of about 8–15 km. Only the village of Georgievka and surrounding districts required some form of decontamination.

As with Tomsk-7, little quantitative information is available on waste management practices and environmental contamination at Krasnoyarsk-26. The Krasnoyarsk-26 complex known as the Mining and Chemical Combine was built underground to ensure survival from a nuclear strike. The first graphite-moderated plutonium production reactor at Krasnoyarsk-26 was commissioned in 1958. A second graphite reactor began operations in 1962 followed by the commissioning of a closed-circuit production reactor and spent-fuel reprocessing plant in 1964. The original single-pass defense reactors were decommissioned in 1992 (ITAR-Tass, 1992), while the closed circuit

reactor and reprocessing plant have apparently remained in operation. Construction of a separate reprocessing plant, known as RT-2, began operation in 1983 to reprocess VVER-1000 spent reactor fuel but prospects for its completion are uncertain (Bradley, 1997). As of January 1995, the spent-fuel storage facility within the RT-2 plant contained 1100 t of VVER-1000 fuel and had a holding capacity of 3000 t. Solid and liquid high-level radioactive wastes are also stored on site in underground tanks.

Sources of radioactive contamination of the Yenisey River from Krasnoyarsk-26 include discharges of coolant water from the original single-pass defense reactors (Tass World Service, 1992) and water overflow from open reservoirs contaminated with chemical reprocessing waste. The present inventory of radioactive waste released to the Yenisey River from reactor operations is estimated to be around 3.9 PBq (Table 1). The four main reservoirs at Krasnoyarsk-26 contain about 0.7 PBq of radioactivity but, unlike the reservoirs at Mayak and Tomsk-7, the activity composition of the residual radioactivity is dominated by plutonium and other long-lived radionuclides. Radioactive contamination of soils within the floodplain of the Yenisey River have been traced over distances of 1500 km, ranging from a maximum of 1.5 TBq km⁻² near the discharge point to less than 4 GBq km⁻² at 500 to 1500 km distance (Bradley, 1997). The principal radionuclides contained in bottom sediments include ^{51}Cr , ^{54}Mn , ^{60}Co , ^{90}Sr , ^{137}Cs , ^{238}Pu , and $^{239+240}\text{Pu}$. External exposure rates near the discharge point in the Yenisey River are known to have exceeded permissible standards of radiation protection. Local inhabitants continue to be exposed to elevated levels of ^{32}P , ^{24}Na , ^{65}Zn , and ^{60}Co by consuming fish

contaminated by discharges from the Krasnoyarsk-26 site.

Liquid wastes generated from spent fuel reprocessing have been injected into the ground at Krasnoyarsk-26 since 1963. The main injection site is located on a high terrace about 750 m from the Yenisey River. Radioactive (and industrial) wastes of different types are injected into a sandy clay deposit about 450 m thick—the deposit is further divided into beds of quartz-feldspar, sandstones, and kaolin/mica clays. The current inventory of radioactive waste injected underground at Krasnoyarsk-26 is estimated at 16,700 PBq (Table 1). The residual inventory of radioactive waste injected underground at this site is much greater than that discharged into surface water bodies. Although the geologic formation is favorable to radioactive waste disposal, a number of concerns have been raised concerning reported leaks in a 15-km-long pipeline used to transport radioactive waste from the main site to the well-injection installation. Other Russian scientists are skeptical about the impermeability of the geologic formation and concerned about possible long-term infiltration of contaminated ground water into local water supplies and the Yenisey River. Deep-well injection has also been used to dispose of radioactive waste at the Scientific Research Institute for Nuclear Reactors in Dimitrovgrad (Bradley et al., 1996) (Table 1).

Other potential land-based sources of radioactive contamination of surface and subsurface waters in the Siberian Basin include nuclear weapons testing, peaceful

nuclear explosions, power reactor operations, the 1986 Chernobyl accident in Ukraine, and disposal of ore tailings and liquid wastes from uranium mining and milling operations (Table 1). However, the total radioactivity released from all these events including Chernobyl is much less than 1% of the 63,000 PBq discharged directly into the environment from nuclear weapons production activities. The Western Siberian Artesian Basin is one of largest shallow water basins in the world. Surface water hydrology within the basin is dominated by an extensive system of rivers, lakes, and marshes that act as sinks for runoff and contaminant flow to the Arctic. The region is also believed to be a single groundwater basin with pervasive artesian character (Bradley, 1997). As a result, discharges of radioactive waste from the Mayak, Tomsk-7, and Krasnoyarsk-26 sites are potential long-term radioactive source terms to both the local and regional surface and subsurface hydrologic systems. Much has been done to improve waste management practices used by nuclear weapons complexes over the past five decades. The Russian Federation and the United States are also working together to evaluate the impacts of past (and present) releases including assessments on the long-term impacts on the Arctic region. Research is also continuing on quantifying the regional hydrology of the Western Siberian Basin and establishing boundary conditions to model contaminate flow and evaluate options for cleanup and remediation of contaminated sites.

Nuclear Weapons Testing

The full military potential of nuclear fission was first realized with the successful detonation of an “atomic bomb” in July 1945 over a New Mexico desert near the town of Alamogordo in the United States. The atomic bombings of Hiroshima and Nagasaki followed a few weeks later where the immediate loss of human life led to an end of World War II. Early nuclear weapons were pure fission devices fueled by either ^{239}Pu or ^{235}U . During the fission of a heavy element, the compound nucleus consisting of the original nuclei plus a neutron splits into two lighter elements, simultaneously emitting gamma rays, 200 to 220 MeV of fission energy, and two or three neutrons that propagate the fission of other target nuclei. The probability that fission will occur is defined by the effective critical mass of the fissionable material, the energy of the neutrons inducing fission, and the relative number of protons and neutrons of the target nucleus. To achieve rapid supercriticality of fissionable materials, a conventional explosive device was used either to bring two or more subcritical masses together to exceed the critical mass, or to compress a subcritical mass to become supercritical. Uranium-235 and ^{239}Pu contain an even number of protons and odd number of neutrons and are capable of undergoing fission with neutrons of virtually any energy. Nuclei having an even number of protons and neutrons (e.g., ^{240}Pu , ^{238}U , and ^{232}Th) can only be fissioned by high-energy (fast) neutrons above a threshold value of about 1 MeV—the higher the energy the greater the probability of fission.

The first thermonuclear or hydrogen bomb was detonated by the United States on

Enewetak Atoll in the Marshall Islands on 1 November 1952. A significant fraction of the energy released by thermonuclear devices comes from nuclear fusion where the high temperature generated by the primary fission reaction is used as a trigger to fuse deuterium and tritium with the concomitant release of neutrons and vast amounts of energy.

The explosive energy of a nuclear explosion is conventionally expressed in units of energy released by a ton (907 kg) of the explosive TNT (1 kiloton (kt) = 4.18×10^{12} J). The complete fission of one kilogram of ^{239}Pu containing about 2.5×10^{24} atoms will produce an explosive equivalent of 17.5 kt. The terms “dirty” and “clean” bombs have sometimes been used to describe the relative amounts of radioactivity produced by nuclear tests (Eisenbud and Gesell, 1997). Pure fission devices typically generate more radioactivity than weapons whose energy is primarily derived from maximizing the fusion yield.

A nuclear explosion produces a cloud of incandescent gas and vapor in excess of 10 to 100 million degrees called a fireball (Figure 3). The size and height of stabilization of the fireball are a function of the explosive yield of the device, the altitude of denotation, and the meteorological conditions at the time of the blast. The fireball of a 1-Mt explosion reaches a diameter of around 2000 after about 10 seconds, rising rapidly at initial speeds of several hundred kilometers per hour, and then slowing as it cools (Kathryn, 1984). As the fireball dissipates it assumes a toroidal shape where strong convective forces uplift cool air and surface debris into the cloud. The fireball from an atmospheric nuclear detonation spreads out into a typical mushroom shape as

it reaches the top of the troposphere (15-26 km) and may continue to rise well into the

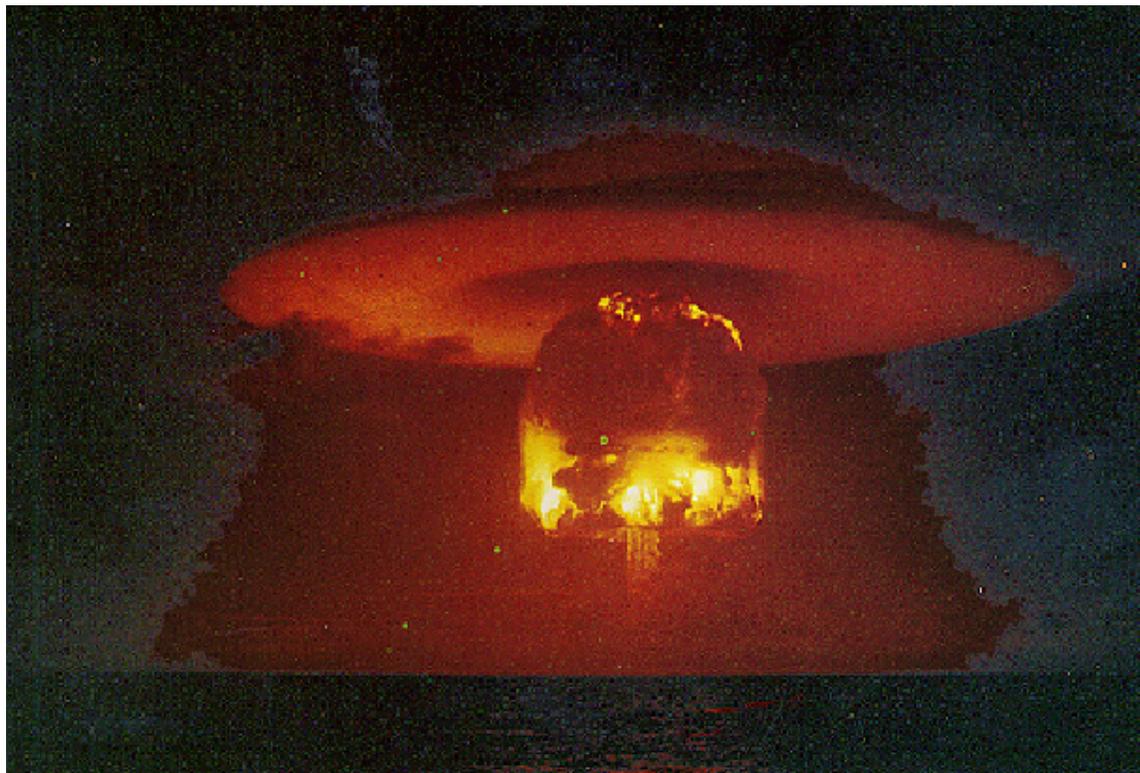


Figure 3. **Detonation of a high-energy nuclear device showing the formation of a fireball**

stratosphere. The cloud from a 1-Mt nuclear explosion reaches a maximum height about 10 minutes after denotation and may exceed 40-km in altitude (Kathryn, 1984).

Modes of Production of Radioactive Debris

Nuclear fission produces hundreds of short-lived, neutron-rich, nuclides of about 35 different elements with half-lives ranging from fractions of a second to 17 million years. Fission products normally decay by beta-emission through isobaric chains to longer-lived and finally stable nuclides. The detailed mass distribution of fission products depends on the target nucleus and the energy of the

fissioning neutron. The production probabilities or mass yields for ^{239}Pu and ^{235}U target nuclides are shown in Figure 4. The highest fission yields are associated with those isotopes with closed nuclear shells centering on mass numbers from 85 to 104 and from 130 to 139. The principal radionuclides of environmental significance produced from fission by slow and fast neutrons on ^{235}U , ^{239}Pu or ^{238}U include ^{79}Se , ^{85}Kr , ^{90}Sr (^{90}Y), ^{93}Zr , ^{95}Zr , ^{99}Tc , ^{103}Ru , ^{106}Ru (^{106}Rh), ^{106}Rb , ^{107}Pd , $^{113\text{m}}\text{Cd}$, $^{121\text{m}}\text{Sn}$, ^{126}Sn , ^{125}Sb ($^{125\text{m}}\text{Te}$), ^{129}I , ^{131}I , ^{134}Cs , ^{135}Cs , ^{137}Cs , ^{140}Ba , ^{141}Ce , ^{144}Ce , ^{147}Pm , and ^{151}Sm . The $^{137}\text{Cs}/^{90}\text{Sr}$ ratio in fallout can be used to provide a measure of the relative fission yields from target nuclides

within a mix of fuel components (IAEA, 1998).

The chemical and physical properties of particles formed in nuclear explosions (e.g., the size, distribution, shape, composition, and color) are known to vary according to the altitude of the denotation and composition of materials incorporated into the fireball (Crocker et al., 1966). The energy released from a large, near-surface denotation is sufficient to lift and immediately vaporize several hundred thousand t of soil and associated material. Vapors formed within the fireball begin to condense within seconds of denotation and under certain conditions fractionation between volatile and refractory materials may occur. Decay chain dynamics also play an important role in early condensation and fractionation of radioactive debris, especially for nuclides formed from short-lived noble gas precursors such as ^{137}Cs

and ^{90}Sr . Soil particles entering the fireball may also serve as nuclei on which radioactive debris or other condensation products attach. Refractory nuclides tend to be incorporated into larger particles (0.4–4mm) formed from condensation of iron, aluminum, and other refractory materials. Larger particles settle to earth quickly and produce what is known as local or close-in fallout deposition. The volatile elements tend to be associated with, or deposited onto, the surfaces of smaller-sized particles (<0.3mm in diameter) and are more likely to enter the global environment. It is assumed, on average, about 50% of the volatile fission products produced in near-surface detonations are deposited in the local or regional environment, while the remainder of the radioactive debris is widely dispersed into the global atmosphere (Peterson, 1970).

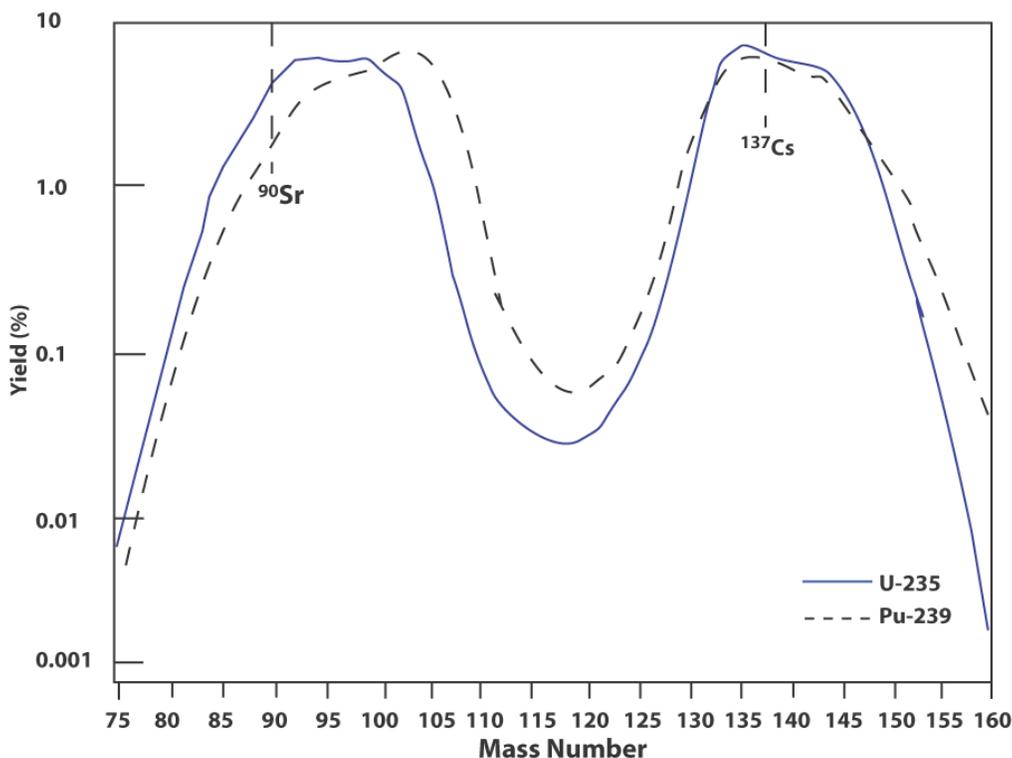


Figure 4. Yield curves for fission of ^{235}U and ^{239}Pu .

In addition to fission products, radioactive species are produced from neutron activation of non-fuel bomb and mounting materials, the atmosphere, and in soil and water within the immediate vicinity of the exploding device. The main activation products produced in atmospheric weapons tests of environmental significance include ^3H , ^{14}C , ^{36}Cl , ^{41}Ca , ^{45}Ca , ^{55}Fe , ^{59}Ni , ^{60}Co , ^{59}Ni , and ^{63}Ni . The residual gamma spectra of typical environmental materials collected near former atmospheric nuclear test sites are usually dominated by ^{137}Cs , ^{60}Co and the europium isotopes 152 and 155. Europium-152 and ^{154}Eu are considered products of neutron capture. Tritium is both a fuel residue and a fuel product of thermonuclear explosions. Radiocarbon (^{14}C) is produced by the interaction of neutrons on atmospheric nitrogen based on the $^{14}\text{N}(n, p)^{14}\text{C}$ reaction. The total residual tritium and radiocarbon released from atmospheric nuclear weapons testing has been estimated at 186,000 and 213 PBq, respectively (UNSCEAR, 2000). As will be shown, these reactions have produced a marked increase in the natural background concentration of tritium and radiocarbon in the surface ocean.

The main components of a nuclear weapon may include tritium, lithium deuteride, uranium or plutonium. Tritium is primarily used for boosting a fission device but may also be used in the secondary stage of thermonuclear weapons.

Tritium is both an intermediate product and fuel for thermonuclear denotations where it is produced through neutron reactions on lithium. Weapons grade uranium contains greater than 90% ^{235}U . Weapons grade plutonium contains greater than 93% ^{239}Pu and less than 7% ^{240}Pu . Some ^{241}Pu is normally present in a typical isotope mix of weapons grade plutonium. Plutonium-241 has a half-life of 14.35 years and decays to ^{241}Am ;

therefore, the amount of ^{241}Am present in weapons grade plutonium will depend on the age of the nuclear fuel.

Reactions of a nuclear explosion would ideally consume all the available nuclear fuel. Early nuclear weapons used a solid sphere or hollow shell (known as a pit) of ^{239}Pu or ^{235}U and consumed a relatively small fraction of the fuel. It was soon realized that introduction of deuterium-tritium gas could produce additional neutrons. These neutrons caused new fission chains and helped boost the overall yield of the device. Much higher yields were obtained by separating the fission trigger from the thermonuclear package in a Teller–Ulam “H bomb” configuration. The secondary stage of thermonuclear weapons produce very high neutron fluences with sufficient energy to fission and consecutive neutron capture reactions on uranium, in staged weapons. Natural uranium will capture a single neutron to form ^{239}Pu from decay of ^{239}U . The high-mass uranium isotopes are short-lived, and they decay by beta emission to longer-lived plutonium isotopes (e.g., ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu) over the course of a few days. The relative abundance of mass chains 239, 240, 241, and 242 in global fallout is 1 to 0.18 to 0.013 to 0.0043 (UNSCEAR, 2000; Krey et al., 1976). ^{237}Np may also be formed in thermonuclear detonations by $(n,2n)$ reactions on ^{238}U . Plutonium isotopes in the primary capture single neutrons to form ^{240}Pu , ^{241}Pu , and ^{242}Pu . A $(n,2n)$ reaction takes place when one neutron is absorbed and two neutrons are ejected; in this case, to form ^{237}U which then decays to ^{237}Np by beta emission. High-energy neutrons also produce ^{238}Pu and ^{234}U . Early weapons containing large quantities of uranium were known as ‘dirty bombs’ and produced a significant fraction of the total radioactive debris in global fallout.

Other radioactive species associated with atmospheric nuclear testing programs included activation products of neutron fluence monitors. A number of stable elements incorporated into critical components of nuclear devices were commonly used to determine the neutron energy spectrum and flux (and for other diagnostic purposes) by measuring single and multiple (n, 2n) products. Residual quantities of ^{235}U and ^{238}U remaining in the environment from nuclear weapons testing are often overlooked because of the high natural uranium content of soils, seawater, and other environmental materials.

Atmospheric Nuclear Weapons Testing

The main contribution to the global man-made radiation environment has come from the testing of nuclear weapons in the atmosphere. Updated listings of the date, type, total number, and explosive yields of individual nuclear tests as reported by country have recently been compiled by UNSCEAR (2000). There were reportedly 543 atmospheric nuclear denotations carried out between 1945 and 1980 with an estimated yield of 440 Mt. The annual number and total fission yields of atmospheric nuclear tests performed by all countries are summarized in Table 2, and the location of test sites shown in Figure 1. The number of reported detonations includes 39 safety trails used to assess the impacts of handling or operations accidents. Atmospheric nuclear tests include detonations performed as airdrops, suspensions from balloons, launchings by rockets, mountings on towers, placement on barges, and from anchorage points under water. Through the

end of 1998, an additional 1876 underground nuclear detonations have been reportedly carried out with a combined yield of 90 Mt (UNSCEAR, 2000). Cratering test are known to have released radioactive debris into the atmosphere but, in general, releases to the near-surface environment from underground explosions are only occasionally observed.

The first atomic bomb—code named Trinity—was detonated by the United States in July 1945 near the town of Alamogordo in New Mexico. Less than a month later, nuclear weapons were detonated under wartime conditions over the Japanese cities of Hiroshima and Nagasaki. The United States established a nuclear weapons testing program shortly after the end of World War II on Bikini Atoll in the northern Marshall Islands. Operation Crossroads commenced in June of 1946 and involved two nuclear tests: the ABLE shot (21 kt) detonated at an altitude of about 150 m and the BAKER shot (21 kt) detonated 30 m underwater inside the lagoon. Three more nuclear devices were detonated in 1948 on Enewetak Atoll before the Former Soviet Union tested their first nuclear device on 29 August of 1949. The United Kingdom followed with their first nuclear test during October of 1952. Atmospheric nuclear weapons testing continued at an accelerated rate from 1954 to 1958. A nuclear weapons-test moratorium was declared in the fall of 1958; by which time a total of 261 nuclear detonations had taken place having a cumulative explosive yield of 152 Mt. Virtually all these tests were carried out in the atmosphere with unrestrained release of

Table 2. Atmospheric nuclear tests (1954–1980) by number, country, site, year, and fission yield (Data source: UNSCEAR, 2000).

Region	Test Site	Country	Years	Number of	Yield (Mt)		
				Tests	Total	Fission	
Equatorial Pacific	Bikini Atoll	United States	1946-1858	23	76.8	42.2	
	Christmas Island	United Kingdom	1958	6	6.65	3.35	
	Christmas Island	United States	1962	24	23.3	12.1	
	Enewetak Atoll	United States	1947-1958	42	31.7	15.5	
	Johnson Atoll	United States	1958-1962	12	20.8	10.5	
	Pacific Ocean	United States	1955-1962	4	0.102	0.102	
			Total	111	159	84	
Northern temperate latitudes	Algeria	France	1960-1961	4	0.073	0.073	
	Japan ^a	United States	1945	2	0.036	0.036	
	Kapustin Yar	Former Soviet Union	1957-1962	10	0.98	0.68	
	Lop Nor	China	1964-1980	22	20.72	12.2	
	New Mexico	United States	1945	1	0.021	0.021	
	Nevada Test Site (NTS)	United States	1951-1962	86	1.05	1.05	
	Semipalatinsk	Former Soviet Union	1949-1962	116	6.59	3.74	
	Totsk, Aralsk	Former Soviet Union	1954-1956	2	0.04	0.04	
				Total	243	29.5	17.8
	Polar-north	Novaya Zemlya	Former Soviet Union	1955-1962	91	239.6	80.8
			Total	91	239.6	80.8	
Southern Hemisphere	Atlantic	United States	1958	3	0.0045	0.0045	
	Fangataufa Atoll	France	1966-1970	4	3.74	1.97	
	Malden Island	United Kingdom	1957	3	1.2	0.69	
	Maralinga/Emu Test Ranges	United Kingdom	1953-1957	9	0.080	0.08	
	Monte Bello Islands	United Kingdom	1952-1956	3	0.1	0.1	
	Mururoa Atoll	France	1966-1974	37	6.38	4.13	
				Total	59	11.5	7.0
Total	all sites	all countries		504#	440	189	

Includes 39 safety tests: 22 by the U.S.A., 12 by the United Kingdom, and 5 by France.

^a Two cases of military combat use.

radioactivity. Nine detonations had estimated explosive yields equal to or greater than 4 Mt representing more than 55% of the estimated total cumulative fission yield for all tests to 1958. These high-energy tests (≥ 4 Mt) were all detonated in the near-surface environment of Bikini and Enewetak Atolls in the northern Marshall Islands.

A nuclear weapons testing moratorium was observed through 1959 before France detonated its first nuclear weapon on 13 February of 1960 in Algeria. The Former Soviet Union resumed nuclear testing in September of 1961 prompting the United States to take a similar course of action in April of 1962. This second phase of atmospheric nuclear testing (1960–1962) involved 180 detonations having a total estimated explosive yield of 257 Mt. There were 118 atmospheric nuclear tests conducted in 1962 alone. There were also 15 atmospheric nuclear tests with total yields equal to or greater than 4 Mt and in contrast to high-energy tests detonated between 1954 and 1958, the majority of these tests were located in the polar atmosphere. The two exceptions were airdrops detonated over Johnson Atoll and Christmas Island in the equatorial mid-Pacific. China conducted the only additional high-energy nuclear test on 17 November of 1976 at the Lop Nor test site in Sinkiang Province.

The Former Soviet Union, the United States, and the United Kingdom agreed to an atmospheric test-ban treaty in 1963. France and China were not signatories to the treaty. China detonated its first nuclear weapon in October of 1964, and through 1980 conducted a total of 22 nuclear tests at the Lop Nor test site. In 1966, France moved its atmospheric nuclear testing program to the Tuamotu Islands, in French Polynesia, where they

detonated 4 tests on Fangataufa Atoll and 37 tests on Mururoa Atoll.

As will be shown, the injection and partitioning of radioactive debris in the atmosphere can be estimated from the location and yield of each test. There were 111 atmospheric nuclear tests conducted in the equatorial-north Pacific Ocean, 243 nuclear tests at northern temperate latitudes, 91 nuclear tests in the polar north, and 59 tests conducted in the Southern Hemisphere (Table 2). Therefore, the total fission energy released for partitioning of radioactive debris in the atmosphere is largely divided between the equatorial Pacific (43%) and the polar north (44%). Approximately 64% of the total fission energy released in the polar north was delivered by 13 high-energy detonations (≥ 4 Mt) between 1961 and 1962, and 64% of the total fission energy released in the equatorial Pacific was delivered by 11 high-energy (≥ 4 Mt) denotations, most of which took place between 1954 and 1958. Taken together, high-energy denotations account for 66% of the total yield and 56% of the fission yield of all atmospheric nuclear tests carried out between 1945 and 1980.

Dispersion and Deposition of Radioactive Debris

The nature and partitioning of radioactive fallout between the local environment, the troposphere, and the stratosphere are determined by (1) the type, location, and altitude of the test; (2) the energy yield; and (3) the quantity and type of materials interacting with the device (Hamilton et al., 1996). Radioactive debris deposited at or near test sites within a few hours from the time of detonation is described as local or close-in fallout deposition. The extent to which a given nuclear explosion will produce local or

regional fallout depends on the explosive yield of the test, its height above ground, and the physical characteristics of the particles formed. Airbursts are defined as those tests occurring in the atmosphere at or above a height of $55Y^{0.4}$ m (where Y is the total yield in kiloton) (Petersen, 1970). Below this height the fireball is expected to interact with the earth's surface and produce a significant amount of local and regional fallout contamination. The apportionment of radioactive debris in the atmosphere is coupled to the stabilization height of cloud formation following a nuclear explosion (Petersen, 1970) and can be estimated empirically from partitioning yield estimates. It is assumed that on average about 50% of the volatile radionuclides (e.g., ^{90}Sr , ^{137}Cs , and ^{131}I) produced in near-surface denotations entered the local or regional environment. The remainder of the debris from near-surface denotations and all the debris from airbursts entered the global environment (UNSCEAR, 2000) producing a worldwide pattern of global fallout deposition.

Updated annual partitioning yields for injection of radioactive debris into the various atmospheric regions were recently published by UNSCEAR (2000) and are shown graphically in Figure 5. The total partitioning yield contributing to worldwide dispersion of radioactive debris is estimated to be around 160.5 Mt compared with about 29 Mt deposited locally or regionally (UNSCEAR, 2000). The latter estimate is somewhat uncertain because of varying conditions between tests and the seasonality of atmospheric transport processes, but the uncertainty is small compared with the total fraction injected into the global atmosphere. Partitioning yields into the troposphere,

stratosphere, and high equatorial atmosphere were 15.6, 139 and 6.4 Mt, respectively. About 17 Mt was released into the Southern Hemisphere compared with 144 Mt in the Northern Hemisphere. Injection of nuclear debris into the northern equatorial stratosphere was most pronounced in 1954 followed by a significant pulse into the polar north stratosphere during 1961–1962. About 44.6 and 87.4 Mt of radioactive debris was injected into the northern equatorial and polar stratospheres, respectively, and together these two regions account for more than 82% of the total radioactive debris dispersed globally by atmospheric nuclear weapons testing.

Concern over the fate and transport of radioactive debris from nuclear weapons tests conducted in the late 1950s led to the establishment of a series of global monitoring networks. A surface-air monitoring program was initially established by the United States Naval Laboratory (1957–62) and continued by the DOE Environmental Measurements Laboratory in New York. Comprehensive measurement databases were developed for a number of fission products with particular emphasis on ^{90}Sr . Strontium-90, a beta emitter with a relatively long half-life ($T_{1/2} = 28.7$ years), is readily incorporated into the biosphere and shares chemical properties with calcium, an essential element for most organisms including humans. A primary concern over the fate of radioactivity released by nuclear weapons testing programs was assimilation of ^{90}Sr into marine and terrestrial foods and exposure to the human population. At the same time, measurements of radioactivity in air and deposition on the earth's surface served as internal tracers to study and model the dynamics of atmospheric transport processes.

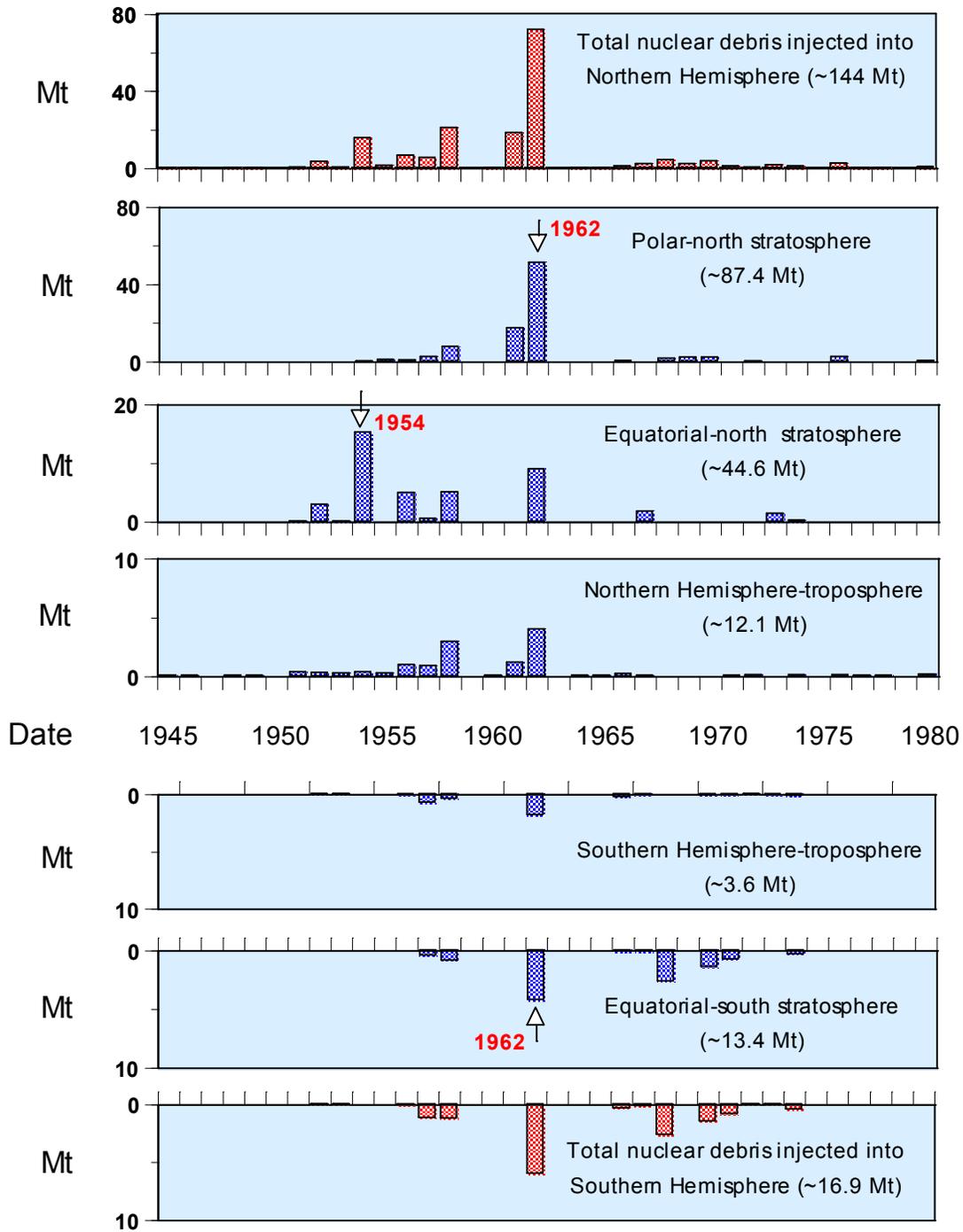


Figure 5. Annual partitioning yields from nuclear detonations and apportionment of debris in the atmosphere. Partitioning from equatorial sites such as Christmas Island and high-altitude tests on Johnson Island were assumed equally divided between the Northern and Southern Hemispheres. For tests conducted at temperate sites (30°–60°) releases were essentially averaged between the equatorial and polar atmospheres depending on the month of the year the nuclear test was conducted (Data source: UNSCEAR, 2000).

A schematic diagram of major atmospheric regions, predominant atmospheric transport processes, and the measured ^{90}Sr deposit averaged over 10-degree latitude bands is shown in Figure 6. Empirical models used to describe atmospheric dispersion and deposition of radioactive debris usually divide the atmosphere into an equatorial region (0° – 30°) and a polar region (30° – 90°). General air movement and atmospheric mixing processes control the dispersion of radioactive debris in the atmosphere. The top of the troposphere averages about 9 km in the polar region and 17 km in the equatorial region. The lower stratosphere extends to 17 km and 24 km within the two regions, respectively, and the upper stratosphere to about 50 km in both regions (UNSCEAR, 2000; Figure 6). Radioactive debris injected into the troposphere from low-yield detonations of 100 kT or less has a mean residence time of about 3 weeks. The most rapid removal of radioactive debris in the troposphere takes place during rainout events occurring locally or regionally within a few thousand kilometers from the test site. Radioactive particles injected into the stratosphere behave as aerosols and descend more slowly by gravitational settlement in the upper stratosphere and by eddy diffusion processes in the lower stratosphere. The mean residence time of radioactive debris injected into the upper stratosphere is around 24 months, and ranges from 3 to 12 months in the polar lower

stratosphere and from 8 to 24 months in the equatorial lower stratosphere (UNSCEAR, 2000). Air circulation in the lower stratosphere and troposphere at lower latitudes is driven by Hadley cell formation. Hadley cells tend to increase or decrease in size and shift in latitude with season (Newell, 1971). The transfer of radioactive debris from the lower stratosphere into the troposphere often occurs through gaps in the tropopause in the winter months and produces a characteristic increase in fallout deposition during the spring at mid-latitudes. The normalized production rate of ^{90}Sr in a nuclear detonation, assuming 1.45×10^{26} fissions per Mt, is around 3.88 PBq Mt^{-1} (UNSCEAR, 2000). The corresponding amount of ^{90}Sr produced in all atmospheric nuclear tests to 1980 is around 733 PBq. The estimated global release inferred from partitioning of radioactive debris in the atmosphere is about 623 PBq excluding releases associated with local and regional deposition. The latter value may be compared with a hemispheric ^{90}Sr deposit of 612 PBq using global fallout measurements (after UNSCEAR, 2000; Monetti, 1996). The partitioning yield estimate of 160.5 Mt provides better agreement with the measured deposition than previous fission yield estimates. Moreover, the measured results indicate that about 2% of the ^{90}Sr injected into the global atmosphere decayed before deposition and infers that the average residence time of radioactive debris in the atmosphere was ~ 1 year.

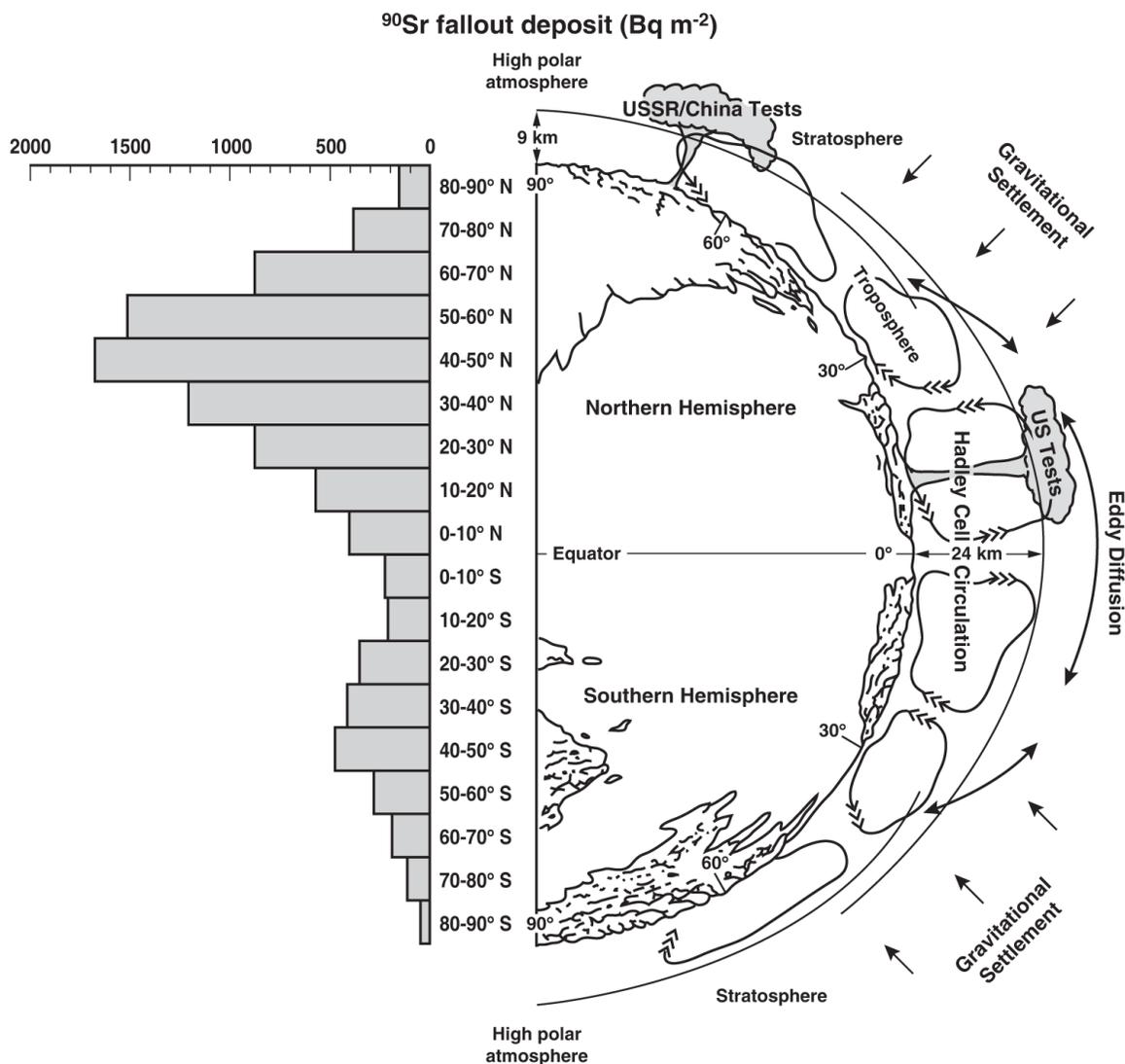


Figure 6. Schematic diagram of transfer mechanisms between atmospheric compartments (to right) and the measured latitudinal fallout deposit of ^{90}Sr (through 1990, to left) (Monetti, 1996; modified after Kathren, 1984).

The measured global cumulative deposit reached a maximum of around 460 PBq in 1967–1972 (UNSCEAR, 2000); since this time the cumulative deposit has decreased because radioactive decay of the global ^{90}Sr burden has been more rapid than inputs from the atmosphere (Figure 7). The calculated annual deposition of ^{90}Sr derived from model calculations shows close agreement with

measured values up until the early 1980s but becomes more uncertain with time (Figure 7). Deviations between the measured deposition and that calculated using empirical models can be attributed to the uncertainty of the measurements, resuspension of previously deposited material, and the influence of ^{90}Sr deposited during 1986 after the Chernobyl accident in the Ukraine.

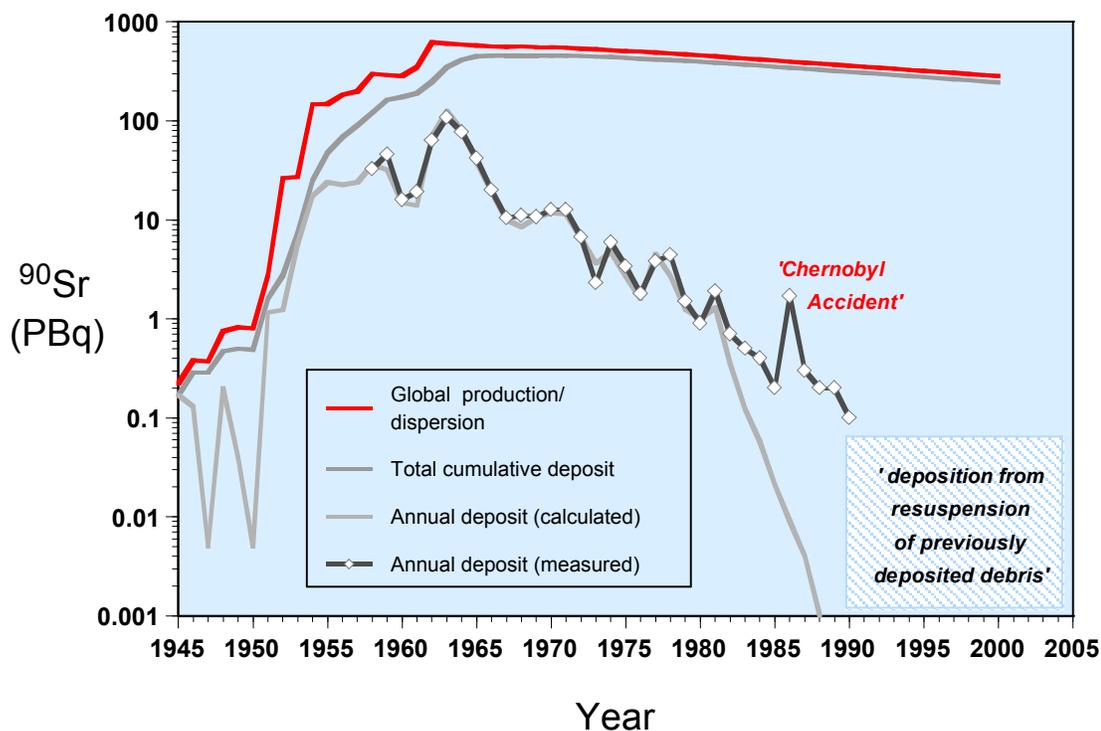


Figure 7. Estimated global production, and the cumulative and annual deposition of ^{90}Sr calculated from individual fission yields of tests and atmospheric model predictions (after UNSCEAR, 2000) compared with the measured fallout deposit of ^{90}Sr through 1990 (Monetti, 1996).

About 1.4 PBq of the ^{90}Sr deposit was associated with the Chernobyl accident. The measured global deposit of ^{90}Sr in 1990 was

around 311 PBq. By 2000, the ^{90}Sr deposit would have decayed to about 245 PBq.

Sources of Anthropogenic Radionuclides in the Oceans

Key radionuclides produced and dispersed globally by weapons testing, and their respective half-lives, fission yields, production modes, and global releases into the atmosphere are shown in Table 3 (updated after UNSCEAR, 2000).

Partitioning of fission yield estimates between the local, regional, and global environment vary from test to test, and on the fractionation of volatile and refractory elements. The global production and

dispersion estimates shown in Table 3 do not include debris injected into local and regional environments. As previously formulated, the measured cumulative deposit of volatile radionuclides in integrated fallout is consistent with global dispersion and deposition of all the debris from airbursts and 50% of the debris from near-surface detonations. For the volatile radionuclides, about 29 Mt of fission energy is assumed to be deposited locally and regionally; of which, about 28 Mt comes from

injections from near-surface detonations at test sites in the Pacific. Partitioning estimates for refractory radionuclides in near-surface detonations assume 50% of the debris is deposited within the immediate vicinity of the test site while an additional 25% is deposited regionally (Beck and Krey, 1983; Hicks, 1982). In general, the radionuclide composition of fallout debris produced in near-surface detonations can be adequately described as having all the volatile radionuclides and half of the refractory radionuclides present in unfractionated debris (Hicks, 1982). The corresponding partitioning

yield for dispersion of refractory radionuclides into the global environment is estimated at 130 Mt. The remainder of the debris (or equivalence of about 59 Mt of fission energy) was presumably deposited as local or regional fallout. The most significant localized releases were derived from high-energy, near-surface nuclear detonations on Bikini and Enewetak Atolls in the Northern Marshall Islands (Figure 8). Much of the local and regional deposition from these tests entered the marine environment forming a significant source-term, especially in the northwest Pacific Ocean.

Table 3. Production and global release of key radionuclides in atmospheric nuclear tests.

Nuclide	Half-Life (years)	Fission Yield (%)	Production Mode	Global Release into the Atmosphere ^a (excluding local fallout)	
				(PBq)	(kg)
³ H	12.33	—	fuel residue & fuel product	186,000	518
¹⁴ C	5730	—	(n, p) in device & environment; ¹⁴ N (n, p); ¹³ C(n,γ) (n,γ), (2, 2n) & (n, α) in device, and (n, γ) in the environment	213	1,290
⁵⁵ Fe	2.73	—		1,530	17
⁹⁰ Sr	28.78	3.5	fission product	623	122
⁹⁹ Tc	211,100	5.8	fission product	0.14	222
¹²⁵ Sb	2.76	0.4	fission product	741	20
¹²⁹ I	15,700,000	1.7	fission product; ¹²⁹ Xe(n,p)	0.0006	87
¹³⁷ Cs	30.07	5.6	fission product; ¹³⁷ Ba(n,p)	948	295
²³⁷ Np ^b	2,200,000	—	²³⁸ U(n,2n) ²³⁷ U, β ⁻ -decay	0.03	1,270
²³⁸ Pu ^b	87.7	—	fuel residue & fuel product; ²³⁹ Pu(n,2n)	0.28	0.18
²³⁹ Pu ^b	24,100	—	fuel residue & fuel product; ²³⁸ U(n,β ⁻) ²³⁹ Np, β ⁻ -decay	6.52	2,835
²⁴⁰ Pu ^b	6,500	—	fuel residue & fuel product; ²³⁸ U(2n, 2β ⁻)	4.35	512
²⁴¹ Pu ^b	14.4	—	fuel residue & fuel product; ²³⁸ U(3n, 2 β ⁻)	142	37
²⁴² Pu ^b	375,000	—	fuel residue & fuel product; ²³⁸ U(4n, 2 β ⁻)	0.002	11
²⁴¹ Am ^c	433	—	fuel residue & fuel product; β ⁻ -decay of ²⁴¹ Pu	(4.8)	(38)

^a Total global dispersion of radioactive debris resulting from 160.5 Mt of fission energy and 250.6 Mt of fusion energy (after UNSCEAR, 2000)
^b Estimated from isotopic ratios in integrated global fallout
^c Maximal deposit from decay of the global ²⁴¹Pu deposit (expected in 2032 at a level equivalent to ~44% of the present global ²³⁹⁺²⁴⁰Pu activity deposit).

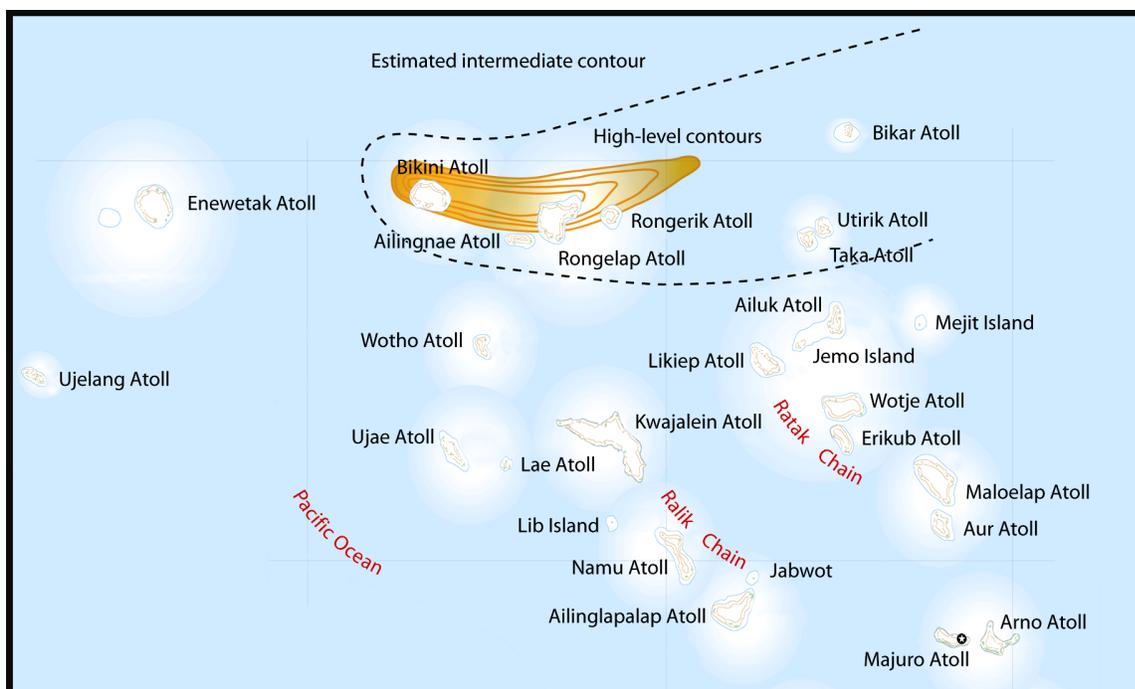


Figure 8. Local and intermediate fallout following the detonation of a 15 Mt thermonuclear test “Bravo” on Bikini Atoll on 1 March of 1954 in the northern Marshall Islands.

Sixty-six nuclear devices were detonated on Bikini and Enewetak Atolls. Near-surface blasts on towers, barges or underwater produced large quantities of partially or completely vaporized CaO , Ca(OH)_2 , and CaCO_3 , (Joseph et al., 1971; Adams et al., 1960). The physical/chemical characterization of local and regional fallout from the Pacific Proving Grounds is different to that found in globally dispersed debris. High-temperature vaporization and condensation processes produced different types and sizes of high-specific activity particles (Schell et al., 1980), which interacted with seawater (Adams et al., 1960). On hydration the particles swell forming a “crumbly or fluffy structure” (Adams et al., 1960); this is accompanied by release of hydroxyl ions and interaction with magnesium ions in seawater to form an inert shell of magnesium hydroxide on the particles (Buessler, 1997). These relatively insoluble particles deposited in the lagoon and continental shelf or slope sediments formed a reservoir and secondary source term to the equatorial Pacific. As of 1972, the $^{239+240}\text{Pu}$ inventory in lagoon slope sediments of Bikini and Enewetak Atoll was 54.4 TBq and 44.4 TBq, respectively (Noshkin and Wong, 1979). The corresponding annual export of $^{239+240}\text{Pu}$ to the open ocean was estimated at 0.12 TBq and 0.10 TBq, respectively. Similar studies at test sites in the South Pacific indicate that the annual export of $^{239+240}\text{Pu}$ from Mururoa Atoll lagoon to the open ocean is around 0.02 TBq (Bourlat et al., 1995). Radionuclide remobilization processes at these test sites are thought to be responsible for addition of up to 2 PBq of ^{137}Cs and 8–9 TBq of $^{239+240}\text{Pu}$ to the Pacific basin (Hamilton et al., 1996).

The total oceanic inventories of selected fission products and transuranic elements produced in atmospheric nuclear weapons tests are shown in Table 4. Radionuclide inventories were calculated by multiplying the measured latitudinal deposit in 10-degree bands with the corresponding fractional sea surface area across the world's oceans (taken from Baumgartner and Reichel, 1975). Marine contributions from nuclear weapons testing include 189 PBq of ^{90}Sr and 300 PBq of ^{137}Cs (Table 4). This compares with about 90 PBq of ^{90}Sr and 142 PBq of ^{137}Cs deposited globally in the terrestrial environment. Neglecting transfers between oceans, the Pacific contains about 60% of the total oceanic pool of volatile radionuclides and up to 70% of the refractory radionuclides (with the exception of ^{238}Pu at ~50%). Readers are reminded that 90% of the debris injected into the global atmosphere was deposited prior to the 1970s so present-day distributions of fallout radionuclides in the oceans will largely be controlled physical mixing and biogeochemical processes rather than a function of deposition patterns through the 1950s and 1960s (Hamilton et al., 1996 and references therein). The inventory estimates shown here for ^{90}Sr and ^{137}Cs are consistent with the results of the GEOSECS (Geochemical Ocean Section) expeditions with the exception of the Arctic Ocean. Discrepancies between the measured and predicted radionuclide inventories in the Arctic Ocean will be discussed in detail in the following sections. It is estimated that the Arctic, Atlantic, Indian, and Pacific Oceans contain about 2.0, 51, 22, and 114 PBq of ^{90}Sr , and about 3.2, 81, 31, and 182 PBq of ^{137}Cs , respectively. Other important long-lived fallout radionuclides of radiological or geochemical significance include the

transuranic elements along with ^{99}Tc and ^{129}I (Table 4).

The radionuclide inventories shown in Table 4 include estimated contributions from local and regional fallout as well as global dispersion of ^{238}Pu from the abortive reentry of a navigational satellite over the Indian Ocean. The SNAP-9A satellite was launched on April 21, 1964 but failed to reach orbital velocity and burned up in the high stratosphere. The SNAP-9A power unit contained about 0.63 PBq of ^{238}Pu and almost tripled the global deposit of ^{238}Pu from nuclear weapons testing (Hardy et al., 1973; Krey, 1967). Moreover, the SNAP-9A ^{238}Pu deposition distribution is an entirely different pattern compared with fallout from nuclear weapons tests. Approximately 80% of the SNAP-9A ^{238}Pu deposit was found in the Southern Hemisphere compared with about 20% from weapons fallout. In addition, about 1.7 PBq of ^{238}Pu and 1.2 TBq of ^{239}Pu entered the Tonga Trench in the North Pacific when an Apollo lunar probe was aborted in flight (Dobry, 1980). It is thought that the plutonium inside the lunar probe has remained intact in its containers at the bottom of the sea (Aarkrog, 1988). Localized inputs of plutonium into the marine environment also occurred in January of 1968 when an American B-52 aircraft carrying four nuclear weapons crashed on ice in Bylot Sound near Thule, Greenland. Plutonium contained in the weapons was released into the environment by a conventional chemical explosion and further distributed in the fuel fire following the crash. It has been estimated that about 1 TBq (or about 0.5 kg) of plutonium remained in bottom sediments of Bylot Sound after the initial cleanup operation (Smith et al., 1994; Aarkrog et al., 1987; Aarkrog et al., 1984).

Table 4. Oceanic inventory of fission products and transuranium elements originating from globally dispersed debris, and local and regional deposits from atmospheric nuclear weapons test.

Radionuclide	Arctic Ocean		Atlantic Ocean		Indian Ocean		Pacific Ocean		Total Oceanic Inventory	
	PBq	kg	PBq	kg	PBq	kg	PBq	kg	PBq	(kg)
⁹⁰ Sr	2.0	0.40	51	10	22	4	114 ^a	23	189	37
⁹⁹ Tc	0.001	1.8	0.03	46	0.01	20	0.07 ^b	110	0.11	178
¹²⁹ I	0.000005	0.72	0.00012	18	0.000050	7.7	0.0003 ^c	43	0.0005	69
¹³⁷ Cs	3.2	1.0	81	25	34	11	182 ^d	57	300	93
²³⁷ Np	0.0003	11.3	0.007	263	0.002	94	0.02 ^e	888	0.03	1,256
²³⁸ Pu	0.002 ^m	0.0032	0.13 ⁿ	0.2	0.11 ^o	0.17	0.50 ^{f,p}	0.78	0.73	1.16
²³⁹ Pu	0.054	23	1.4	591	0.58	250	4.5 ^g	2,960	6.5	2,820
²⁴⁰ Pu	0.036	4.2	0.90	106	0.38	45	4.0 ^h	477	5.4	632
²³⁹⁺²⁴⁰ Pu	0.090	28	2.3	697	0.96	295	8.6 ⁱ	2,436	12	3,456
²⁴¹ Pu	0.17	0.046	4.3	1.1	1.8	0.48	24 ^j	6.2	30	7.9
²⁴² Pu	0.00001	0.091	0.0003	2.3	0.0002	1.0	0.003 ^k	22	0.004	26
²⁴¹ Am	0.04	0.29	0.92	7.2	0.39	3.1	3.7 ^l	29	5.1	40

decay date 1 January 2000

^aIncludes an estimated 36 PBq of ⁹⁰Sr in local and regional fallout.

^bIncludes an estimated 0.025 PBq of ⁹⁹Tc in local and regional fallout.

^cIncludes an estimated 0.0001 PBq of ¹²⁹I in local and regional fallout.

^dIncludes an estimated 58 PBq of ¹³⁷Cs in local and regional fallout

^eIncludes an estimated 0.013 PBq of ²³⁷Np in local and regional fallout calculated from measured isotope ratios in global fallout deposition.

^fIncludes an estimated 0.11 PBq of ²³⁸Pu in local and regional fallout calculated from measured isotope ratios in global fallout deposition.

^gIncludes an estimated 2.4 PBq of ²³⁹Pu in local and regional fallout calculated from apportionment of radioactive debris in the atmosphere.

^hIncludes an estimated 2.7 PBq of ²⁴⁰Pu in local and regional fallout using isotopic ratios observed in fallout from the Mike detonation on Eneweak Atoll.

Includes an estimated 5.1 PBq of ²³⁹⁺²⁴⁰Pu in local and regional fallout using isotopic ratios observed in fallout from the Mike detonation on Eneweak Atoll.

^jIncludes an estimated 17 PBq of ²⁴¹Pu in local and regional fallout using isotopic ratios observed in fallout from the Mike detonation on Eneweak Atoll.

^kIncludes an estimated 0.003 PBq of ²⁴²Pu in local and regional fallout using isotopic ratios observed in fallout from the Mike detonation on Eneweak Atoll.

^lIncludes a maximal deposit of 2.3 PBq of ²⁴¹Am in local and regional fallout from decay of ²⁴¹Pu.

^mIncludes 0.0003 PBq of ²³⁸Pu to the Arctic Ocean from SNAP-9A burnup (the ²³⁸Pu deposit was distributed globally after Hardy et al., 1973).

ⁿIncludes 0.083 PBq of ²³⁸Pu to the Atlantic Ocean from SNAP-9A burnup (the ²³⁸Pu deposit was distributed globally after Hardy et al., 1973).

^oIncludes 0.090 PBq of ²³⁸Pu to the Indian Ocean from SNAP-9A burnup (the ²³⁸Pu deposit was distributed globally after Hardy et al., 1973).

^pIncludes 0.15 PBq of ²³⁸Pu to the Pacific Ocean from SNAP-9A burnup (the ²³⁸Pu deposit was distributed globally after Hardy et al., 1973)

For comparison, about 3.6 t of plutonium was dispersed into the oceans from nuclear weapons tests (Table 4). The Thule plutonium was initially present in the form of discrete oxide particles of low solubility and is not thought to constitute a significant source of plutonium contamination in the surrounding environment (AMAP, 1988). A similar incident involving the crash of a U.S. aircraft carrying nuclear weapons occurred near Palomares, Spain during 1966. In this instance, plutonium contamination was largely confined to agricultural lands surrounding the crash site. Impacts on the marine environment were apparently limited by the recovery of an intact nuclear weapon that fell into the Mediterranean Sea (unofficial source).

The world's oceans contain about 12 PBq of $^{239+240}\text{Pu}$ from global and local fallout deposition (Table 4). The Arctic, Atlantic, Indian, and Pacific Oceans contain about 0.1, 2.3, 1.0, and 8.6 PBq of $^{239+240}\text{Pu}$, respectively. These data predict that a significant fraction of the plutonium present in the Pacific Ocean comes from local and regional injections rather than global fallout deposition. Water column inventories in the North Pacific do reflect an excess of plutonium above what is expected from global fallout deposition (Bowen et al., 1980). For example, the $^{239+240}\text{Pu}$ inventory in the North Pacific water column during the 1973–74 GEOSECS expeditions was estimated to be around 6.4 PBq or about 50% more than that expected from global fallout alone. Recent observations of anomalously high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in the water column and dated coral records (Buesseler, 1997) provide strong evidence the main source of this excess plutonium is local fallout from the Pacific Proving Grounds in the Marshall Islands. The average $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in integrated global fallout is around 0.18 (Kjode et al.,

1985; Krey et al., 1976). Much higher ratios have been observed in debris from high-energy nuclear tests conducted in the Marshall Islands; for example, debris from the Mike thermonuclear test conducted on Enewetak Atoll in 1952 contained a $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio of ~ 0.36 (DOE, 1982). Plutonium isotopic abundances for the Mike test were used to predict the local and regional inputs to the Pacific shown in Table 4. A mixture of 4.6 PBq of global fallout with a $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio of ~ 0.18 combined with 5.1 PBq of plutonium distributed as Mike debris over the North Pacific would give rise to an elevated $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in the water column of around 0.24. Observed $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios reported in North Pacific seawater and sediment samples over a wide geographical area of the North Pacific range from ~ 0.19 to ~ 0.34 (Buesseler, 1997); the average ratio is ~ 0.23 in close agreement with the predicted value given above. The elevated levels of plutonium in the water column combined with available source-specific plutonium isotopic information appear to confirm earlier speculation that local fallout from nuclear weapons tests conducted in the Marshall Islands, typically characterized by an elevated $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios (>0.2), may form a major plutonium source-term in the North Pacific. Moreover, the trend toward finding higher $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in deep water samples and underlying sediments suggests plutonium geochemistry differs between sources of plutonium. Plutonium contained in local fallout is preferentially removed from the water column whereas global fallout plutonium behaves as a more soluble tracer. Source specific behaviors of Pu-bearing fallout particles can also be used to describe the relatively low $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios observed in some coastal sediments of the United States (Buesseler, 1986; Scott et al.,

1983) because of preferential removal of Nevada fallout containing an average $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio of 0.035 (Hicks and Barr, 1984).

Other important sources of artificial radioactivity in the marine environment include the dumping of nuclear waste; controlled effluent discharges associated with the nuclear fuel cycle and nuclear weapons production activities; accidental releases from land-based nuclear installations; and other accidents and/or losses at sea involving nuclear materials. Accurate estimates of the potential incremental or “pulse-like” releases of radionuclides contained in dumped nuclear waste or in nuclear materials lost at sea (e.g., nuclear reactors or intact nuclear weapons aboard sunken vessels; and other accidents involving nuclear weapons and/or sealed radioactive sources) are difficult to obtain and will only be reviewed here on a comparative basis (Figure 9).

Ocean Dumping of Nuclear Waste

Approximately 85 PBq of radioactive waste has been deliberately dumped into the oceans at more than 80 different locations worldwide (Figure 9). The first sea disposal operation took place in 1946 off the coast of California in the Northeast Pacific. Pursuant to principals adopted by an inter-government conference held in London in 1972, a convention was adopted for the Prevention of Marine Pollution by Dumping of Wastes and Other Matter (IMO, 1972). The Contracting Parties to the London Convention 1972 agreed to promote the effective control of all sources of pollution of the marine environment. The International Atomic Energy Agency (IAEA) further pledged to take practicable steps to prevent pollution of the sea by dumping of waste and other matter liable to (1) create

hazards to human health, (2) harm living resources and marine life, (3) damage amenities, or (4) interfere with other legitimate uses of the sea (IAEA, 1999). The convention was ratified on 30 August of 1975 and became known as the London Dumping Convention (LDC). The contracting parties to the convention designated the IAEA as the international organization responsible for matters related to sea disposal of radioactive waste. The IAEA was mandated to establish criteria for sea disposal of radioactive waste in order to minimize impacts on man and the environment, and provide recommendations for environmental assessment methodologies. Through 1977, sea disposal operations took place under national authority using consultative mechanisms developed through the IAEA and United Nations Organization for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA).

Between 1946 and 1982, five countries including Belgium, Japan, the Former Soviet Union, the United Kingdom, and the United States regularly used sea disposal as a nuclear waste management option. During this period, about 46 PBq of low-level nuclear waste was dumped at sea, mostly in the Northeast Atlantic dumpsite (Figure 9). The OECD/NEA maintained records of the waste disposal operations carried out by its Member States. Some indication of the composition and origin of the waste was usually given with beta-gamma emitters making up about 98% of the radionuclide inventory. The tritium inventory of radioactive waste disposals at sea to 1982 was around 15.6 PBq (IAEA, 1991) but represents less than 0.02% of the oceanic tritium deposit from atmospheric nuclear weapons testing (GESAMP, 1990). Other important radionuclides contained in

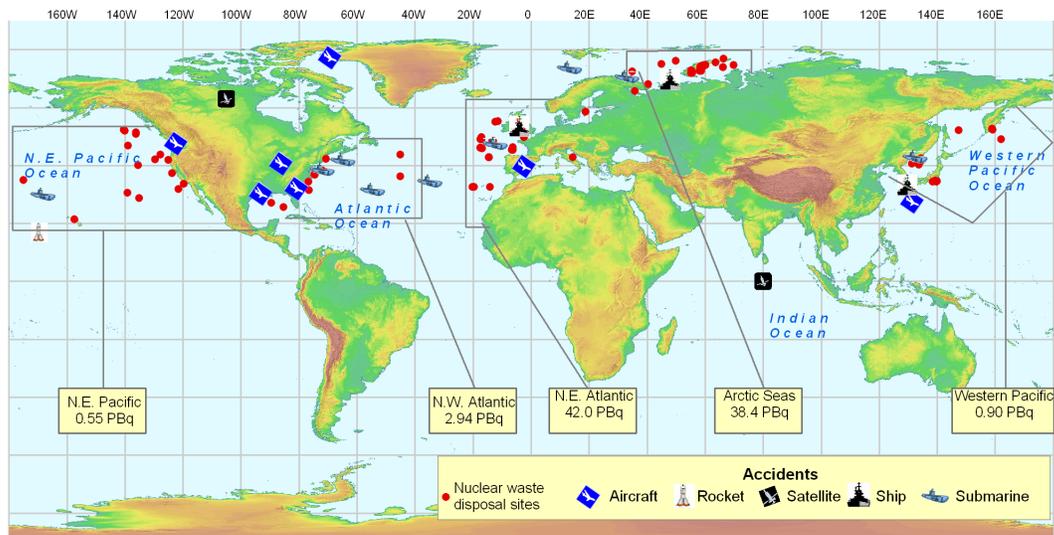


Figure 9. Inventory of radioactive waste disposals at sea.

radioactive waste dumped at sea include ^{90}Sr , ^{137}Cs , ^{55}Fe , ^{59}Co , ^{60}Co , and ^{14}C , and lesser quantities (<2%) of alpha-emitting radionuclides such as plutonium and americium. Radioactive waste dumped at sea consisted largely of low-level packaged waste from research, medical, industry, and military activities. The waste packages contained mostly protective clothing, glass, and contaminated concrete, piping, and other building materials encased in concrete or bitumen and/or placed in metal drums.

The OECD Council decided to develop a Coordinated Research and Environmental Surveillance Programme (CRESP) in 1977 as a key provision within a multilateral consultation and surveillance mechanism to keep sea disposal operations under review. The programme has conducted regular surveys of the Northeast Atlantic dumpsite since this time and the results have been published in reports (NEA/OECD, 1996). The IAEA Monaco Laboratory in cooperation with the Bundesforschungsanstalt für Fischerrei (BFA), Germany, and the Fisheries

Laboratory of the Ministry of Agriculture, Fisheries and Food, in the United Kingdom, conducted a major assessment of the Northeast Atlantic dumpsite in 1992. Analyses of seawater, sediments, and biota indicate a local source of radioactivity entering the environment but negligible radiological impact (Baxter et al., 1995). The U.S. Environmental Protection Agency and the U.S. National Oceanic and Atmospheric Administration have also carried out radiological surveys of the Pacific and Northwest Atlantic dumpsites. Again, the results of these surveys show that migration of radioactivity is limited to transfers to water and sediment in close proximity to waste packages residing on the sea floor.

A voluntary moratorium on disposal of low-level radioactive waste at sea was introduced in 1985 by the Contracting Parties to the London Convention-Resolution LDC.21(9); a new resolution was adopted in 1993 prohibiting sea disposal of radioactive waste. Then in May 1993, the Russian Federation provided IAEA with information

on previously undisclosed sea disposal operations adjacent to the territories of the Russian Federation in the Arctic and Far Eastern Seas. The Russian Federation published a report of historical waste disposal practices of the Former Soviet Union and the Federation through 1993 in what became commonly known as the “White Book” (White Book, 1993). Radioactive materials dumped in the Arctic Ocean have included liquid waste, solidified packaged and unpackaged solid waste from nuclear installations, and, perhaps most alarmingly, high-level radioactive waste including reactor and reactor compartments with and without spent nuclear fuel. Information gathering about the dumping operations, extensive surveys of the dumpsites and surrounding environment, and radiological impact assessments are continuing (AMAP 1998; IAEA, 1997; NRPA, 1996). According to information published by the Russian Federation, the total amount of radioactivity dumped in the Arctic Ocean was approximately 90 PBq at the time of disposal (White Book, 1993). The dumped objects included a total of 16 reactor assemblies or compartments, six nuclear reactor objects from submarines, and a reactor shielding assembly from the icebreaker Lenin, all containing spent nuclear fuel. Five of the reactor objects, including the reactor assembly from the icebreaker Lenin, were dumped in shallow fjords off the island of Novaya Zemlya between water depths of 20–50 m. The remaining reactor object with spent nuclear fuel was dumped in the Novaya Zemlya Trough at a depth of 300 m (Figure 2). All the dumped objects containing spent nuclear fuel were filled with a special polymer mixture, identified as FurfuroI™, prior to dumping. Deterioration of this protective barrier will eventually lead to the

corrosion of the materials housing the reactor and the nuclear fuel, releasing residual radioactivity into the environment. A detailed assessment of the source terms indicate that the radionuclide inventory in the dumped reactors is about 40% of the value given in the White Book (IAEA, 1997; Sivintsev, 1994a; Sivintsev, 1994b; Yefimov, 1994). The Northeast Atlantic and Arctic Ocean dumpsites combined received about 95% of the radionuclide inventory of waste dumped in the world's oceans. The remaining sea disposal operations occurred in the Northeast Pacific, Western Pacific, and Northwest Atlantic (Figure 9); a small quantity of radioactive waste (about 1 TBq) was also dumped off the east coast of New Zealand. Sea disposal of radioactive waste has been prohibited under a resolution adopted by the Contacting Parties to the London Convention since 1993 (IMO, 1993).

The last documented waste disposal operation at sea occurred in 1993 when the Russian Federation released low-level liquid radioactive waste into the Sea of Japan (Danilov-Danilyan, 1993). A series of Japanese–Korean–Russian expeditions to the region during 1994–95 indicated that the concentration of ^{90}Sr , ^{137}Cs , and plutonium isotopes in seawater and underlying sediments were within the range expected from global fallout from atmospheric nuclear weapons tests (Hirose et al., 1999; Pettersson et al., 1999). Approximately 98.6% of all radioactive waste disposal operations at sea were dumped in the form of packaged or unpackaged solid waste. The feasibility of using deep-ocean dumping was based on the premise that the integrity of the waste packages would remain intact, limiting the infiltration of seawater and subsequent remobilization of radionuclides and allowing the bulk of the radioactive materials to decay before entering the

environment. Radioactive decay of the waste has already reduced the oceanic inventory of radioactive waste dumped at sea to about 10 PBq or a factor of 10 times less compared with the time of disposal. Low-level liquid effluents make up only a small fraction of the waste stream (<1.4%) dumped in the oceans (IAEA, 1999) and are not expected to impact significantly on the marine radiation environment.

The International Arctic Sea Assessment Project (IASAP) was established in 1993, partly at the request of the Convention, but also to address deepening public concern about the potential health and ecological consequences of radioactive waste disposals in the Arctic region. Exploratory cruises to the Kara and Barents Seas were organized by a number of multidisciplinary teams of scientists, many under the auspices of the Arctic Monitoring and Assessment Program (AMAP). The activity concentrations of artificial radionuclides in the region are generally very low and, for the most part, can be explained by the deposition of global fallout or transfers of previously deposited debris. Consequently, the nuclear waste dumped in the Arctic Ocean appears to represent a negligible risk to human health and the environment. Nevertheless, elevated levels of selected radionuclides in the immediate vicinity of dumped objects were indicative of a gradual loss in waste containment (AMAP, 1998).

Other Sources of Artificial Radioactivity in Regional Seas

Discharges of radioactive effluents from nuclear fuel reprocessing facilities into coastal waters have resulted in a significant increase in the inventories of a number of radionuclides in the North Atlantic (Livingston et al., 1982).

Large quantities of radioactive waste have been discharged from the Sellafield nuclear complex (formally known as Windscale) on the west coast of Cumbria, in the United Kingdom. Reprocessing wastes have also been discharged into the English Channel from the La Hague reprocessing facility located on the northwest coast (Goury, Cherbourg Harbor) of France. The principal radionuclides and isotope ratios of radiological or oceanographic interest include ^3H , ^{90}Sr , ^{95}Zr , ^{95}Nb , ^{99}Tc , ^{106}Ru , ^{125}Sb , ^{129}I , ^{134}Cs , ^{137}Cs , ^{238}Pu , $^{239,240}\text{Pu}$, ^{241}Am , ^{241}Pu , $^{137}\text{Cs}/^{134}\text{Cs}$, $^{129}\text{I}/^{137}\text{Cs}$, $^{129}\text{I}/^{99}\text{Tc}$, $^{238}\text{Pu}/^{239+240}\text{Pu}$, $^{241}\text{Am}/^{239+240}\text{Pu}$, $^{237}\text{Np}/^{239}\text{Pu}$, and $^{241}\text{Pu}/^{239}\text{Pu}$.

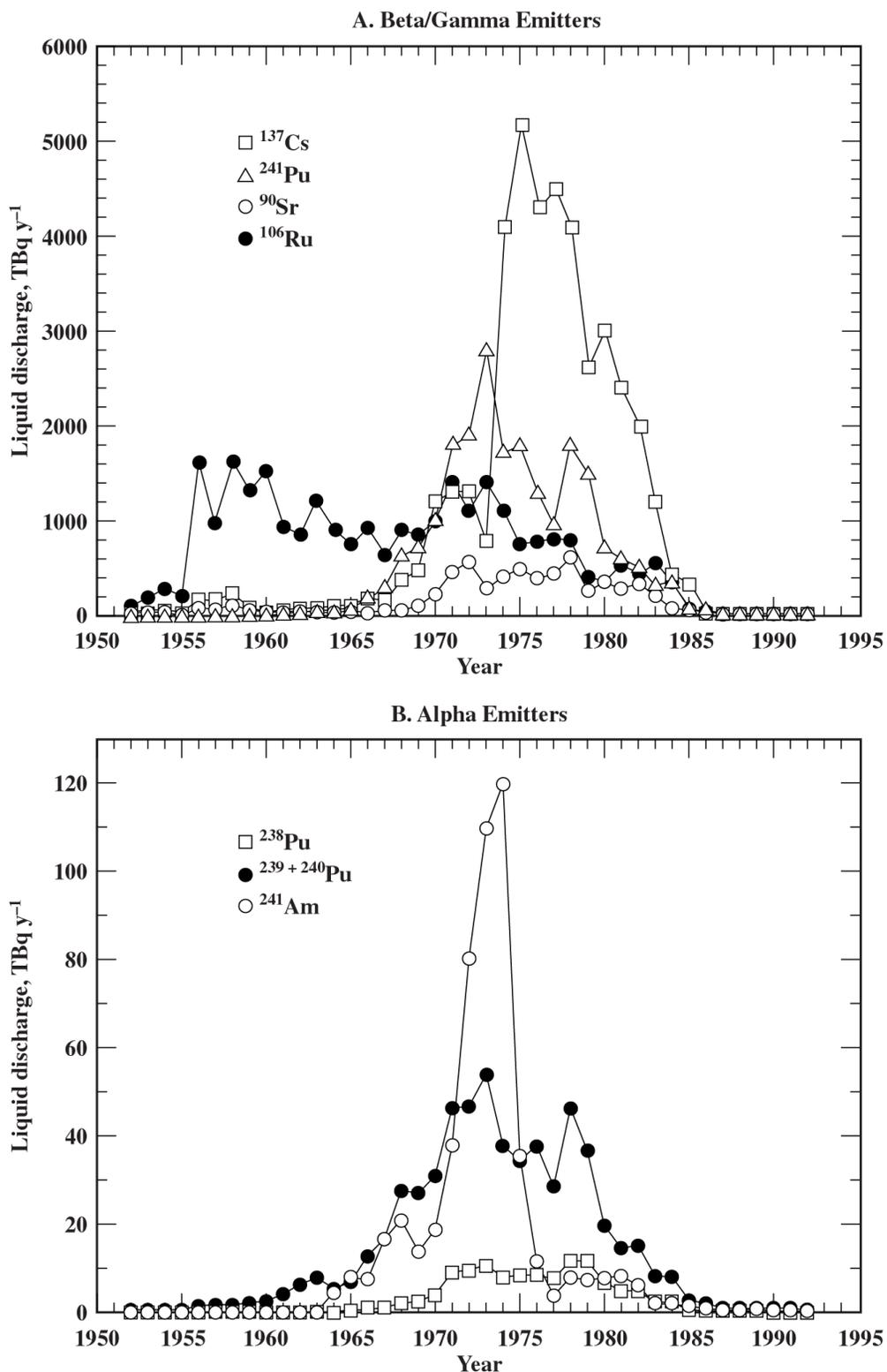
Two air-cooled plutonium reactors, known as the Windscale Piles, were first brought into operation during 1951. Irradiated fuel from reactors was stored in open-water filled ponds and subsequently processed in a primary separation plant. The plutonium separation plant and two associated purifications plants (B202, B203) were transferred to an integrated facility in 1964; the second purification plant (B203) remained in operation until February 1987. The Windscale Piles were closed down following a fire in Pile 1 during October of 1957. The unit was replaced with spent nuclear fuel shipped in from the Calder Hall and other Magnox reactor facilities.

The two main sources of liquid radioactive effluents at Sellafield are chemical process liquors and purge waters from spent fuel storage ponds. Fuel storage pond water was discharged into the Irish Sea without treatment through until the late 1970s. Radioactive waste disposals increased significantly over this period because of increased fuel storage time and corrosion of the Magnox fuel (Figure 10A). Historically, salt bearing liquors were an important source of ^{106}Ru that made up a significant fraction of the radionuclide inventory of the effluent stream (Gray et al.,

1995) (Figure 10A). Discharges of the transuranium elements also increased through the mid-1970s because of increased production levels and the addition of residues to the process stream (Figure 10B). Major developments and advances in waste management technologies have proved very successful in reducing Sellafield discharges. Beginning in the early 1970s, discharges of radionuclides were progressively reduced by introducing primary treatments systems (e.g., sand filtration, ion-exchange, and flocculation precipitation) and increasing the storage time of intermediate-level waste liquors prior to discharge. The disposal of intermediate-level waste liquors into the Irish Sea was terminated in 1980 (Gray et al., 1995). The significant reduction in radioactive waste disposals at sea after 1985 (Figures 10a and 10b) coincide with commissioning of the Site Ion Exchange Effluent Plant (SIXEP) and the Salt Evaporator. The SIXEP was used for treatment of storage pond waters. The Salt Evaporator allowed concentration and storage of chemical process liquors and other reprocessing effluents, reducing the discharge of short-lived fission products such as ^{106}Ru , ^{95}Zr , and ^{95}Nb (Gray et al., 1995). The Enhanced Actinide Removal Plant (EARP) was commissioned in 1994 to treat the backlog of stored concentrates. Many by-products arising from the treatment are now being converted into vitrified forms for long-term storage. Contemporary discharges from the La Hague plant in France have also been reduced since commencement of operations at the site in 1966 (Betis, 1993).

In general, radionuclide discharges from western European reprocessing facilities were likely to have had a pronounced affect on radionuclide inventories in the Arctic Ocean, especially during periods of peak discharge in

the mid-1970s (Kershaw and Baxter, 1995; Aarkrog et al., 1983). Discharges from La Hague and export of La Hague labeled seawater into the Norwegian and Barents Seas have largely been masked by Sellafield inputs, with the possible exception of ^{129}I and ^{125}Sb (Kershaw and Baxter, 1995; Raisbeck et al., 1993). The total contribution of La Hague to the marine inventory of artificial radionuclides discharged into the North Atlantic, expressed as a percentage of the Sellafield releases to 1992, is around 12.2%, 12.6%, 2.3%, and 0.4% for ^{90}Sr , ^{99}Tc , ^{137}Cs , and $^{238+239+240}\text{Pu}$, respectively (Kershaw and Baxter, 1995). Cumulative effluent discharges to sea from the Dounreay civilian reprocessing facility in Scotland are at least half as much again. Through 1992, fallout from nuclear weapons testing contributed about 79 PBq of ^{137}Cs and 1.8 PBq of $^{239+240}\text{Pu}$ to the North Atlantic. The comparative contributions from Sellafield are 41 PBq and 0.6 PBq, respectively. The Sellafield contribution inside the latitude band where the discharges occurred is obviously even more pronounced. For example, the inventories of ^{90}Sr , ^{99}Tc , ^{137}Cs , and $^{238+239+240}\text{Pu}$ discharged through 1992 exceed the estimated global fallout deposit in the 50° to 60°N latitude band by factors of 0.8, > 2000, 3.4, and 2.1, respectively. The spatial and temporal distributions of ^{90}Sr , ^{99}Tc , and ^{137}Cs in the North Atlantic over the past few decades have been described in many studies (Kershaw et al., 1999; Guegueniat et al., 1997; Kershaw and Baxter, 1995; Dahlgaard 1993; Kershaw et al., 1992; Dahlgaard et al., 1991; Smith et al., 1990; Kautsky, 1989; Dahlgaard et al., 1988; Dahlgaard et al., 1986; Pentreath, et al., 1985; Livingston et al., 1985; Aarkrog et al., 1983; Holm et al., 1983; Livingston et al., 1982; Baxter et al., 1979). The initial dispersion of



Figures 10a and 10b. Effluent discharges from the Sellafield reprocessing facility in the United Kingdom (1952–1992) to the Irish Sea: **A.** Beta/Gamma Emitters; **B.** Alpha Emitters (Data source: Gray et al., 1995).

radionuclides in effluent streams is controlled by a number of factors including the physical-chemical form of the element, local hydrographic conditions, and the composition of bottom sediments. Strontium-90, ^{99}Tc , ^{129}I , and the radiocesiums are relatively soluble in seawater and serve as conservative tracers for the passage of waters from western European reprocessing facilities to the Arctic (Guegueniat et al., 1997). Of interest, discharges of ^{99}Tc and ^{129}I from Sellafield have increased significantly through the mid-1990s reversing the progressive reductions seen for many other radionuclides (Kershaw et al., 1999). The pulsed release of ^{99}Tc provides a tracer to study and more accurately determine the transit times of North Atlantic water through the Norwegian Sea to the Arctic shelf seas and deep basins.

In contrast, reactive radionuclides released in particulate or hydrolyzed forms (e.g., plutonium and americium) are quickly removed to the underlying sediments by direct precipitation reactions or scavenging onto suspended particulate matter (Hetherington, 1975). About 0.6 PBq of $^{239+240}\text{Pu}$ was discharged into the Irish Sea from Sellafield through 1992 (Kershaw et al., 1995). Most of the plutonium was retained in a relatively defined coastal zone of the eastern Irish Sea bounded by muddy subtidal and intertidal sediments. It has only been in recent years, as discharges from Sellafield have declined, that the Irish Sea has been reconciled as a potential source of plutonium to the North Atlantic rather than a sink (Kershaw et al., 1995). Based on plutonium isotope ratio measurements, resuspended or resuspended plutonium leaving the Irish Sea can be distinguished from that contained in contemporary discharges and more closely reflects the cumulative Sellafield deposit found in sediments (Mitchell et al., 1999). The

solubilized plutonium from Sellafield is exported into the North Sea and possibly as far as the Barents and Greenland Seas (Kershaw and Baxter, 1995; Holm et al., 1986).

Sources of Anthropogenic Radionuclides in the Arctic

Over the past decade, a great deal of scientific and public attention has been given to investigations of radioactive contamination of the Arctic Ocean (Strand and Jolle, 1999; Strand and Cooke, 1995). It has been reported that the "Russian Arctic is filled with nuclear perils on sea and land, atomic waste litters the Murmansk Region" (International Herald Tribune, 1996). The Norwegian environmental organization, Bellona, has focused attention on Arctic contamination issues, especially the lack of adequate facilities for handling of liquid and solid radioactive wastes generated by naval and civilian operations on the Kola Peninsula. Tens of thousands of spent nuclear fuel assemblies from nuclear-powered submarines litter harbors and naval shipyards awaiting transfer to reprocessing facilities. In addition, a large quantity of liquid nuclear waste on the Kola Peninsula is held in temporary storage in tanks on sea and land, and aboard service ships and tenders (Nilsen, et al., 1996). The Russian Federation face a difficult task to secure, store, and treat existing legacy waste while continuing the process of decommissioning nuclear submarines and dismantling nuclear weapons under strategic arms reduction treaties with the United States. According to Bradley (1997), the Russian Federation decommissioned a total of 147 submarines and service ships through the beginning of 1995, including 76 submarines in the Northern Fleet. About half of the

decommissioned submarines remain in floating storage with the fuel on-board, and an additional 130 nuclear submarines remain to be decommissioned.

Extensive monitoring of radionuclides in seawater (and sediments) of the Barents, Kara and other regional seas suggests that current levels of radioactive contamination in the Arctic region remain relatively low and pose no immediate radiological concern.

Concentrations of ^{137}Cs and $^{239+240}\text{Pu}$ from surface activities in the Barents and Kara Seas between 1985 and 1996 are shown in Figures 11 & 12, respectively. Radionuclide distributions in the region can be adequately described by global fallout from nuclear weapons testing and the export of radionuclides from European reprocessing facilities in northwest Europe and the North Sea. Water flows northward into the Norwegian Sea via the Norwegian Coastal Current then splits off to the east into the Barents Sea or continues with the West Spitzbergen Current through the Fran Strait into the Nansen Basin. About 1 Bq m^{-3} of ^{137}Cs in surface water of the Barents and Kara Seas is derived from Chernobyl fallout (Dahlgaard et al., 1995) or as much as 10 to 20% of the total ^{137}Cs activity concentration. In all, about 100 PBq of ^{137}Cs was dispersed into the atmosphere from the 1986 Chernobyl nuclear reactor accident and deposited in the northern hemisphere, especially over the Ukraine, Belarus, and western Russia. The plume crossed central Norway and Sweden

with ^{137}Cs deposition exceeding 200 kBq m^{-2} (AMAP, 1998). Chernobyl fallout within the vicinity of the Arctic Circle was considerably less. For example, northern Finland received around $1\text{--}2 \text{ kBq m}^{-2}$ but impacts of Chernobyl on the burden of artificial radioactivity in the Arctic Basin is difficult to quantify because of the heterogeneous or patchy nature of the deposition. Furthermore, the ^{137}Cs content of the Baltic Sea increased about 10-fold after the Chernobyl accident from immediate deposition and surface-water runoff. Export of Chernobyl labeled waters from the Baltic Sea and North Sea through the Norwegian Sea constitutes another important pathway for transport of artificial radioactivity into the Arctic Ocean. The influence of marine transport processes on the export of global fallout in the North Atlantic from low latitudes to the Arctic has often been overlooked. The maximal global fallout deposit from atmospheric nuclear weapons testing occurred at mid-latitudes and decreased substantially toward the North Pole (Figure 6). Redistribution of just 5% of the North Atlantic global fallout deposit essentially doubles the contribution in the Arctic Ocean. This scenario seems entirely plausible based on our knowledge of marine transport processes developed from studies of Sellafield-labeled waters. Sea ice rafted sediments may also play a role in the long-range transport and biogeochemical cycling of artificial radionuclides in the region (Cooper et al., 1998).

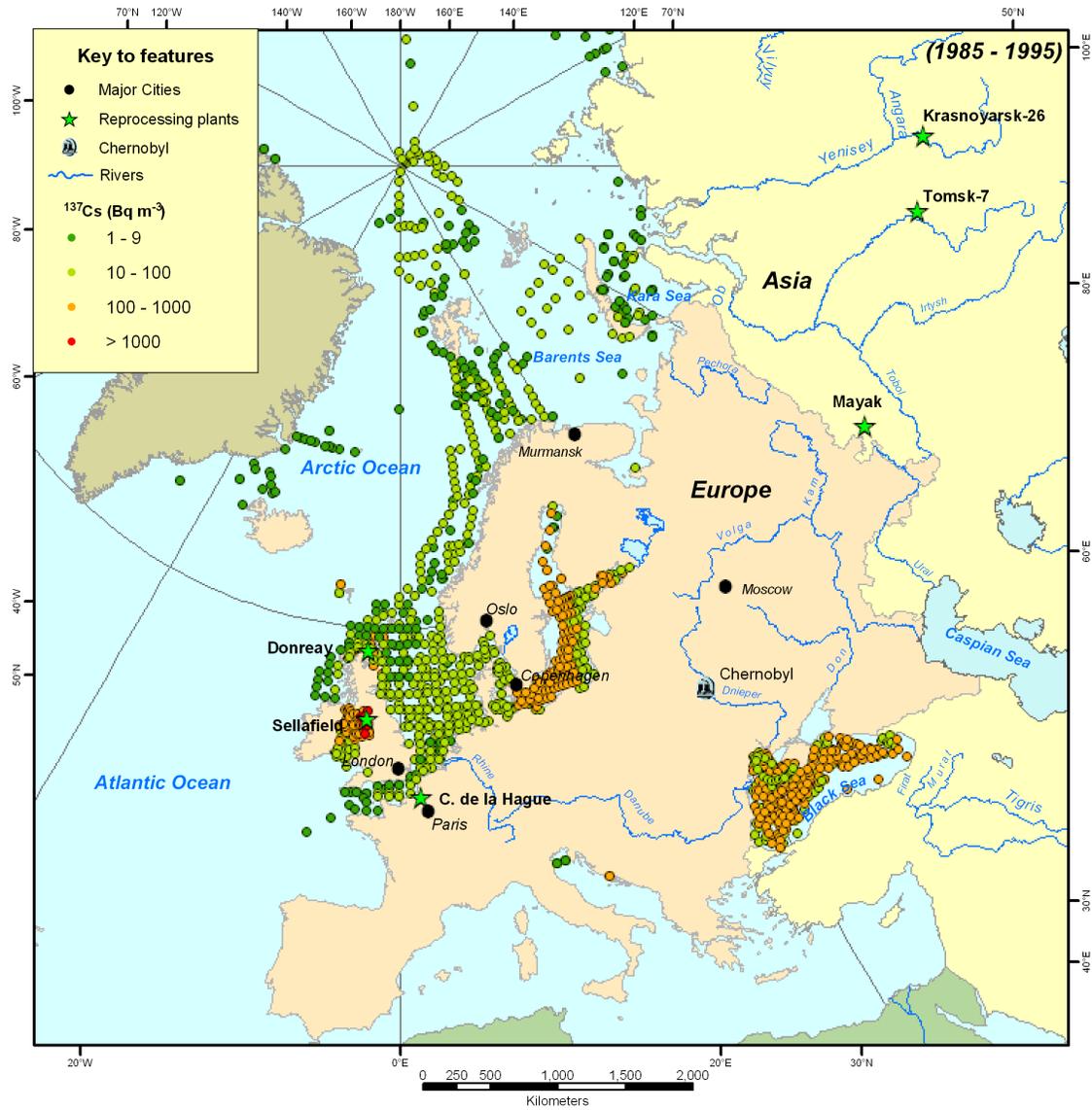


Figure 11. Activity concentration of ^{137}Cs in surface waters of the Northeast Atlantic, Black Sea, and Arctic Ocean (1985–1995) in relation to reprocessing facilities (Data source: Dr. Kathy Crane, Office of Naval Research, U.S.A.)

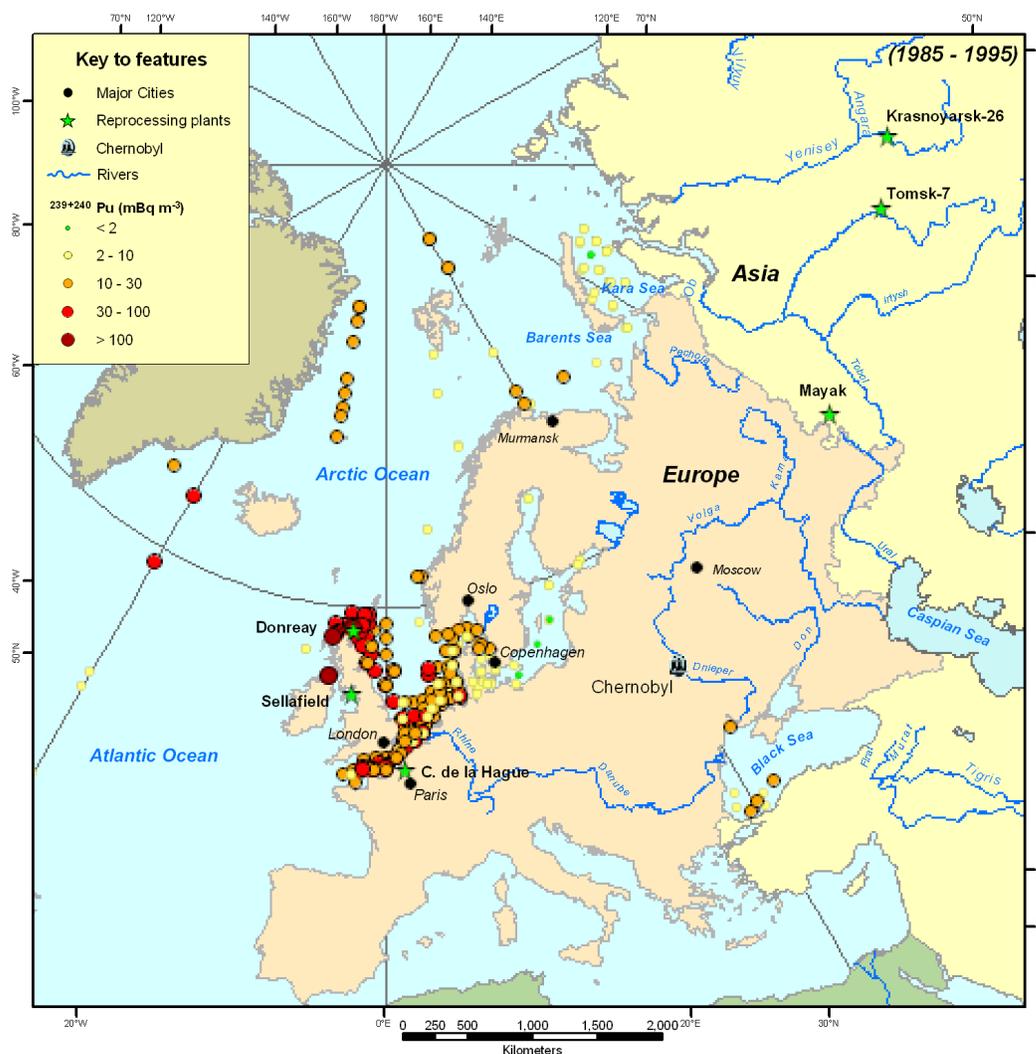


Figure 12. Activity concentrations of $^{239+240}\text{Pu}$ in surface waters of the Northeast Atlantic, Black Sea, and Arctic Ocean (1985–1995) in relation to reprocessing facilities (Data source: Dr. Kathy Crane, Office of Naval Research, U.S.A.)

About 22% of the ^{137}Cs Sellafield discharge passes into the Barents Sea en route to the Nansen Basin with another 13% passing through the Fran Strait (Kershaw and Baxter, 1995). Quantifying the fluxes of other radionuclides can be more problematic (Kershaw and Baxter, 1995) because inflowing North Atlantic water entering and mixing with Arctic waters is diluting radioactive contamination previously dominated by discharges from Sellafield

during the 1950s and 1960s. Conversely, an increase in activity concentration of ^{137}Cs in western waters east of Greenland is attributed to old “ ^{137}Cs rich” Sellafield-labeled waters entering the Arctic circulation and being transported back to the North Atlantic through the East Greenland Current (AMAP, 1998). Transit times for export of ^{137}Cs to the Barents Sea are estimated to be around 5 to 6 years for Sellafield, and 17 to 36 months for La Hague-

labeled waters (Guegueniat et al., 1997; Kershaw and Baxter, 1995).

Another major mechanism for water transport of radioactive contamination in the Arctic region relates to the unique surface-water hydrology and complex system of rivers, lakes, and marshes that make up the Russian Arctic territories. Several large, northward flowing rivers serve as sinks for terrestrial contaminant transport into the Russian Arctic, including ground water contamination from what is described as the largest shallow water artesian basin in the world. The main sources of artificial radioactivity transported in river systems are previously deposited global fallout from atmospheric nuclear weapons testing and environmental releases from Russian nuclear production facilities. The former Soviet Union also conducted a number of Peaceful Nuclear Explosions (PNE) for civilian use including cratering tests, denotations designed to stimulate gas and oil production or increase mineral recovery. The total yield from all PNEs conducted by the Former Soviet Union was around 1.6 Mt (Bradley 1997). Many of these nuclear tests caused significant contamination adjacent to the Ground Zero (GZ) but it is not clear what impact these detonations may contribute to Arctic contamination. It is expected that releases to the Ob and Yenisey were negligible. It has been reported that about 3.2 PBq of radioactivity was released into the Irtysh River (Bradley, 1997), presumably in runoff from the Semipalatinsk test site (Table 1).

Other sources of radioactive contamination in the Arctic region include offshore transport and redistribution of radioactivity from nuclear sites on the southern coastline of Novaya Zemlya and other accidents involving nuclear ships, submarines, and/or nuclear weapons. The

Former Soviet Union submarine SSN Komsomolets sank off Bear Island (Figure 2) on 7 April of 1989 after a fire broke out in the stern section of the vessel. The reactor was shut down prior to the vessel sinking but concerns have been raised about leakage of radioactivity from the reactor and two nuclear torpedoes aboard, one of which was fractured (Bergman and Baklanov, 1998). The reactor core and warheads reportedly contain a total of about 22 TBq of ^{239}Pu (10 kg), 2.4 PBq of ^{90}Sr , and 2.7 PBq of ^{137}Cs (decay corrected to 1 January, 1995) (Høibråten and Thoresen, 1995). Scientific expeditions to the site indicate very little loss of radioactivity from the submarine (Kolstad, 1995).

The Former Soviet Union conducted three underwater nuclear detonations near Chernaya Bay, a 15-km fjordic inlet on the southern coastline of Novaya Zemlya. It is estimated that approximately 11 TBq of $^{239+240}\text{Pu}$ from the tests has been retained in the local sediments (Smith et al., 2000). Chernaya Bay is among the world's most contaminated marine environments. Plutonium-239+240 levels in the central region of Chernaya Bay exceed 8000 Bq kg^{-1} and are characterized by low $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios (~ 0.03) associated with the detonation of low-yield nuclear tests (Smith et al., 1995). By exploiting the large difference in $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios between the Chernaya Bay fallout (~ 0.03) and atmospheric fallout (~ 0.18) end-members, Smith et al. (2000) estimated that an additional 2 TBq of Chernaya Bay labeled plutonium resides in the eastern Barents Sea from offshore transport from the embayment. A plume of low $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio plutonium is observed in a northwestern direction along the southern coastline of Novaya Zemlya indicating an additional pathway for transfer of previously deposited fallout debris into the Arctic Ocean.

The Mayak production association complex is located along the Techa River on the upper reaches of the Ob–Irtysh–Tobol–Iset river system. Approximately 37,000 PBq of radioactive waste has been generated since commencement of operations at Mayak in 1948. Today, authorities acknowledge that at least 4800 PBq of radioactivity was released directly into surface waters adjacent to the site (Hamilton, this report). Most of the radioactive contamination is held back in lakes and reservoirs on the Upper Techa; however, historical releases are known to have caused significant contamination of the riverbed and associated floodplain. A large number of residents living on the Techa River during the early 1950s received significant radiation exposures. Health studies on the population continue to this day. Overflow and seepage water from the dams, as well as migration of radionuclides deposited in the floodplain and especially runoff from the Asanov swamps, provide a possible source-term for transport of radioactive contaminants from Mayak to the Kara Sea. The Tomsk-7 and Krasnoyarsk-26 reprocessing facilities are located on tributaries to the Ob and Yenisey rivers, respectively (Figure 2). Despite the large releases of radioactivity from Russian reprocessing facilities and potential for riverine transport of the contamination to the Kara Sea, the major fraction of ^{137}Cs and plutonium deposited in delta sediments of the Ob and Yenisey rivers is said to be derived from fallout from atmospheric nuclear weapons tests (Sayles et al., 1997; Baskaran et al., 1995). The latter findings are supported by analyses of Pu isotopes, and $^{238}\text{Pu}/^{239+240}\text{Pu}$ and $^{239+240}\text{Pu}/^{137}\text{Cs}$ activity ratios, all of which were indistinguishable from global fallout. Using high quality mass-spectrometric measurements, Cooper et al. (2000) has shown that plutonium isotopic signatures in Ob and

Yenisey River sediments are distinctly different from those of northern-hemisphere stratospheric fallout, arguing that the ratios are more indicative of the presence of weapons grade plutonium from Russian reprocessing facilities located thousands of kilometers upstream. Observed ratios in sediments from the Eurasian Arctic Ocean were also shown to be inconsistent with significant contributions of plutonium to arctic sediments from western European facilities, namely Sellafield (Cooper et al., 2000). This work has re-addressed the need for a more thorough and accurate assessment of the fate and transport of radioactive effluents discharged into surface water from Russian reprocessing facilities (Hamilton, this report).

The USSR Hydrometeorological Service has maintained detailed records of the amount of ^{90}Sr entering the Kara Sea from the Ob and Yenisey rivers. It is estimated about 1.1 PBq of ^{90}Sr was transported to the Kara Sea between 1961 and 1989 (SCRF, 1995; Vakulovsky et al., 1993). A concurrent set of data does not exist for ^{137}Cs . However, using the average $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratio observed in river water over the same period (~ 0.1), the associated ^{137}Cs discharge to the Kara Sea is estimated at around 0.11 PBq. According to Aarkrog (1993), radionuclide contributions to the Arctic Ocean from atmospheric nuclear weapons fallout, discharges from Sellafield, and runoff from global fallout over land are 2.6 PBq, 1–2 PBq, and 1.5 PBq for ^{90}Sr , and 4.1 PBq, 10–15 PBq, and 0.5 PBq for ^{137}Cs , respectively. About 1–5 PBq of Chernobyl fallout was also delivered to the Arctic Ocean by direct deposition from the atmosphere and transport of Chernobyl labeled waters from the Baltic and North Seas.

In summary, large-scale contamination of the Arctic Ocean with artificial radionuclides (past and present) is controlled by four

primary sources: global fallout from atmospheric nuclear weapons testing, discharges from European reprocessing facilities, fallout from the Chernobyl accident, and runoff from Siberian rivers. The relative contributions from different sources are both radionuclide and transport pathway specific. Through 1993, the inventory of ^{90}Sr in the Arctic increased about twofold over what was expected from global fallout deposition alone. The associated ^{137}Cs inventory increased about 4 to 6 times. The main source of the additional ^{90}Sr and ^{137}Cs in the Arctic has been traced to discharges from European reprocessing entering the Arctic via the Norwegian Coastal Current. Sellafield contributes as much as 90% of the excess ^{137}Cs inventory and 60% of the excess ^{90}Sr inventory. Chernobyl fallout is a significant contributor to the ^{137}Cs inventory in the Arctic Ocean and supplies anywhere from 5–40% of the additional ^{137}Cs inventory above the expected global fallout deposit. Chernobyl does not contribute significantly to the total ^{90}Sr inventory in the Arctic. The Dvina, Pechora, Ob and Yenisey Rivers in northwestern Russia and Siberia all drain into the Kara Sea and constitute an important pathway for export of previously deposited global fallout debris over land to the Arctic. Based on several studies conducted during the 1990s, about 40–60% of the additional ^{90}Sr and 3–5% of the additional ^{137}Cs in the Arctic Ocean may be from export of previously deposited fallout debris over land. This conjecture is mainly based on the lack of convincing evidence for significant riverine inputs to the Kara Sea of artificial radionuclides from Russian reprocessing facilities located upstream with the exception of weapons grade plutonium and, possibly, ^{129}I . Discharges of ^{99}Tc and ^{129}I from Sellafield and La Hague dominate the total

oceanic inventory, especially those from La Hague. The estimated combined discharge of ^{129}I from reprocessing through 1994 is approximately 1440 kg (8.7 TBq) (Yiou, 1995) or about 16 times the global release from atmospheric nuclear weapons testing (Table 3). Other sources of radioactive contamination in the Arctic include indiscriminant dumping of nuclear waste, accidents involving nuclear ships and submarines, and offshore transport of plutonium from tests sites on Novaya Zemlya. Gaps in current understanding will require a continuing level of radiological surveillance of known and potential source-terms, source-term related assessments, and studies relating ocean dynamics and contaminant transport processes. There remain a number of critical challenges in developing appropriate long-term strategies to protect the Arctic and the wider regional environment from past, present, and potential future sources of artificial radioactivity. These are seen as negligent nuclear waste management practices in the handling of spent nuclear fuel, the management of legacy waste and nuclear materials in inventory, and risk of a catastrophic release from waste containment systems holding back high-level radioactive waste from Mayak and other Russian reprocessing facilities on the Ob and Yenisey rivers. There is also a need to improve the safety of nuclear power plants at Kola and Bilibino. Some agreements involving Russian and bi- and trilateral cooperations with Norway, the United States, and the European Community have already taken steps to improve the management of waste and spent nuclear fuel on the Kola Peninsula.

The Ocean Water Column

Early measurements of artificial radionuclides in the oceans demonstrated that global fallout from atmospheric nuclear weapons tests penetrated the deep ocean. Interpretation of these data was complicated because of the heterogeneous nature of the global fallout deposition and questions concerning the quality of the measurements. No consensus could be reached about the significance of ^{90}Sr penetration into the deep ocean or on the total oceanic inventory (Volchok et al., 1971). These early studies highlighted the need for more systematic and long-term studies, and raised expectations about the value of using global fallout radionuclides as tracers of oceanographic processes. Modern day “radionuclide oceanography” really only began with the commencement of the GEOSECS (Geochemical Ocean Section) program in 1973–74 where very carefully collected, large volume hydrographic stations were occupied and fractions of these samples made available for analysis of ^3H , ^{14}C , ^{90}Sr , ^{137}Cs and the transuranic elements (Broecker et al., 1985; Livingston et al., 1985; Bowen et al., 1980). In general, ^{90}Sr and ^{137}Cs concentrations in the surface ocean have decreased steadily since the early 1960s to present-day levels of around <1 to 4 Bq m^{-3} (Bourlat et al., 1996). Early measurements showed clear latitudinal concentration gradients as expected from global fallout patterns but these trends have since become more obscure (Hamilton et al.,

1996). Regional anomalies were also identified in association with discharges from reprocessing plants; upwelling and other oceanographic processes; and localized inputs from nuclear tests conducted in the Marshall Islands. As expected, the observed activity concentration of ^{90}Sr (and ^{137}Cs) in surface waters of the Southern Hemisphere was much lower than those reported for northern latitudes.

Depth distributions of ^{137}Cs and ^{90}Sr in the water column were characterized by sharp concentration gradients extending down to depths of 1000–2000 m (Nagaya and Nakamura, 1987; Nagaya and Nakamura, 1984). The depth distribution patterns were similar to those observed for fallout ^3H (Roether, 1974) and attributed to the conservative nature of ^{90}Sr and ^{137}Cs fallout. However, a significant fraction of ^{137}Cs and ^{90}Sr penetrated the deep ocean, and there was no clear understanding of the processes involved. The most probable explanation came from Martin (1970), Honjo (1980), Iseki (1981), and others who postulated that global fallout radionuclides are actively transported to the deep ocean on biogenic particles. Elevated levels of ^{137}Cs were also observed in waters immediately above the seafloor. This was attributed to preferential scavenging of ^{137}Cs into bottom sediments and remobilization processes, and/or advection of nuclide-rich water from other regions (Nakamura and Nagaya, 1985; Nakamura and Nagaya, 1975; Noshkin and Bowen, 1973).

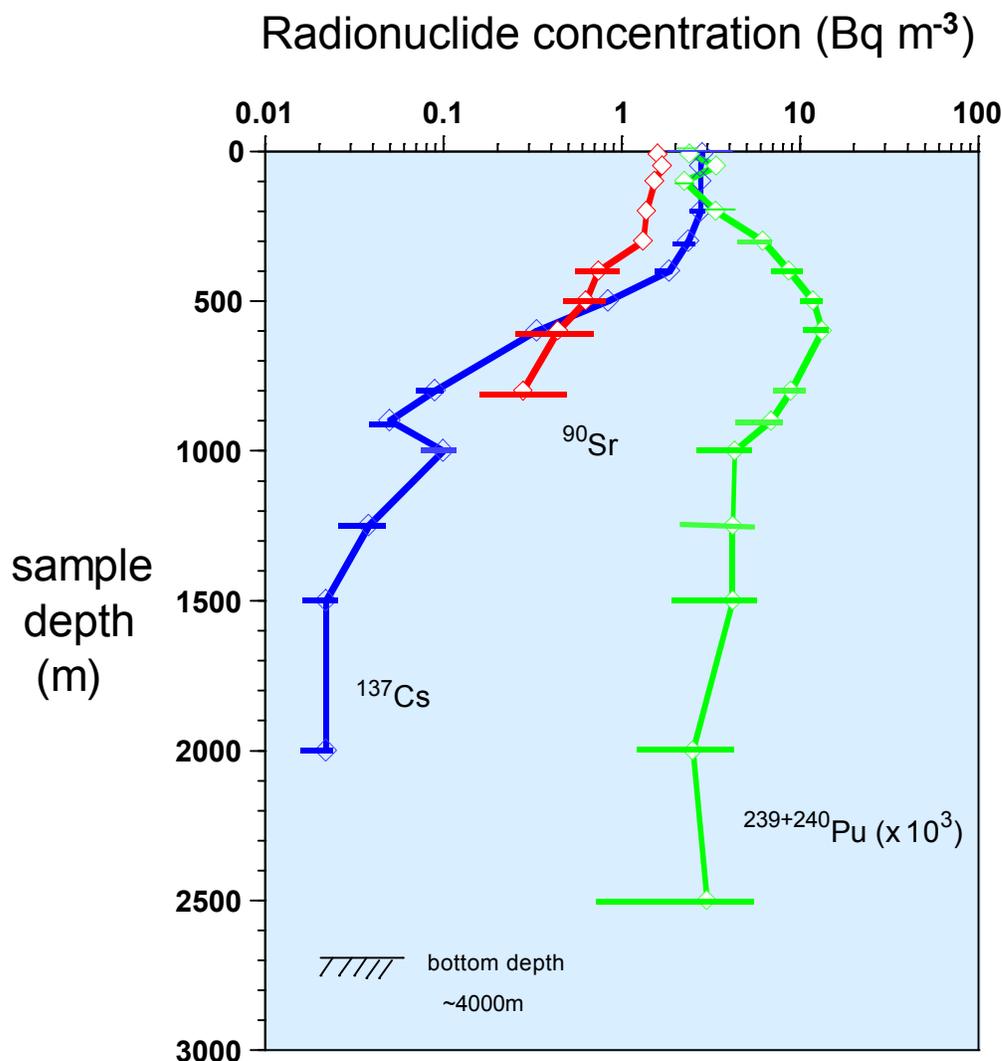


Figure 13. Depth distribution of ^{90}Sr , ^{137}Cs , and $^{239+240}\text{Pu}$ in the ocean, Tuamoto Archipelago, French Polynesia (Data source: Hamilton et al., 1996)

The general depth distribution profiles of key radionuclides such as ^{90}Sr , ^{137}Cs , and $^{239+240}\text{Pu}$ do not appear to have changed significantly over the past 25 years (Hamilton et al., 1996) (Figure 13). One of the more interesting features of plutonium behavior in the oceans is the widespread occurrence of a subsurface maximum ranging in depth from 250 to 1000 m (Nakanishi et al., 1995; Bowen et al., 1980; Noshkin and Wong, 1975). Although there is some evidence to suggest

the depth of the subsurface maximum and proportion of plutonium residing in the deep ocean has increased over time (Nakanishi et al., 1984; Bowen et al., 1980), early transport models based on irreversible particle scavenging predicted a more rapid transfer of plutonium to the deep ocean. A reversible process of biologically mediated scavenging that varies in space and time best explains the depth distribution and transport of long-lived radionuclides such as plutonium in the deep

oceans. Highly productive surface waters enhance the vertical transport of particle reactive species and provide a possible mechanism where radionuclides can be preferentially transported to the deep ocean and onto the seafloor. There is also strong evidence to suggest that the main vector for transport of particle reactive radionuclides to the deep ocean are large, rapidly sinking zooplankton faecal pellets (Fowler et al., 1983; Fisher and Fowler, 1983). Smaller particles and micropellets sink slowly and, hence, are more subject to biogeochemical cycling. Therefore, the appearance of a subsurface maximum for plutonium may be a function of both the particle reactivity of plutonium and dynamics of these transformation processes. A short residence time and higher affinity for removal of plutonium on sinking particles is supported by

the general findings that particulate $^{239+240}\text{Pu}$ concentrations range up to ~20% of the total activity concentrations in surface waters compared with less than 1% for ^{137}Cs (Hirose et al., 1992; Fowler et al., 1983; Noshkin and Wong, 1980). Although there is a small but significant flux of artificial radionuclides depositing on the sea floor, about 80% of the total $^{239+240}\text{Pu}$ inventory and 95% of the total ^{137}Cs inventory in the open ocean remains in the water column. Long-term monitoring studies in French Polynesian waters indicate ^{137}Cs concentrations in surface water decrease with an apparent half-life of about 14 years (Figure 14, Bourlat and Martin, 1992). Using a simple box model, Hirose et al. (1992) estimated that the residence times of $^{239+240}\text{Pu}$ and ^{137}Cs in the surface-mixed layer of the western North Pacific were around 4 and 9.1 years, respectively.

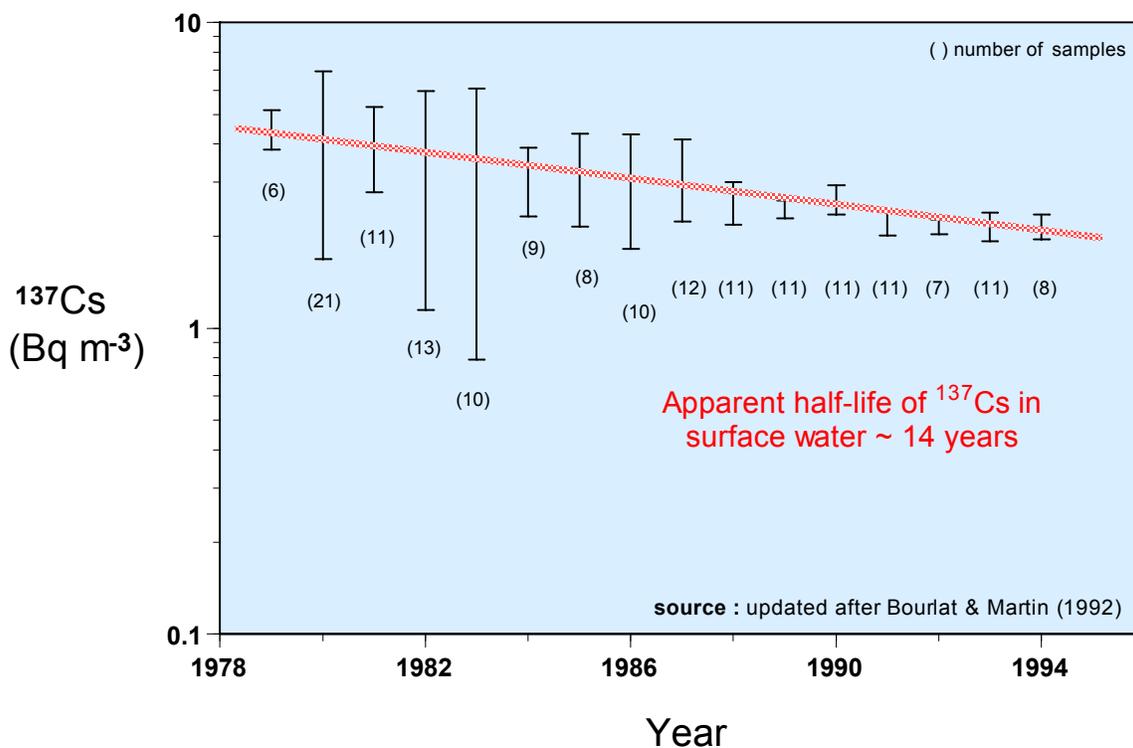


Figure 14. Temporal pattern in the activity concentration of ^{137}Cs in French Polynesian surface waters (modified after Hamilton et al., 1996)

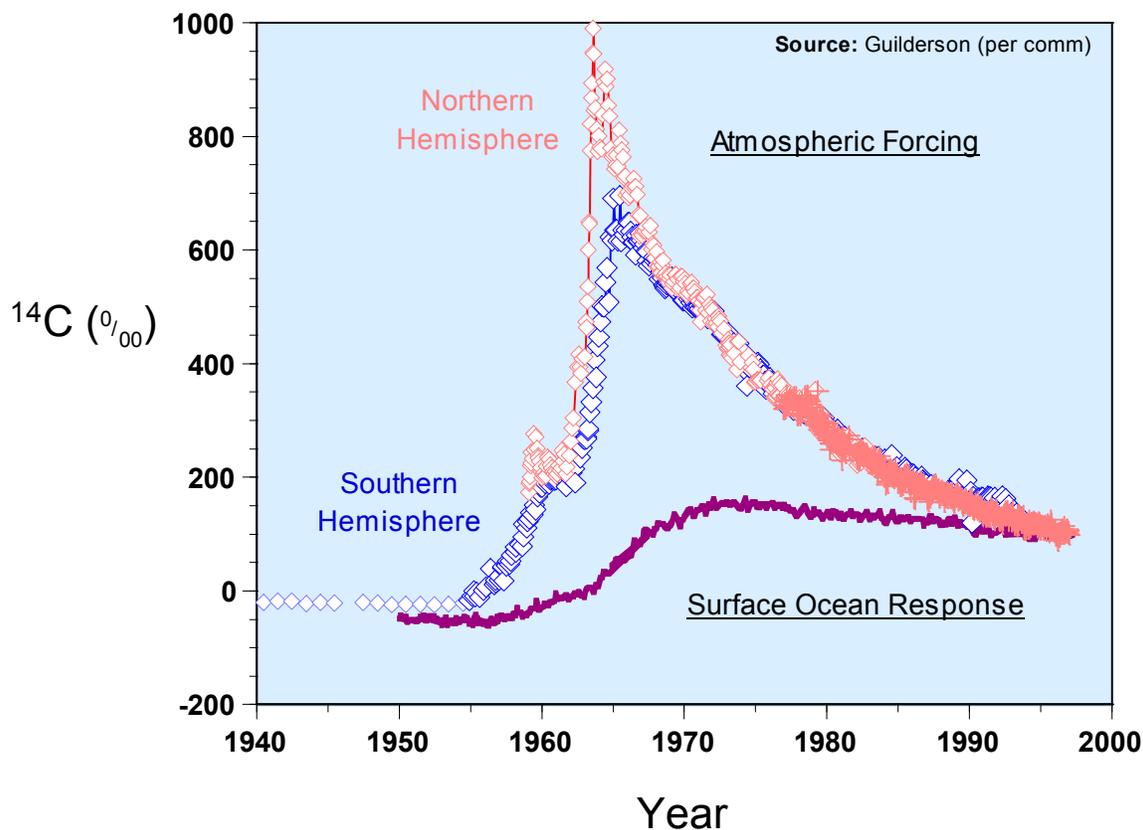


Figure 15. Surface ocean response to global dispersion and deposition of ^{14}C in the ocean: a subtropical South Pacific high-resolution coral record from Rarotonga (21°S , 159°W) (Data source: Guilderson, personal communication).

Today, the global dispersion and deposition of fallout radionuclides on the earth's surface is largely controlled by tropospheric resuspension of previously deposited debris on land (Monetti, 1996; Nakanishi et al., 1995). For example, seasonal patterns in the deposition of ^{137}Cs and $^{239+240}\text{Pu}$ over Japan and on the Korean Peninsula are directly linked to 'yellow dust' events (Lee, 1994). According to Hamilton et al., (1996), the annual deposition rates of ^{137}Cs and $^{239+240}\text{Pu}$ into the Japan Sea from resuspension of dust are about 0.8 and 0.03 MBq km^{-2} , respectively; and are very similar to the leveling-off aerosol flux measured by the Meteorological Research Institute (MRI) in Japan.

Other important global fallout radionuclides of oceanic significance include tritium and ^{14}C . Very large quantities of tritium ($T_{1/2} = 12.3$ years) and ^{14}C ($T_{1/2} = 5730$ years) were produced in atmospheric testing of high-yield nuclear weapons during the 1950s and 1960s (Table 3). Bomb tritium generated in nuclear detonations was quickly incorporated into stratospheric water molecules and upon transfer to the troposphere; the tritiated water was rapidly scavenged onto raindrops and deposited by rainout over sea or land (Broecker et al., 1995). Bomb tritium preferentially resides in the high-latitude northern hemispheric oceans (Weiss and Roether, 1980) and is only present as tritiated water. Much of the bomb tritium

deposited over land was transferred to the ocean in runoff or via evaporation. The delivery of tritium from land to sea is a major pathway but the input is poorly constrained. With the exception of small losses by atmospheric exchange processes there is no sink for oceanic tritium other than loss by radioactive decay. The concentration of tritium in the ocean is normally reported in TU (tritium units) used to denote the number of tritium atoms per 10^{18} atoms of hydrogen or synonymously, by the tritium ratio (TR). One TU is equivalent to 0.12 Bq, or in terms of atoms, 6.7×10^7 atoms per liter of water. The tritium oceanic inventory in 1973 was estimated to be around 60,000 PBq. The activity concentration of tritium in near-surface ocean waters is about 0.3 Bq kg^{-1} of water, but its isotopic concentration (i.e., the molar ratio of tritium to stable hydrogen) is extremely low (on the order of 10^{-18}). In contrast, ^{14}C was incorporated into CO_2 and transferred to the sea by invasion from the

atmosphere. Differences in environmental pathways between ^{14}C and tritium led to a major geographic separation between the two isotopes in the oceans. Naturally occurring ^{14}C is produced in the atmosphere by the action of neutrons from cosmic rays on nitrogen. Atmospheric nuclear testing more than doubled the natural cosmogenic atmospheric inventory. Bomb ^{14}C was spread uniformly throughout the atmosphere and into the oceans, and taken up into the biosphere by photosynthesis. Penetration of bomb ^{14}C in the surface ocean is illustrated in Figure 15, showing a rapid increase in the amount of ^{14}C incorporated into a coral core extending back to the pre-nuclear age. The global ocean bomb ^{14}C inventory as of 1972 was about 305×10^{26} atoms (Radiocarbon Units-RCU or 116 PBq) (Broecker et al., 1985) increasing to about 435×10^{26} atoms (RCU or 167 PBq) by 1985 from post-1972 transfer of ^{14}C to the ocean (Lassey et al., 1988).

Acknowledgments

I thank Dr. Kathy Crane, Office of Naval Research (ONR), and Dr. Tom Guildersen, Lawrence Livermore National Laboratory (LLNL), for providing data for graphics. Ms. Lynn Wilder assisted with the preparation of maps and figures. Work performed under the auspices of the United States Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

References

- Aarkrog A., J. Simmonds, P. Strand, G. Christensen, and B. Salbu (2000). Radiological Assessment of Past, Present and Potential Sources to Environmental Contamination in the Southern Urals and Strategies for Remedial Measures (SUCON, Risø-R-1243 (EN), Risø National Laboratory, Roskilde, December 2000, 71 pp.
- Aarkrog A. (1993). Radioactivity in the polar regions-main sources. In: *Environmental Radioactivity in the Arctic and Antarctic*,

- (editors, P. Strand and E. Holm), Norwegian Radiation Protection Authority, Østerås, Norway, pp. 15-34.
- Aarkrog A. (1988). Worldwide data on fluxes of $^{239,240}\text{Pu}$ and ^{238}Pu to the oceans, In: *Inventories of Selected Radionuclides in the Oceans*, IAEA-TECDOC-481, International Atomic Energy Agency, Vienna, pp. 103-138.
- Aarkrog A., S. Boellskifte, H. Dahlgaard, S. Duniec, E. Holm, and J.N. Smith (1987). Studies of transuranics in an arctic marine environment, *J. Radioanal. Nucl. Chem.*, 1115, 39-50.
- Aarkrog A., H. Dahlgaard, K. Nilsson, and E. Holm (1984). Further studies of plutonium and americium at Thule, Greenland. *Health Phys.*, 46, 29-44.
- Aarkrog A., H. Dahlgaard, L. Hallstadius, H. Hansen, and E. Holm (1983). Radiocesium from Sellafield effluents in Greenland water, *Nature*, 304, 49-51.
- Adams C.E., N.H. Farlow, and W.R. Schell (1960). The compositions, structure and origins of radioactive fall-out particles. *Geochim. Cosmochim. Acta*, 18, 42-56.
- AMAP (1998). *Radioactivity*, AMAP Assessment Program Arctic Pollution Issues, Chapter 8, Arctic Monitoring and Assessment Programme (AMAP) (editors, P. Strand, M. Balonoc, A. Aarkrog, M.J. Bewers, B. Howard, A. Salo, and Y.S. Tsururov), Oslo, Norway, pp 525-610.
- Baskaran M., S. Asbill, P. Santchi, T. Davis, J. Brooks, M. Champ, V. Makeyev, and V. Knlebovich (1995). Distribution of $^{239+240}\text{Pu}$ and ^{238}Pu concentrations in sediments from the Ob and Yenisey rivers and the Kara Sea, *Appl. Radiat. Isotope*, 46, 1109-1119.
- Baumgartner A., and E. Reichel (1975). *World Water Balance: Mean, Annual Global, Continental and Maritime Precipitation, Evaporation and Run-off*, Elsevier, N.Y., 179 pp.
- Baxter M.S., S. Ballestra, J. Gastaud, T.F. Hamilton, I. Harms, L. Huynh-Ngoc, L. Liong Wee-Kwong, I. Osvath, P. Parsi, H. Petterssen, P.P. Povinec, and A. Sanchez (1995). Marine Radioactivity studies in the vicinity of sites with potential radionuclide releases, International Symposium on Environmental Impact of Radioactive Releases, International Atomic Energy Agency (IAEA), Vienna, Austria, 8-12 May, 1995.
- Baxter M.S., I.G. McKinley, A.B. Mackenzie, and W. Jack (1979). Windscale radiocesium in the Clyde Sea Area. *Marine Pollut. Bull.*, 10, 116-120.
- Beck H.L., and P.W. Krey (1983). Radiation exposure in Utah from Nevada nuclear tests. *Science*, 220, 18-24.
- Bergman R., and A. Baklanov (1998). Radioactive sources of main radiological concern in the Kola-Barents region, Defense Research Establishment, FOA-B-98- 00343-861-SE, Sweden, 78 pp.
- Betis J. (1993). Impact of the La Hague reprocessing plant on the surrounding environment, In: *Environmental Impact of Nuclear Installations*, (editors, H. Volke and S. Pretre), Les Éditions de Physique, Les Ulis Cedex A, France, pp. 33-36.

Bowen V.T., V.E. Noshkin, H.D. Livingston, and H.L. Volchok (1980). Fallout radionuclides in the Pacific Ocean: vertical and horizontal distributions, largely from GEOSECS stations, Earth Planet. Sci. Lett., 49, 411-434.

Bol'shakov V.N., R.M. Aleksakhin, L.A. Bol'shov, V.N. Chukanov, L.A. Kochetkov, V.I. Petukhov, and A.F. Tsyb (1991). *Conclusion of the Commission for Evaluating the Ecological Situation in the sphere of Influence of the Industrial Plant (IP) 'Mayak' of the USSR Ministry of Atomic Energy*, Organized by the Presidium of the USSR Academy of Sciences No. 1140-501, on June 12, 1990, Moscow, Russia.

Botov N.G. (1992). ALWP-67: A Little-Known Big Nuclear Accident, Proceedings of the Third International Conference on High Level Radioactive Waste Management, April 12-16, 1992, Las Vegas, Nevada.

Bourlat Y., J.-C. Millies-LaCroix, G. Le Petit, and J. Bourlignon (1996), ^{90}Sr , ^{137}Cs and $^{239+240}\text{Pu}$ in world ocean water samples collected from 1992 to 1994, In: Radionuclides in the Oceans, Inputs and Inventories, (coordinators; P. Guéguéniat, P. Gwermain and H. Métivier), Chapter 2, Les Éditions de Physique, Les Ulis Cedex A, France, pp. 75-93.

Bourlat Y., J.-C. Millies-LaCroix, and R. Nazard (1995). Determination of plutonium radioactivity in Mururoa lagoon water, J. Radioanal. Nucl. Chem., 197, 387-408.

Bourlat Y., and G. Martin (1992), Precise determination of the concentration of radiocesium in the water of Mururoa lagoon, J. Environ. Radioactivity, 17, 13-29.

Bradley D.J. (1997). Behind the Nuclear Curtain: Radioactive Waste Management in the Former Soviet Union (editor, D. R. Payson), Battelle Press, 716 pp.

Bradley D.J., C.W. Frank, Y. Mikerin (1996). Nuclear Contamination from Weapons Complexes in the Former Soviet Union and the United States, Physics Today, 49, 40-45.

Bradley D.L., and U.P. Jenquin (1995). Radioactive Inventories and Sources for Contamination of the Kara Sea by Rivers, PNWD-2316, Richland, Washington.

Broecker W.S., S. Sutherland, and W. Smethie (1995). Oceanic radiocarbon: Separation of the natural and bomb components, Global Biogeochem. Cycles, 9, 263-288.

Broecker W.S., T.H. Peng, G. Ostland, and M. Stuiver (1985). The distribution of bomb radiocarbon in the ocean, J. Geophys. Res., 90, 6953-6970.

Buesseler, K.O. (1997). The isotopic signature of fallout plutonium in the North Pacific, J. Environ. Radioactivity, 36, 69-83.

Buesseler, K.O., (1986). Plutonium isotopes in the North Atlantic, Ph.D. Thesis, MIT/WHOI Joint Program in Oceanography, 220 pp.

- Christensen G.C., S.V. Malyshev, P. Strand, B. Salbu, G.N. Romanov, T.D. Selnaes, D. Oughton, Y.V. Glagolenko, I. Amundsen, A.L. Rudjord, T.O. Bjerck, and B. Lind (1995). *Radioactive Contamination in the Environment of the Nuclear Enterprise 'Mayak' PA. Results from the Joint Russian-Norwegian Field Work in 1994*. Presented at the Second International Conference on Environmental Radioactivity in the Arctic, August 21-25, 1995, Oslo, Norway.
- Cochran T.B., R.S. Norris, and A.O. Bukharin (1995). *Making the Russian Bomb: From Stalin to Yeltsin*, Westview Press.
- Cochran T.B., R.S. Norris, and K.L. Suokko (1993). Radioactive contamination at Chelyabinsk-65 Russia, *Annu. Rev. Energy Environ.*, 18: 507-528.
- Compton A.A. (1956). *Atomic Quest, A Personal Narrative*, Oxford University Press, New York, 370 pp.
- Cooper L.W., J.M. Kelley, L.A. Bond, K.A. Orlandini, and J. M. Grebmeier (2000). Sources of the transuranic elements plutonium and neptunium in arctic marine sediments, *Marine Chemistry*, 69, 253-276.
- Cooper L.W., I.L. Larsen, T.M. Beasley, S.S. Dolvin, J. M. Grebmeier, J. M. Kelley, M. Scott, and A. Johnson-Pyrtle (1998). The distribution of radiocesium and plutonium in sea ice-entrained Arctic sediments in relation to potential sources and sinks, *J. Environ. Radioactivity*, 39, 279-303.
- Crocker G.R., J.D. O'Connor, and E.C. Freiling (1966). Physical and radiochemical properties of fallout particles, *Health Phys.*, 12, 1099-1104.
- Dahlgaard H., Q. Chen, H. Nies, R.D. Ibbett, and P.J. Kershaw (1995). On the background level of ^{99}Tc , ^{90}Sr , and ^{137}Cs in the North Atlantic, *J. Mar. Syst.*, 571-578.
- Dahlgaard H. (1993). Where does all the Cs and Sr in Greenland come from? In: *Environmental radioactivity in the Arctic and Antarctic*, (editors, P. Strand and E. Holm), Norwegian Radiation Protection Authority, Østerås, Norway, pp. 121-124.
- Dahlgaard H., Q.J. Chen, and S.P. Nielsen (1991). Radioactive tracers in the Greenland Sea, In: *Radionuclides in the Study of Marine Processes*, (editors; P.J. Kershaw and D.S. Woodhead), Elsevier Applied Science, London, pp. 12-22.
- Dahlgaard H., Q. Chen, H. Nies, R.D. Ibbett, and P.J. Kershaw (1988). On the background level of ^{99}Tc , ^{90}Sr , and ^{137}Cs in the North Atlantic, *J. Mar. Syst.*, 6, 571-578.
- Dahlgaard H., A. Aarkrog, L. Hallstadius, E. Holm, and J. Rioseco (1986). Radiocesium transport from the Irish Sea via the North Sea and Norwegian Coastal Current to East Greenland, *Rapport et Proces-Verbaux Reunion Conseil Internationale Exploration de Mer*, 186, 70-79.
- Dailov-Danilyan V.I. (1993). Statement (LC/16/J/4) made at the 16th Consultative Meeting of the Contracting Parties to the London Convention 1972, 10 November, 1993.

Degteva M.O., M.I. Vorobiova, V.P. Kozheurov, E.I. Tolstykh, L.R. Anspaugh, and B.A. Napier (2000). Dose reconstruction system for the exposed population living along the Techa River, *Health Phys.*, 78, 542-554.

Degteva M.O., V.P. Kozheurov, and M.I. Vorobiova (1994). General Approach to Dose Reconstruction in the Population Exposed as a Result of the Release of Radioactive Waste into the Techa River, *Sci. Total Environ.*, Special Issue, Radiation Exposure in the Southern Urals.

Dobry T.J. Jr. (1980). Transuranic Elements in Space Nuclear Power Systems, In: *Transuranic Elements in the Environment*, (editor; W.C. Hanson), DOE/TIC-22800, pp 83-85.

DOE/EM (1997). Linking Legacies: Connecting the Cold War Nuclear Weapons Production Processes to their Environmental Consequences, U.S. Department of Energy/Office of Environmental Management, DOE/EM-0319, Washington DC, 229 pp.

DOE (1982). Enewetak Radiological Support Project, NVO-213, United States Department of Energy, Nevada Operation Office, Nevada, p 158.

Drozhko E.G., B.V. Nikipelov, A.S. Nikiforov, A.P. Suslov, and A.F. Tsarenko (1989). *Experience in radioactive waste management at the Soviet radiochemical plant and main approaches to waste reliable confinement development*, Paper presented at the International symposium on the Safety Assessment of Radioactive Waste Repositories, Paris, France, October 9-12, 1989.

Eisenbud M., and T. Gesell (1997), *Environmental Radioactivity, From Natural, Industrial, and Military Sources*, 4th edition, Academic Press, San Diego, U.S.A., 656 pp.

Fisher N.S. , and S.W. Fowler (1983). The role of biogenic debris in the vertical transport of transuranic wastes in the sea, In: *Oceanic Processes in Marine Pollution*, (editors; T.P. O'Connor, W.V. Burt, and I.W. Duedall), Volume 2, Chapter 16, Robert E. Krieger Publishing Co., Florida, pp. 197-207.

Fowler S.W., Ballestra S., La Rosa J. and R. Fukai (1983). Vertical transport of particulate-associated plutonium and americium in the northwest Pacific, *Deep-Sea Res.*, 7: 35-47.

GESAMP (1990). The State of the UNEP Regional Seas Report and Studies No. 115, UNEP, Group of Experts on the Scientific Aspects of Marine Pollution (GESAMP).

Gosling F.G. (1994). *The Manhattan Project: Making the Atomic Bomb*, DOE/HR-0096, U.S. Government Printing Office, Washington, D.C., September, 1994.

Gray J., S.R. Jones, and A.D. Smith (1995). Discharges to the environment from Sellafield Site, 1951-1992, *J. Radiol. Prot.*, 15, 99-131.

Guéguéniat P., P. Kershaw, J. Hermann, and P. Bailly du Bois (1997). New estimation of La Hague contribution to the artificial radioactivity of Norwegian water (1992-1995) and Barents Sea (1992-1997), *Sci. Total Environ.*, 202, 249-266.

- Hamilton T.F., J.-C. Milliès-LaCroix., and G.H. Hong (1996). ^{90}Sr (^{137}Cs) and Pu isotopes in the Pacific Ocean: Sources and Trends, In: *Radionuclides in the Ocean: Inputs and Inventories*, Chapter 2 (Coordinators; P. Guèguèniat, P. Germain and H. Mètivier), Les Éditions de Physique, Les Ulis cedex A, France, pp. 29-58.
- Hardy E.P., P.W. Krey, and H.L. Volchok (1973). Global inventory and distribution of fallout plutonium, *Nature*, 241, 444-445.
- Hetherington J.A. (1975). Some investigations into the behavior of plutonium in the marine environment, In: *Impacts of Nuclear Releases into the Aquatic Environment*, IAEA symposium (SM-198/29), pp. 193-212.
- Hewlett R.G., and O. Anderson Jr. (1962). *The New World, 1939-1946, Volume I, History of the United States Atomic Energy Commission*, Pennsylvania State University Press, University Park.
- Hicks H.G. (1982). Calculation of the concentration of any radionuclide deposited on the ground by offset fallout from a nuclear detonation, *Health Phys.*, 42, 585-600.
- Hicks H.G., and D.W. Barr (1984). Nevada test site fallout atom ratios: $^{240}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$, UCRL-53499/1, Lawrence Livermore National Laboratory, 4 pp.
- Hirose K., H. Amano, M.S. Baxter, E. Chaykovskaya, V.B. Chumichev, G.H. Hong, K. Isogai, C.K. Kim, S.H. Kim, T. Miyao, T. Torimoto, A. Nikitin, K. Oda, H.B.L. Pettersson, P.P. Povinec, Y. Seta, A. Tkjalin, O. Togawa, and N.K. Veletova (1999). Anthropogenic radionuclides in seawater in the East Sea/Japan Sea: Results of the first-stage Japanese-Korean-Russian expedition, *J. Environ. Radioactivity*, 43, 1-13.
- Hirose K., Y. Sugimura, and M. Aoyama (1992). Plutonium and ^{137}Cs in the western north Pacific: estimation of residence time of plutonium in surface waters, *Appl. Radia. Isot.*, 43, 349-359.
- Høibråten S., and P.E. Thoresen (1995). The sunken submarine Komsomolets and its effects on the environment, In: *Environmental Radioactivity in the Arctic*, (editors; P. Strand and A. Cooke), Norwegian Radiation Protection Authority, Østerås, Norway, pp. 2350-2355.
- Holm E., A. Aarkrog, S. Ballestra, and H. Dahlgaard (1986). Origin and isotopic ratios of plutonium in the Barents and Greenland Sea, *Earth Planet. Sci. Lett.*, 79, 27-32.
- Holm E., B.R.R. Persson, L. Hallstadius, A. Aarkrog, and H. Dahlgaard (1983). Radiocesium and transuranium elements in the Greenland and Barents Seas, *Oceanologica Acta*, 6, 457-462.
- Honjo S. (1980). Material fluxes and modes of sedimentation in the mesopelagic and bathypelagic zones, *J. Mar. Res.*, 38: 53-97.
- IAEA (1999). *Inventory of Radioactive Waste Disposal at Sea*, IAEA-TECDOC-1105, International Atomic Energy Agency, Vienna, 121 pp.
- IAEA (1998). *The Radiological Situation at the Atolls of Mururoa and Fangataufa, Main Report*, Radiological Assessment Report Series, Report by the International Advisory Committee, International Atomic Energy Agency, Vienna, 282 pp.

- IAEA (1997). *Predicted Radionuclide Release from Marine Reactors Dumped in the Kara Sea*, IAEA-TECDOC-938, International Atomic Energy Agency, Vienna, 83 pp.
- IAEA (1991). *Inventory of Radioactive Material Entering the Marine Environment: Sea Disposal of Radioactive Waste*, IAEA-TECDOC-588, International Atomic Energy Agency, Vienna, 83 pp.
- IMO (1993). Resolution LC.51(16): Report of the 16th Consultative Meeting of the Contracting Parties to the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, LC 16/14. International Maritime Organization, London, 1993.
- IMO (1972). International Conference on the Convention on the Dumping of Wastes at Sea, London 30 October-13 November 1972, Final Act of the Conference with Attachment Including the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, International Maritime Organization, London.
- International Herald Tribune, October 12-13, 1996.
- Iseki K. (1981). Vertical transport of particulate organic matter in deep Bering Sea and Gulf of Alaska, *J. Oceanogr. Soc. Japan*, 37: 101-110.
- ITAR-Tass (1992). Nuclear Reactors at Krasnoyarsk Shut Down After 30 Years, June 30, 1992.
- Jachmenev V. (1995). *Problems of Radioactive Contamination of the Chelyabinsk Region and the Way to Solve Them*, IIASA Meeting, Luxemburg, Austria, March 6-10, 1995.
- Jones V.C. (1985). *The Making of the Atomic Bomb*, U.S. Government Printing Office, Washington D.C., 1985
- Joseph, A.B., P.F. Gustafson, I.R. Russel, E.A. Schuert, H.L. Volchok, and A. Tamplin (1971). Sources of radioactivity and their characteristics, In: *Radioactivity in the Marine Environment*, National Academy of Sciences, Washington, Chapter 2, pp. 6-41.
- Kathren R.L. (1984). *Radioactivity in the Environment: Sources, Distribution and Surveillance*, Harwood Academic Publishers, Chur, Switzerland, 397 pp.
- Kautsky H. (1989). Content of radiocesium in the water of the northern North Atlantic during the year 1985, *Deutches Hydrographisches Zeitung*, 42 , 21-26.
- Kennedy J.W., G.T. Seaborg, E. Segre, and A.C. Wahl (1946). *Phys. Rev.*, 70, 555.
- Kershaw K., D. McCubbin, and K.S. Leonard (1999). Continuing contamination of north Atlantic and Arctic waters by Sellafield radionuclides, *Sci. Total Environ.*, 237/238, 119-132.
- Kershaw P., and A. Baxter (1995). The transfer of reprocessing waste from north-west Europe to the Arctic, *Deep-sea Res. II*, 42, 1413-1448.

- Kershaw P., D.S. Woodhead, M.B. Lovett, and K.S. Leonard (1995). Plutonium from European reprocessing operations-its behavior in the marine environment, *Appl. Radiat. Isot.*, 46, 1121-1134.
- Kershaw P.J., R.J. Pentreath, D.S. Woodhead, and G.J. Hunt (1992). A review of radioactivity in the Irish Sea. Aquatic Environment Monitoring Report, No. 32, MAFF Directorate of Fisheries Research, Lowestoft, U.K., 65 pp.
- Kiode M., K.K. Bertine, T.J. Chow, and E.D. Goldberb (1985). The $^{240}\text{Pu}/^{239}\text{Pu}$ ratio, a potential geochronometer, *Earth Planet. Sci. Lett.*, 72, 1-8.
- Kolstad A.K. (1995). Expeditions to "Komsomolets" in 1993 and 1994. NPRA Stralevern Rapport 1995:7, Norwegian Radiation Protection Authority, Østerås, Norway.
- Krey P.W. (1967). Atmospheric burnup of a plutonium-238 generator, *Science*, 158, 769-771.
- Krey P.W., E.P. Hardy, C. Pachucki, F. Rourke, J. Coluzza, and W. Benson (1976). Mass isotopic composition of global fallout plutonium in soil. In: *Transuranium Nuclides in the Environment*, IAEA-SM-199/39, International Atomic Energy Agency, Vienna.
- Lassey K.R., M.R. Manning, and B.J. O'Brien (1988). Assessment of the inventory of carbon-14 in the oceans: An overview, In: *Inventories of Selected Radionuclides in the Oceans*, IAEA-TECDOC-481, International Atomic Energy Agency (IAEA), Vienna, Austria, pp. 7-50.
- Lee Y.K. (1994). The study of the atmospheric deposition of anthropogenic radionuclides in the Korean Peninsula, MS Thesis, Hanyang University, Seoul, 43 pp. (in Korean).
- Livingston H.D., V.T. Bowen, S.A. Casso, H.L. Volchok, V.E. Noshkin, K.M. Wong, and T.M. Beasley (1985). Fallout nuclides in Atlantic and Pacific waters columns: GEOSECS Data, WHOI-85-19, Woods Hole, MA 02543, 73 pp.
- Livingston H.D., V.T. Bowen, and S.L. Kupferman (1982). Radionuclides from Windscale discharges II, their dispersion in Scottish and Norwegian coastal circulation, *J. Marine Res.*, 40, 1227-1258.
- Martin J.H. (1970). The possible transport of trace metals via moulted copopod exoskeletons, *Limnol. Oceanogr.*, 15: 756-761.
- Mitchell P.I., O.M. Condren, L. Vintro, and C.A. McMahon (1999). Trends in plutonium, americium and radiocesium acculation and long-term bioavailability in the western Irish Sea mud basin, *J. Environ. Radioactivity*, 44 , 223-251.
- Monetti M.A. (1996). Worldwide Deposition of Strontium-90 through 1990. Environmental Measurements Laboratory, EML-579, U.S. DOE, New York, 56 pp.
- Moscow Interfax (1994), 21 January 1994.

Nagaya, Y., and K. Nakamura (1987). Artificial radionuclides in the western northwest Pacific (II): ^{137}Cs and $^{239,240}\text{Pu}$ inventories in water and sediment columns observed from 1980 to 1986. *J. Oceanogr. Soc. Japan*, 43: 345-355.

Nagaya Y., and K. Nakamura (1984). $^{239,240}\text{Pu}$, ^{137}Cs and ^{90}Sr in the central north Pacific, *J. Oceanogr. Soc. Japan*, 40, 416-424.

Nagaya Y., and K. Nakamura (1981). Artificial radionuclides in the western northwest Pacific (II): ^{137}Cs and ^{90}Sr in deep waters, *J. Oceanogr. Soc. Japan*, 37, 135-144.

Nagaya Y., and K. Nakamura (1976). ^{90}Sr and ^{137}Cs contents in the surface waters of adjacent seas of Japan and North Pacific during 1969 and 1973, *J. Oceanogr. Soc. Japan*, 40, 228-234.

Nakamura K., and Y. Nagaya (1985). Accumulation of Cs-137 and Pu-239,240 in sediments of the coastal sea and the north Pacific. In: *Marine and Estuarine Geochemistry*, (editors, A.C. Sigleo and A. Hattori), Chapter 12, Lewis Pub., Chelsea, pp 171-180.

Nakamura K., and Y. Nagaya (1975). Accumulation of radionuclides in coastal sediment of Japan (II): Contents of fission products in some coastal sediments collected in 1966-72, *J. Radiat. Res.*, 16: 184-192.

Nakanishi T., Y. Shiba, M. Muramatsu, and M. Azizul Haque (1995). Estimation of mineral aerosol fluxes to the Pacific by using environmental plutonium as a tracer, In: *Biogeochemical Processes and Ocean Flux in the Western Pacific* (editors; H. Sakai and Y. Nozaki), Terra Scientific (TERRAPUB), Tokoyo, pp. 15-30.

Nakanishi T., M. Tajima, M. Senaga, M. Takei, A. Ishikawa, and K. Sakamoto (1984). Determination of $^{239,240}\text{Pu}$ in seawater, *Nucl. Instru. Meth. Phys. Res.*, 223: 239-242.

NCRP (1938). Radium Protection Report No. 4, National Council on Radiation Protection and Measurements, Bethesda, Maryland, U.S.A.

NCRP (1936). X-ray Protection Report No. 3, National Council on Radiation Protection and Measurements, Bethesda, Maryland, U.S.A.

NEA/OECD (1996). Coordinated Research and Environmental Surveillance Programme Related to Sea Disposal of Radioactive Waste, CRESO Final Report 1991-1995, Nuclear Energy Agency, Organization for Economic Cooperation and Development, Paris, 1996.

Newell R.W (1971). The global circulation of atmospheric pollutants, *Sci. Am.*, 224, 32-47.

Nikipelov B.V., and E. G. Drozhko (1990). Explosion in the South Ural Mountains, *PRIRODA*, May 1990, pp. 48-49.

- Nikipelov B.V., G.N. Romanov, L.A. Buldakov, N.S. Babev, Yu B. Kholina, and E.I. Mikerin (1990). A Radiation Accident in the Southern Urals in 1957, Original article submitted July 14, 1989, Plenum Publishing Co.
- Nilsen T., I. Kudrik, and A. Nitikin (1996). The Russian Northern Fleet: Sources of Radioactive Contamination. Bellona Foundation, Oslo, Norway, 18 April 1996. Internet address: <http://www.grida.no/ngo/bellona/lhome.htm>
- Noshkin V.E. and K.M Wong (1980). Plutonium in the North Equatorial Pacific, In: *Processes Determining the Input Behavior and Fate of Radionuclides and Trace Elements in Continental Shelf Environments*, U.S. DOE Conference Report 790382, pp. 11-28.
- Noshkin V.E., and K.M. Wong (1979). Plutonium mobilization from sedimentary sources to solution in the marine environment, In: *Proceedings of the Third NEA Seminar on Marine Radioecology*, Tokoyo, pp. 165-178.
- Noshkin V.E., and Bowen V.T. (1973). Concentrations and distributions of long-lived fallout radionuclides in open ocean sediments. In: *Radioactive Contamination of the Marine Environment*, International Atomic Energy Agency (IAEA), Vienna, Austria, pp. 671-686.
- NRPA (1996). Dumping of radioactive waste and investigation of radioactive contamination in the Kara Sea, Results from 3 years of investigation (1992-1994) in the Kara Sea. Joint Norwegian-Russian Expert Group for Investigation of Radioactive Contamination in the Northern Areas, Norwegian Radiation Protection Authority (NRPA), Østerås, Norway.
- Nuclear Fuel (1993). Tomsk-7 Environmental Pu Emission About 23 grams, Fact-Finder Says, p 13.
- Nuclear Waste News (1993). Tomsk-7 Caused Minimal Radiation Hazards, IAEA Team Reports, pp 157-158.
- Oi N. (1998). Plutonium Challenges: Changing dimensions of global cooperation, IAEA Bulletin, 40/1/1998, 12-16.
- OTA (1995). Spent Fuel and Waste Management, Presented-by the Russian Delegation to the Congressional Office of Technology Assessment Meeting, January 17-18, 1995, Office of Technology Assessment (OTA), Washington, D.C.
- Pentreath R.J. (1985). *General review of literature relevant to water discharges*, IAEA TECDOC-239, International Atomic Energy Agency, Vienna, Austria, pp. 19-66.
- Petersen K.R. (1970). An empirical model for estimating worldwide deposition from atmosphere nuclear detonations, *Health Phys.*, 18, 357-378.
- Pettersson H.B. L., K. Amano, V.I. Berezhnov, E. Chaykovskaya, V.B. Chumichev, C.K. Chung, J. Gastaud, K. Hirose, G.H. Hong, C.K. Kim, S.H. Kim, S.H. Lee, T. Morimoto, A. Nikitin, K. Oda, P.P. Povinec, E. Suzuki, A. Tkalin, O. Togawa, N.K. Veletova, Y. Volkov, and K. Yishida (1999). Anthropogenic radionuclides in sediments in the NW Pacific Ocean and its marginal seas: results of the 1994-1995

- Japanese-Korean-Russian expeditions, *Sci. Total Environ.*, 237/238, 213-224.
- Raisbeck G.M., F. Yiou, Z.Q. Zhou, L.R. Kilius, and H. Dahlgard (1993). Anthropogenic ^{129}I in the Kara Sea, In: *Environmental Radioactivity in the Arctic and Antarctic*, (editors; P. Strand and E. Holm), Norwegian Radiation Protection Authority, Østerås, Norway, pp. 125-128.
- Rhodes, R. (1986). *The Making of the Atomic Bomb*, Simon and Schuster, New York, 1986.
- Robison W.L., V.E. Noshkin, T.F. Hamilton, C.L. Conrado, and K. Bogen (2001). *An Assessment of Current Day Impact of Various Materials Associated with the U.S. Nuclear Test Program in the Marshall Islands*, UCRL-LR-143980, Lawrence Livermore National Laboratory, 21 pp.
- Roether W. (1974). The tritium and carbon-14 profiles at the GEOSECS I (1969) and GOGO I (1971) North Pacific stations, *Earth Planet. Sci. Lett.*, 23, 108-115.
- Sayles F.L., H.D. Livingston, and G.P. Panteleyev (1997). The history and source of particulate ^{137}Cs and $^{239+240}\text{Pu}$ deposition in sediments of the Ob River delta, Siberia. *Sci. Total Environ.*, 202, 25-71.
- Schell, W.R., F.G. Lowman, and R.P. Marshall (1980). Geochemistry of Transuranic Elements at Bikini Atoll, In: *Transuranic Elements in the Environment* (editor, W.C. Hanson), U.S. Department of Energy, pp 541-577.
- Scott M.R., P.F. Salter, and J.E. Halverson (1983). Transport and deposition of plutonium in the ocean: Evidence from Gulf of Mexico sediments, *Earth Planet. Sci. Lett*, 63, 202-222.
- SCRF(1995). *Problems of Arctic Contamination*, materials prepared by a working group headed by Deputy Minister A.I. Volgin of the Ministry of Affairs for National and Regional Policy, Security Council of the Russian Federation, Moscow, Russia, 1995.
- Seaborg G.T., and W.D. Loveland (1990). *The Elements Beyond Uranium*, John Wiley & Sons, Inc., New York, 359 pp.
- Sivintsev, Y. (1994a). Study of the nuclide composition and characteristics of the fuel in dumped submarine reactors and icebreaker "Lenin". Part 1 - Atomic icebreaker, Working materials of the International Arctic Seas Assessment Project, IAEA-IASAP-1, International Atomic Energy Agency, Vienna.
- Sivintsev, Y. (1994b). Study of the nuclide composition and characteristics of the fuel in dumped submarine reactors and icebreaker "Lenin". Part 2 - Nuclear submarines, Working materials of the International Arctic Seas Assessment Project, IAEA-IASAP-5, International Atomic Energy Agency, Vienna.
- Smith J.N., K.M. Ellis, L. Ployak, G. Ivanov, S.L. Forman, and S.B. Moran (2000). $^{239,240}\text{Pu}$ transport into the Arctic Ocean from underwater nuclear tests in Chernaya Bay, Novaya Zemlya., *Continent. Shelf Res.*, 20, 255-279.

- Smith J.N., K.M. Ellis, K. Naes, S. Dahle, and D. Matishov (1995). Sedimentation and mixing rates of fallout radionuclides in Barents Sea sediments off Novaya Zemlya, *Deep-Sea Res. II*, 42, 1471-1493.
- Smith J.N., K.M. Ellis, A. Aarkrog, H. Dahlgard, and E. Holm (1994). Sediment mixing and burial of the $^{239,240}\text{Pu}$ from the 1968 Thule, Greenland nuclear weapons accident. *J. Environ. Radioactivity*, 25, 135-159.
- Smith J.N., K.M. Ellis, and E.P. Jones (1990). Cesium-137 transport into the Arctic Ocean through Fran Strait, *J. Geophys. Res.*, 95(C2), 1693-1701.
- Strand P., and T. Jolle (1999). *Environmental Radioactivity in the Arctic*, Scientific Committee of the Environmental Radioactivity in the Arctic, Norwegian Radiation Protection Authority, Østerås, Norway, 353 pp.
- Strand P., and A. Cooke (1995). *Environmental Radioactivity in the Arctic*, Scientific Committee of the Environmental Radioactivity in the Arctic, Norwegian Radiation Protection Authority, Østerås, Norway, 415 pp.
- Tass World Service (1992). Two Industrial Nuclear Reactors Scheduled for Closure, April 20, 1992.
- UNSCEAR (2000). Sources and Effects of Ionizing Radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, 2000 Report to the General Assembly, with Scientific Annexes, Volume 1: Sources, United Nations Publication E.00.IX.3, United Nations, New York, 2000.
- UNSCEAR (1993). Sources and Effects of Ionizing Radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, 1993 Report to the General Assembly, with Scientific Annexes, United Nations Publication No. E.94.IX.82, United Nations, New York, 1993.
- Vakulovsky S.M. (1993). Transport of Artificial Radioactivity by the Ob to the Arctic Seas, In: *Environmental Radioactivity in the Arctic and Antarctic*, (editors; P. Strand and E. Holm), Norwegian Radiation Protection Authority, Østerås, Norway, August 1993.
- Vakulovsky S.M., S. Nikitin, and V. Chumichev (1993). Radioactive Contamination of the Barents and Kara Seas, International Conference on Environmental Radioactivity in the Arctic, Woods Hole Oceanographic Institution, Woods Hole, MA, USA, June 7-9, 1993.
- Volchok H.L., V.T. Bowen, T.R. Folsom, W.S. Broecker, E.A. Schuert, and G.S. Bien (1971). Oceanic distributions of radionuclides from nuclear explosions, sources of radioactivity and their characteristics. In: *Radioactivity in the Marine Environment*, National Academy of Sciences, Washington, pp. 42-89.
- Vorobiova M.I., M.O. Degteva, D.S. Burmistrov, N.G. Safronova, V.P. Kozheurov, L.R. Anspaugh, and B.A. Napier (1999). Review of historical monitoring data on Techa Review contamination, *Health ys.*, 76, 605-618.

Yefimov E. (1994). Radionuclide composition, characteristics of shielding barriers and analysis of weak points of the dumped reactors of submarine N. 601, Working materials of the International Arctic Seas Assessment Project, IAEA-IASAP-6, International Atomic Energy Agency, Vienna.

Yiou F., G.M. Raisbeck, Z.Q. Zhou, L.R. Killius, and P.J. Kershaw (1995). Improved estimates of oceanic discharges of ^{129}I from Sellafield and La Hague, (editors; P. Strand and A. Cooke), Norwegian Radiation Protection Authority, Østerås, Norway, pp. 113-116. Weiss, W. and W. Roether (1980). The rates of tritium input to the world oceans, *Earth Planet. Sci. Lett.*, 49 , 435-446.

White Book (1993). Facts and problems related to radioactive waste disposal in sea adjacent to the territory of the Russian Federation, Materials for a report by the Government Commission on matters related to radioactive waste disposal at sea, Created by Decree No. 613 of the Russian Federation President of October 24, 1994, (editors; A.V. Yablokov, V.K. Karasev, V.M. Rumyantsev, M.E. Kokeev and O.J. Petrov), White Book No. 3., Small World Publisher, Moscow, 108 pp.