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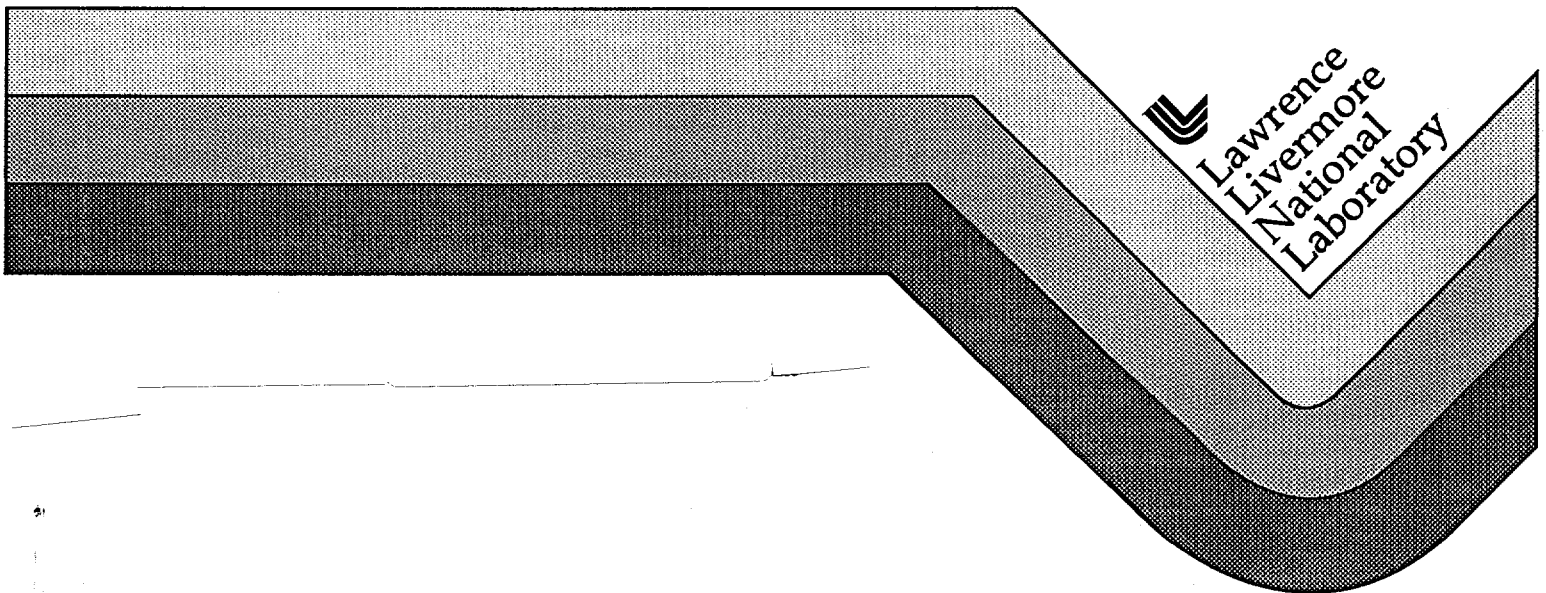
**Predicted Radionuclide Release from Reactor-related
Unenclosed Solid Objects Dumped in the Sea of Japan and
the Pacific Ocean, East Coast of Kamchatka**

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Predicted Radionuclide Release from Reactor-related Unenclosed Solid Objects Dumped in the Sea of Japan and the Pacific Ocean, East Coast of Kamchatka

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Abstract

Between 1978 and 1991 reactor-related solid radioactive waste was dumped by the former Soviet Union as unenclosed objects in the Pacific Ocean, east coast of Kamchatka, and the Sea of Japan. This paper presents estimates for the current (1994) inventory of activation and corrosion products contained in the reactor-related unenclosed solid objects. In addition, simple models derived for prediction of radionuclide release from marine reactors dumped in the Kara Sea are applied to certain of the dumped objects to provide estimates of radionuclide release to the Pacific Ocean, east coast of Kamchatka, and Sea of Japan environments. For the Pacific Ocean, east coast of Kamchatka, total release rates start below 0.01 GBq yr^{-1} and over 1,000 years, fall to 100 Bq yr^{-1} . In the Sea of Japan, the total release rate starts just above 1 GBq yr^{-1} , dropping off to a level less than 0.1 GBq yr^{-1} , extending past the year 4,000.

Background

In the Spring of 1993, the Russian report, *Facts and Problems Related to Radioactive Waste Disposal in Seas Adjacent to the Territory of the Russian Federation* (Yablokov et al. 1993), was released. The findings presented in that report were the result of a scientific study commissioned in October 1992 by the Office of the President of the Russian Federation. Related to the seas adjacent to the Russian Far East, the White Book, as the report was later called, reported on disposal operations at ten separate sites: six in the Sea of Japan (designated as Areas 1, 2, 5, 6, 9, and 10), one in the Sea of Okhotsk (designated as Area 3), and three in the Pacific Ocean, east coast of Kamchatka (designated as Areas 4, 7, and 8). The reported characteristics of the liquid radioactive waste dumped in the seas adjacent to the Russian Far East were

- (1) disposal depths of 1.1 to 3.7 km,
- (2) disposal dates between 1966 and 1992,
- (3) a total activity at the time of disposal of 457 TBq of unspecified origin, and
- (4) a total volume of $118,694 \text{ m}^3$

Work performed under the auspices of the US Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-ENG-48 and the US Navy, Office of Naval Research.

while those of the solid radioactive waste were

- (1) disposal depths of 1.9 to 3.7 km,
- (2) disposal dates between 1968 and 1992,
- (3) a total activity at the time of disposal of 279 TBq of unspecified origin, and
- (4) a total volume of 21,306 m³ within
 - (a) 6,834 containers,
 - (b) 37 ships, and
 - (c) 104 unenclosed objects.

In addition to not identifying the disposed radionuclides, there was also no estimate provided for the current levels of radioactivity or radionuclide release to the environment.

Radionuclide Inventory in Reactor-related Unenclosed Solid Objects

Of the liquid and solid radioactive waste dumped in the seas adjacent to the Russian Far East, only the reactor-related unenclosed solid objects allow for prediction of the current (1994) levels of radioactivity or radionuclide release to the environment.

Reactor-related parts, such as primary loop circulating pumps, steam generators (SGs), a reactor core plate, reactor pressure vessels (RPVs), and a reactor lid, were reportedly dumped as unenclosed objects in either the Pacific Ocean, east coast of Kamchatka (Area 8), or the Sea of Japan (Areas 9 and 10) (Yablokov et al. 1993). Table 1 summarizes the pertinent disposal information for the reactor-related unenclosed solid objects.

In the case of the reactor core plate, RPVs, and reactor lid, the radionuclides contained in each result from neutron activation of the stainless steel or low alloy steel from which they are constructed. For the primary loop circulating pumps and SGs, the radionuclides contained result from primary system corrosion. Whether activation or corrosion product, the long-lived radionuclides of consequence are the same: ¹⁴C, ⁶⁰Co, ⁵⁹Ni, and ⁶³Ni. The difference is in the relative quantity of each.

Using information reported for the activation product inventories in the submarine pressurized water reactors (PWRs) without spent nuclear fuel (SNF) that were dumped in the Kara Sea (Sivintsev 1994), an estimate can be made for the activation product inventories in the reactor core plate, RPVs, and reactor lid. Assuming that each reactor-related object was dumped one year after reactor shutdown, the inventory can be estimated from the product of the total reported activity at time of disposal and the average fraction of ¹⁴C, ⁶⁰Co, ⁵⁹Ni, and ⁶³Ni contained in the submarine PWRs without SNF at one year after shutdown. Simple radioactive decay then provides the inventory in 1994.

In a similar manner, an estimate can also be made for the corrosion product inventories in the primary loop circulating pumps and SGs. With one exception, the procedure is exactly the same

as that for the activation products. The difference is the source of the fraction of ^{14}C , ^{60}Co , ^{59}Ni , and ^{63}Ni contained in the corrosion products: a British calculation for a generic nuclear submarine one year after shutdown (House of Commons Defense Committee 1990).

Table 1. Pertinent disposal information for the reactor-related unenclosed solid radioactive waste dumped in the Sea of Japan and the Pacific Ocean, east coast of Kamchatka, as presented in the White Book (Yablokov et al. 1993).

Site designation ¹	Year of disposal ¹	Dumped unit ¹	Disposal coordinates ¹	Disposal depth (km) ²	Activity at disposal (TBq) ¹
Area 8	1986	Primary loop circulating pump (50 pieces)	52° 31' N 159° 8' E	2.0 - 2.57	1.4
	1988	Steam generator (10 pieces)	52° 30' N 159° 9' E	2.0 - 2.57	2.2
	1989	Submarine core plate	52° 30' N 159° 9' E	2.0 - 2.57	2.6
		Primary loop circulating pump (50 pieces)	52° 30' N 159° 9' E	2.0 - 2.57	0.031
Area 9	1991	Steam generator (5 pieces)	41° 40' N 134° 0' E	3.25 - 3.7	0.24
Area 10	1978	Two submarine reactors	41° 10' N 131° 15' E	2.9 - 3.3	1.7
	1983	Reactor lid (8 pieces)	41° 40' N 131° 26' E	2.9 - 3.3	3.4
Total					11.6

¹ Information is that presented in the White Book (Yablokov et al. 1993).

² The exact depth of disposal was not specified. The depth presented is that associated with the area designation in the White Book.

Table 2 presents the estimated 1994 activity of long-lived activation products in the dumped reactor-related unenclosed solid objects. Overall, about 4.6 TBq of radioactivity were contained in the reactor-related unenclosed solid objects in 1994, with ^{60}Co and ^{63}Ni constituting 77% and 21% of the total, respectively. Furthermore, the reactor core plate in Area 8 and the reactor lid in Area 10 were the two greatest sources of this radioactivity.

Radionuclide Release to the Environment from Selected Solid Objects

In the case of the reactor-related unenclosed solid objects, release of the contained radionuclide inventory occurs through corrosion of the construction materials. Simple models derived for prediction of radionuclide release from marine reactors dumped in the Kara Sea (International Atomic Energy Agency in preparation) can be applied to the reactor core plate, RPVs, and reactor lid to provide estimates of radionuclide release to the Pacific Ocean, east coast of Kamchatka, and Sea of Japan environments. While some question remains as to whether the reactor core

plate and RPVs were actually dumped unenclosed or encased in a container, for the purposes of this estimate, they are assumed to be dumped unenclosed.

Table 2. Estimated 1994 activity of long-lived activation products in the reactor-related unenclosed solid radioactive waste dumped in the Sea of Japan and the Pacific Ocean, east coast of Kamchatka.

Site designation ¹	Dumped unit ¹	Activity in 1994 (GBq) ²			
		¹⁴ C	⁶⁰ Co	⁵⁹ Ni	⁶³ Ni
Area 8	Primary loop circulating pump (50 pieces)	0.00021	500	0.018	3.3
	Steam generator (10 pieces)	0.00031	990	0.028	5.0
	Submarine core plate	0.90	1,000	29	330
	Primary loop circulating pump (50 pieces)	0.0000045	16	0.00040	0.073
Area 9	Steam generator (5 pieces)	0.000035	160	0.0030	0.56
Area 10	Two submarine reactors	0.59	180	19	200
	Reactor lid (8 pieces)	1.2	690	39	420
Total		2.7	3,540	87	959

¹ Information is that presented in the White Book (Yablokov et al. 1993).

² Inventory is based on the total activity at the time of disposal as presented in the White Book.

Pacific Ocean, East Coast of Kamchatka - Submarine Reactor Core Plate

Modelling Strategy

For the submarine reactor core plate dumped in Area 8, it is assumed that it lies flat on the seabed at a depth of 2.0 km. Under these conditions, corrosion of stainless steel is thought to be very slow. There is a wide spread of experimental results for bulk corrosion rates on the deep sea floor. Rates of 0.0005 mils yr⁻¹ for a plate buried in the sediment and 0.0009 mils yr⁻¹ for a plate exposed to free flowing seawater (an average of 0.000018 mm yr⁻¹) are reported (Barth et al. 1989). A later review of all available data for the International Arctic Seas Assessment Project (IASAP) (Heiser et al. 1995), suggests 0.007 mil yr⁻¹, a factor of ten higher at 0.00018 mm yr⁻¹, which will be used for this calculation.

Using the scenarios and methodology of the IASAP models, the activation products are assumed to be released uniformly at a rate proportional to the thickness of steel corroded per year as a fraction of the total thickness of the object. This fraction is multiplied by the inventory of the individual isotopes remaining at time t, to give a release rate to the seawater; in this study, the units are given in GBq yr⁻¹.

From generic engineering drawings of former Soviet Union submarine PWRs, the core plate is estimated to be 90-mm thick. Corroding the core plate on both sides, which is a pessimistic assumption, the material will require 250,000 years to disintegrate. As this is a very unrealistic scenario, it is assumed that the plate will slowly cover with sediment and over a period of 1,000 years, the release rate for the activation products will decrease from a maximum to zero, as the sediment builds up. Since sedimentation types and rates and the mobility of ions through the covering material are unknown, this must represent an estimate at best.

Results

The results of the modelling strategy are illustrated in Figure 1 for the four radionuclides in the activation product inventory: ^{14}C , ^{60}Co , ^{59}Ni , and ^{63}Ni . The relatively short half-life of ^{60}Co shows a steep fall-off, followed by ^{63}Ni ; only the two long-lived isotopes of ^{59}Ni and ^{14}C remain until the end of the next millennia. Figure 2 shows the total rate of release for the four isotopes added together. Total release rates start below 0.01 GBq yr^{-1} and over the 1,000 years, fall to 100 Bq yr^{-1} .

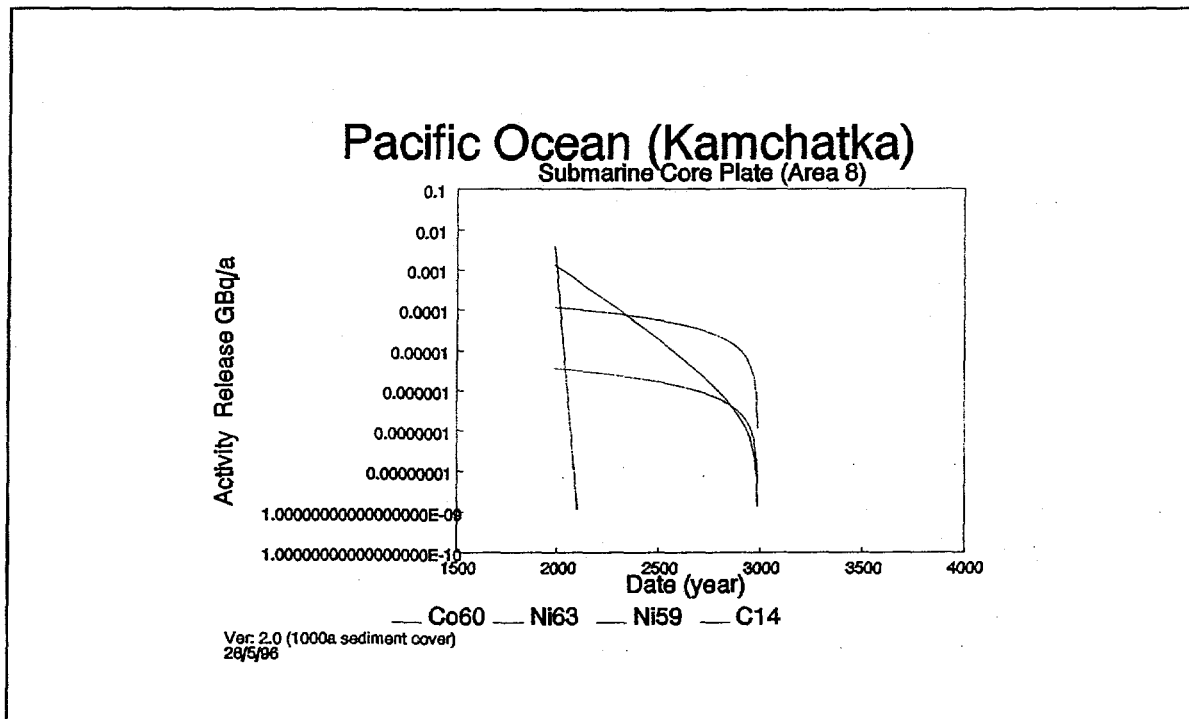


Figure 1. Release rates (GBq yr⁻¹) for individual activation products from the submarine reactor core plate dumped in the Pacific Ocean, east coast of Kamchatka (Area 8).

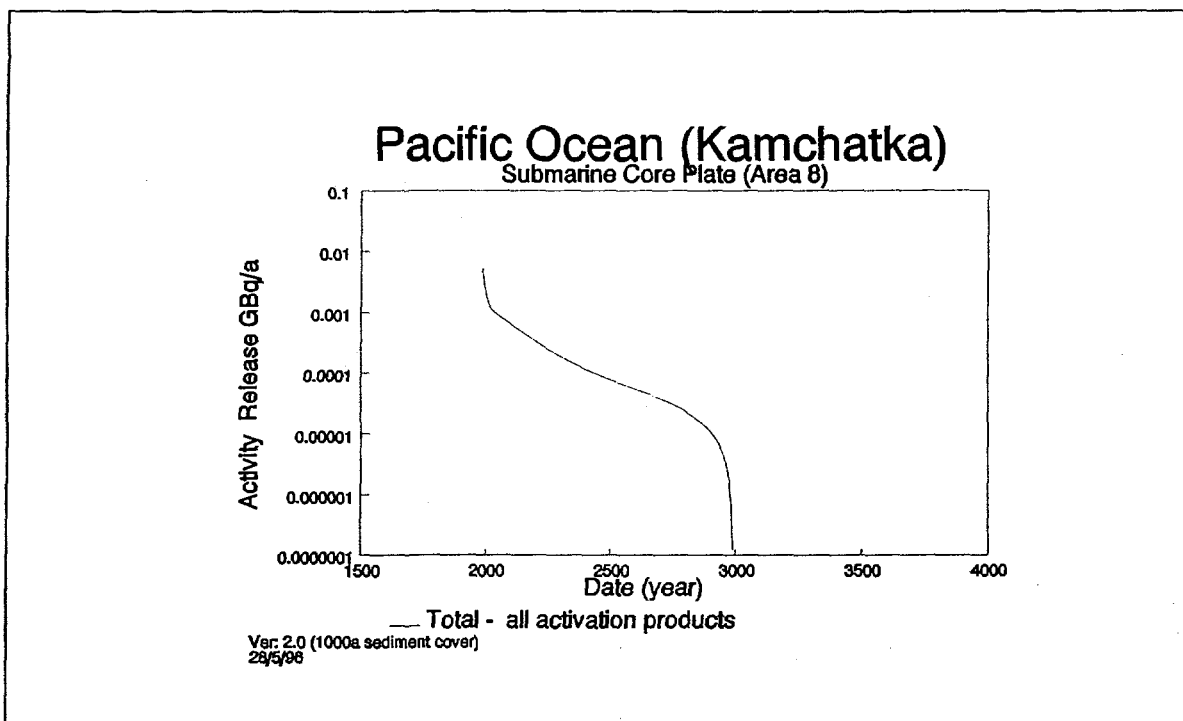


Figure 2. Total release rate (GBq yr⁻¹) of all activation products from the submarine reactor core plate dumped in the Pacific Ocean, east coast of Kamchatka (Area 8).

Sea of Japan - Two Submarine Reactors and A Reactor Lid

Modelling Strategy

For the reactor lid dumped in Area 10, the same philosophy was used for modelling the release rate as that described above for the reactor core plate. The lid was estimated to be 390-mm thick, and although it was divided into eight pieces, the surface area will remain the same and the pieces are assumed to lie flat on the seabed, at a depth of 3.0 km. A bulk corrosion rate of 0.05 mm yr⁻¹ was used from Heiser et al. (1995), which takes into account the sediment interface. Again, the release rate was assumed to be a maximum at the time of dumping, decreasing to zero in 1,000 years, due to sediment accumulation.

The two defueled submarine reactors dumped in Area 10 were treated exactly as in the IASAP models. The dimensions estimated for each RPV were a (1) 100-mm thick wall, (2) 5-mm thick wall clad, (3) 390-mm thick lid, and (4) 14.5-mm thickness for each of five radial thermal shields. Corrosion takes place on the outside surface of the RPV from the moment of dumping. Corrosion of the interior and release of interior activation products from the stainless steel RPV clad and the thermal shields is inhibited due to the lack of oxygen exchange with the outside

seawater. A small breather hole allows initial penetration, followed by the loss of the caps covering the main RPV tube penetrations, increasing the corrosion rates and activity release.

The reactors are assumed to sit on the seabed, largely above the seawater/sediment interface. It was not thought applicable to cover these objects with sediments. Thus, the blanketing effect used on the reactor lid was not applied.

The total release rates for the two reactors and the reactor lid were added together, taking into account their different dates of dumping.

Results

The results for the modelling strategy are illustrated in Figures 3 and 4. In Figure 4, the total release rate starts just above 1 GBq yr⁻¹, dropping off to a level less than 0.1 GBq yr⁻¹, extending past the year 4,000. Beyond this date, prediction would be unrealistic.

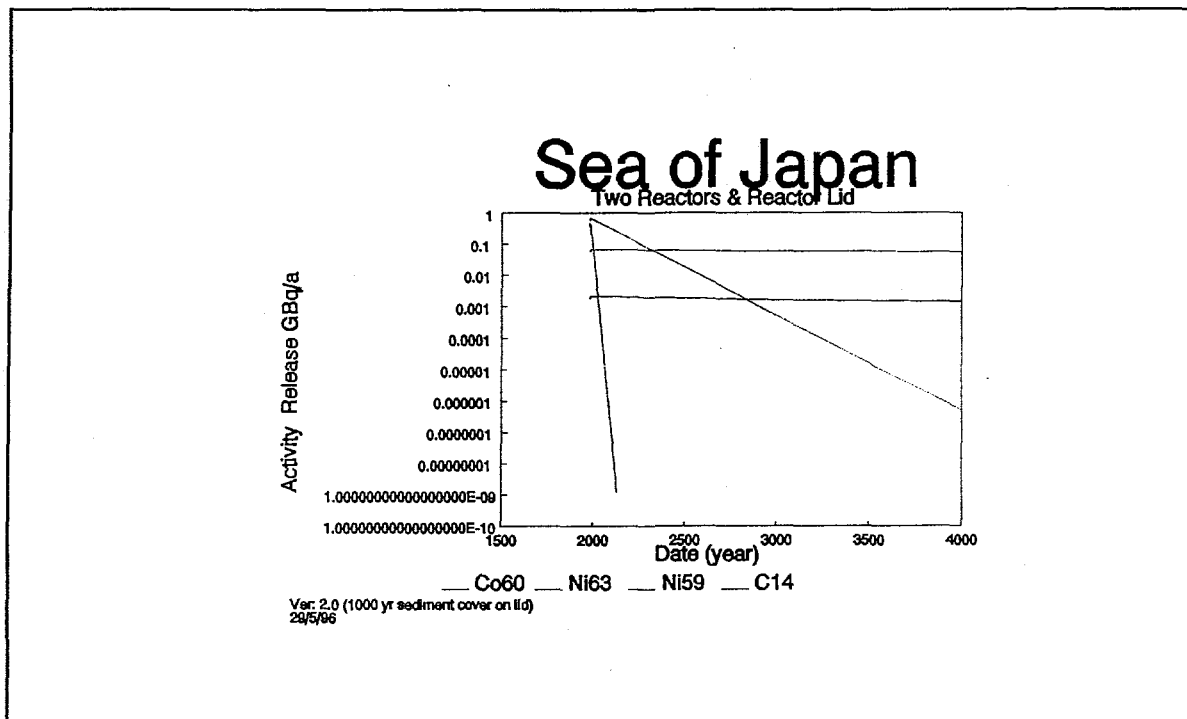


Figure 3. Release rates (GBq yr⁻¹) for individual activation products, from the two submarine reactors and reactor lid dumped in the Sea of Japan (Area 10).

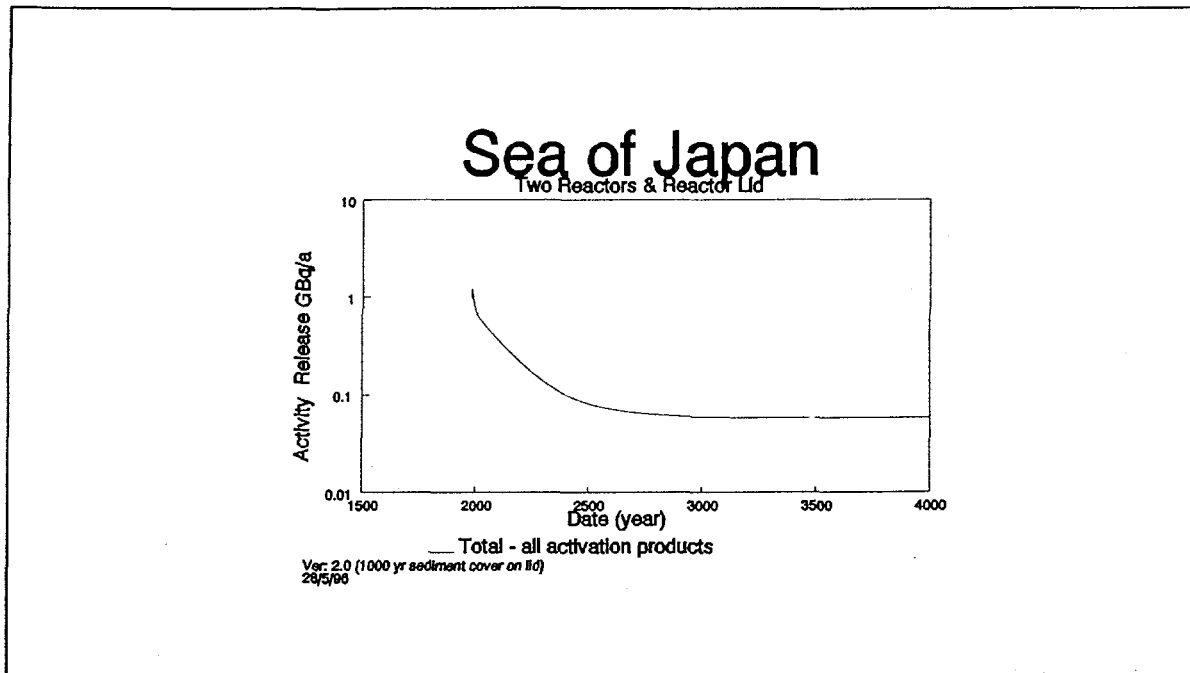


Figure 4. Total release rate (GBq yr⁻¹) of all activation products from the two submarine reactors and reactor lid dumped in the Sea of Japan (Area 10).

References

- Barth, C. H. and Sheldon, R. B. *Corrosion Rates of Structural Materials on the Ocean Floor*, General Electric Co., Schenectady, NY, KAPL-4701 (1989).
- Heiser, J. H. and Soo, P. "Corrosion of Barrier Materials in Seawater Environments," Department of Advanced Technology, Brookhaven National Laboratory, Upton, NY (1995).
- House of Commons Defense Committee. "Decommissioning of Nuclear Submarines," 7th Report, Session 1988-1989, Her Majesty's Stationary Office, London, England (1990).
- International Atomic Energy Agency. *Predicted Radionuclide Release from Marine Reactors Dumped in the Kara Sea*, Technical Report Series No. XXX, IAEA, Vienna, Austria (in preparation).
- Sivintsev, Y. *Study of Nuclides Composition and Characteristics of Fuel in Dumped Submarine Reactors and Atomic Icebreaker "Lenin": Part 2 - Nuclear Submarines*, Russian Research Center "Kurchatov Institute," Moscow, Russia, Report No. 31/7281 (August 1994).

Yablokov, A. V., Karasev, V. K., Rumyantsev, V. M., Kokeyev, M. Y., Petrov, O. I., Lystsov, V. N., Yemelyanenko, A. F., and Rubtsov, P. M. *Facts and Problems Related to Radioactive Waste Disposal in Seas Adjacent to the Territory of the Russian Federation*, Office of the President of the Russian Federation, Moscow, Russia (1993), pp. 29-32, 41-43, and 66-72.

**PREDICTED RADIONUCLIDE RELEASE FROM
REACTOR-RELATED UNENCLOSED SOLID OBJECTS
DUMPED IN THE SEA OF JAPAN AND THE PACIFIC
OCEAN, EAST COAST OF KAMCHATKA**

**GLOBAL PARTNERSHIP: A MULTI-NATIONAL WORKSHOP ON
NUCLEAR WASTE IN AND AROUND THE SEA OF JAPAN, SEA OF
OKHOTSK, AND THE NORTH PACIFIC OCEAN**

**June 14-15, 1996
Niigata, Japan**

**Mark E. Mount
Fission Energy and Systems Safety Program**



Yablokov Commission Report



- **"Facts and Problems Related to Radioactive Waste Disposal in Seas Adjacent to the Territory of the Russian Federation," later called White Book, released in Spring of 1993.**
 - **Disposal of reactor-related unenclosed solid radioactive waste at three separate sites in seas adjacent to Russian Far East.**
 - **One in Pacific Ocean, east coast of Kamchatka (Area 8).**
 - **Two in Sea of Japan (Areas 9 and 10).**

Work performed under the auspices of the US Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-ENG-48 and the US Navy, Office of Naval Research.

Pertinent Disposal Information for Reactor-related Unenclosed Solid Radioactive Waste (White Book)



Site designation ¹	Year of disposal ¹	Dumped unit ¹	Disposal coordinates ¹	Disposal depth (km) ²	Activity at disposal (TBq) ¹
Area 8	1986	Primary loop circulating pump (50 pieces)	52° 31' N 159° 8' E	2.0 - 2.57	1.4
	1988	Steam generator (10 pieces)	52° 30' N 159° 9' E	2.0 - 2.57	2.2
	1989	Submarine core plate	52° 30' N 159° 9' E	2.0 - 2.57	2.6
		Primary loop circulating pump (50 pieces)	52° 30' N 159° 9' E	2.0 - 2.57	0.031
Area 9	1991	Steam generator (5 pieces)	41° 40' N 134° 0' E	3.25 - 3.7	0.24
Area 10	1978	Two submarine reactors	41° 10' N 131° 15' E	2.9 - 3.3	1.7
	1983	Reactor lid (8 pieces)	41° 40' N 131° 26' E	2.9 - 3.3	3.4
Total					11.6

¹ Information is that presented in White Book (Yablokov et al. 1993).

² Exact depth of disposal not specified. Depth presented is that associated with area designation in White Book.

Inventory Estimate Methodology



- Submarine reactor core plate, reactor pressure vessel (RPV), and reactor lid inventories result from neutron activation of construction materials.
 - Long-lived radionuclides of consequence are ^{14}C , ^{60}Co , ^{59}Ni , and ^{63}Ni .
 - Inventory based on product of total reported activity at time of disposal and average fraction of each radionuclide contained in former Soviet Union submarine pressurized water reactors one year after shutdown (International Arctic Seas Assessment Project [IASAP] data).
- Primary loop circulating pump and steam generator inventories result from primary system corrosion.
 - Long-lived radionuclides of consequence are ^{14}C , ^{60}Co , ^{59}Ni , and ^{63}Ni .
 - Inventory based on product of total reported activity at time of disposal and average fraction of each radionuclide contained in generic British nuclear submarine one year after shutdown.

Estimated 1994 Activity of Activation Products in Reactor-related Unenclosed Solid Radioactive Waste



Site designation ¹	Dumped unit ¹	Activity in 1994 (GBq) ²			
		¹⁴ C	⁶⁰ Co	⁵⁹ Ni	⁶³ Ni
Area 8	Primary loop circulating pump (50 pieces)	0.00021	500	0.018	3.3
	Steam generator (10 pieces)	0.00031	990	0.028	5.0
	Submarine core plate	0.90	1,000	29	330
	Primary loop circulating pump (50 pieces)	0.000004 5	16	0.0004 0	0.073
Area 9	Steam generator (5 pieces)	0.000035	160	0.0030	0.56
Area 10	Two submarine reactors	0.59	180	19	200
	Reactor lid (8 pieces)	1.2	690	39	420
Total		2.7	3,540	87	959

¹ Information is that presented in White Book (Yablokov et al. 1993).

² Inventory is based on total activity at time of disposal as presented in White Book.

Release Rate Estimate Methodology



- Limited to selected reactor-related unenclosed solid objects.
 - Submarine reactor core plate.
 - Estimated thickness of 90 mm.
 - IASAP estimated bulk corrosion rate of 0.00018 mm yr⁻¹.
 - Release rate proportional to thickness of steel corroded per year as a fraction of total object thickness.
 - Object slowly covered with sediment over period of 1,000 years with release rate decreasing from a maximum to zero.

Release Rate Estimate Methodology (continued)



- Two submarine reactors (IASAP model).
 - Estimated RPV wall thickness of 100 mm.
 - Estimated RPV wall clad thickness of 5 mm.
 - Estimated total thermal shield thickness of 72.5 mm.
 - Corrosion takes place on outside surface of RPV from moment of disposal.
 - Corrosion of interior and release of interior activation products inhibited due to lack of oxygen exchange with outside seawater.
 - Small breather holes at top of control rod drive channels allow initial penetration of seawater.
 - Loss of caps covering main RPV tube penetrations increases corrosion rates and activity release.

Release Rate Estimate Methodology (continued)



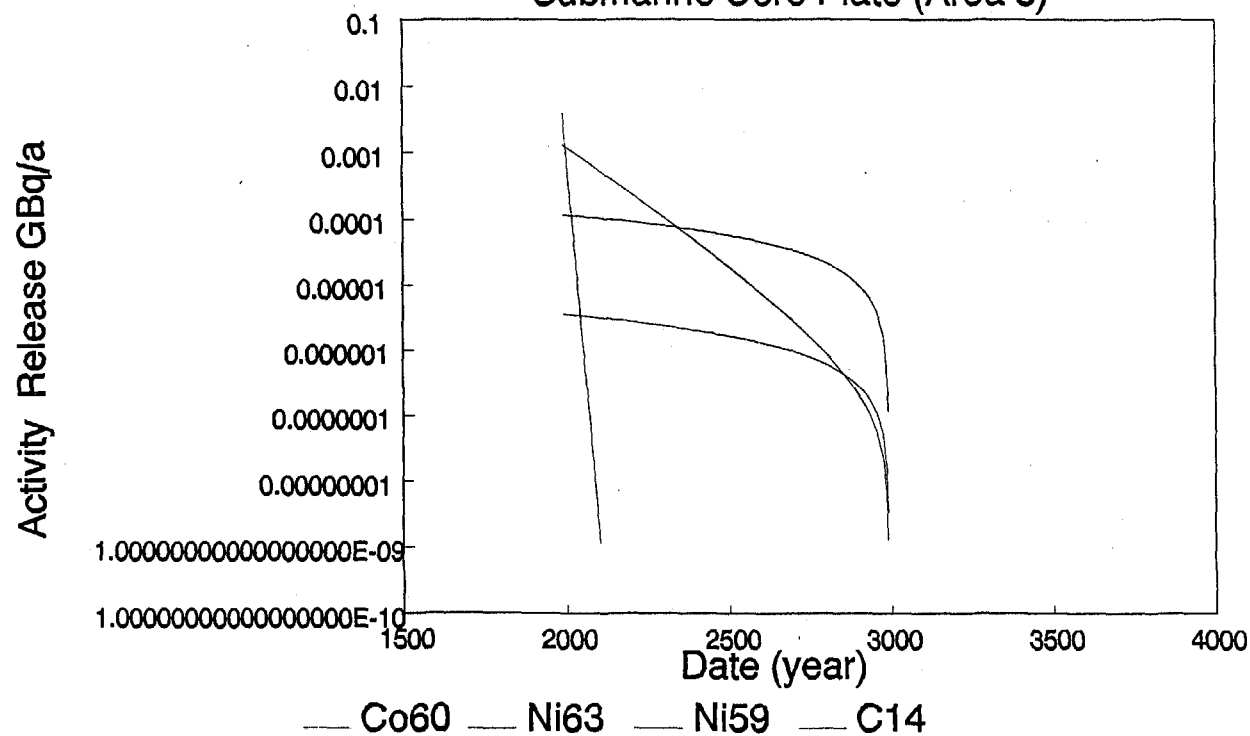
- Reactor lid.
 - Estimated thickness of 390 mm.
 - IASAP estimated bulk corrosion rate of 0.05 mm yr^{-1} .
 - Release rate proportional to thickness of steel corroded per year as a fraction of total object thickness.
 - Object slowly covered with sediment over period of 1,000 years with release rate decreasing from a maximum to zero.

Individual Radionuclide Release Rates (GBq yr⁻¹) from Reactor-related Unenclosed Solid Radioactive Waste



Source: One submarine reactor core plate

Pacific Ocean (Kamchatka) Submarine Core Plate (Area 8)



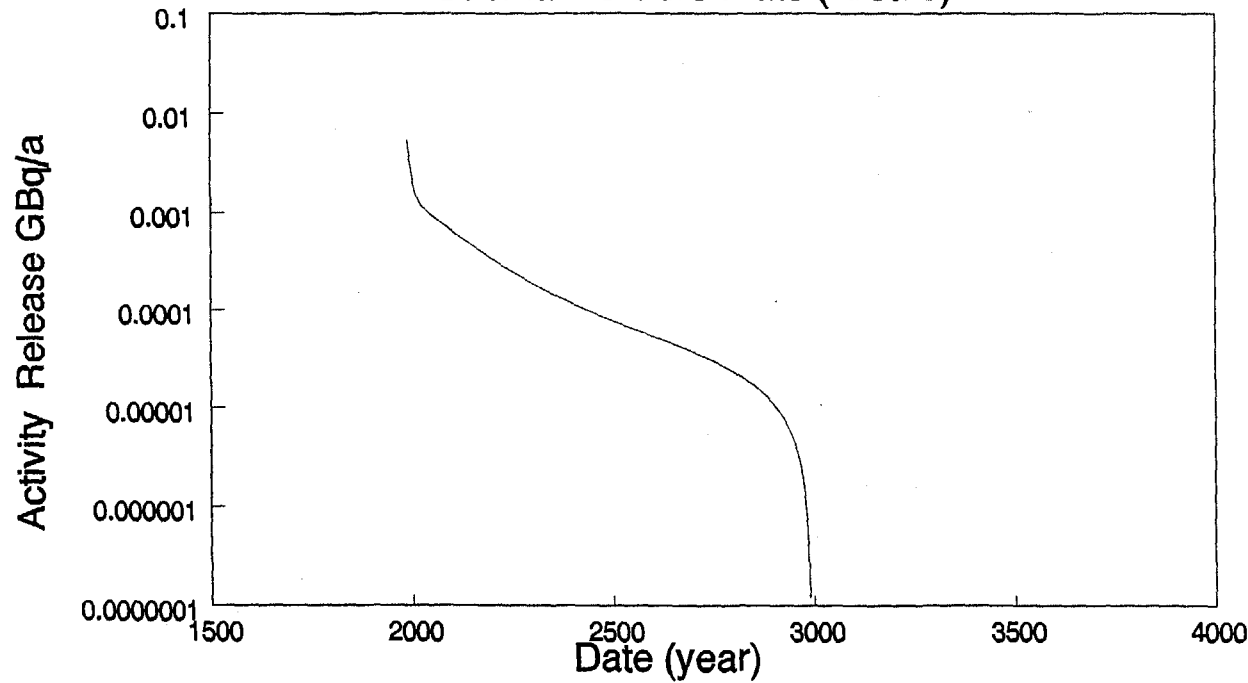
Ver: 2.0 (1000a sediment cover)
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Total Radionuclide Release Rate (GBq yr⁻¹) from Reactor-related Unenclosed Solid Radioactive Waste



Source: One submarine reactor core plate

Pacific Ocean (Kamchatka) Submarine Core Plate (Area 8)



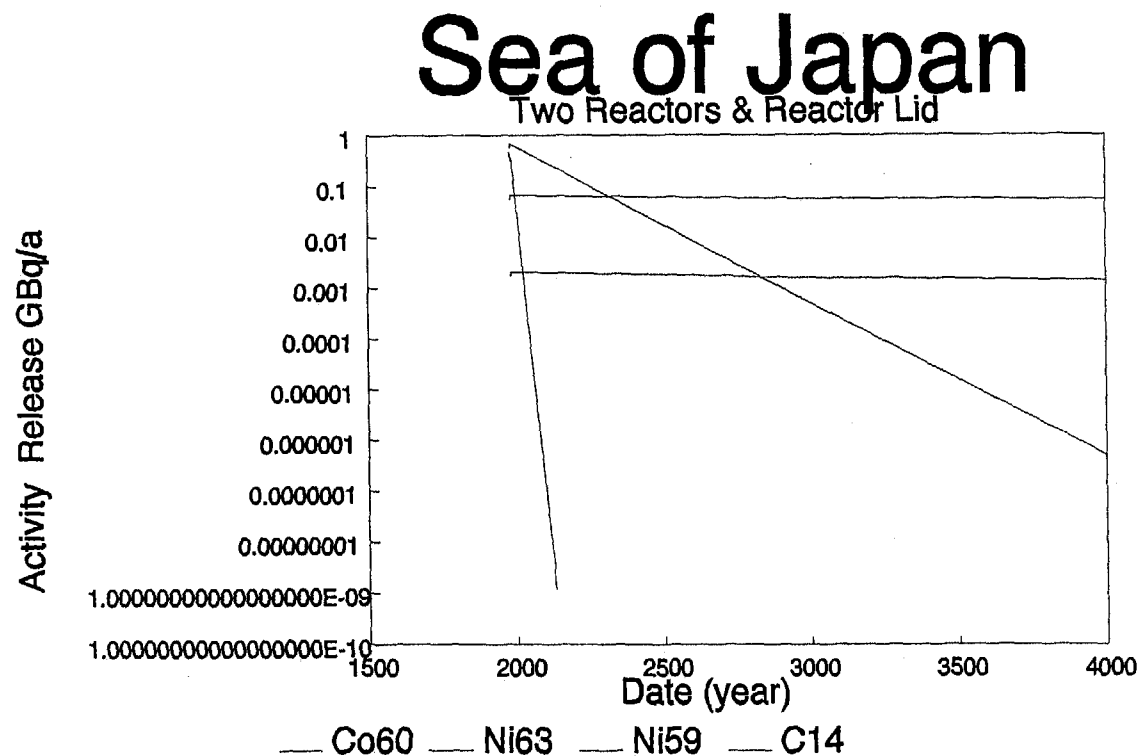
— Total - all activation products

Ver: 2.0 (1000a sediment cover)
28/5/96

Individual Radionuclide Release Rates (GBq yr⁻¹) from Reactor-related Unenclosed Solid Radioactive Waste



Source: Two submarine reactors and one reactor lid



Ver: 2.0 (1000 yr sediment cover on lid)
29/5/96

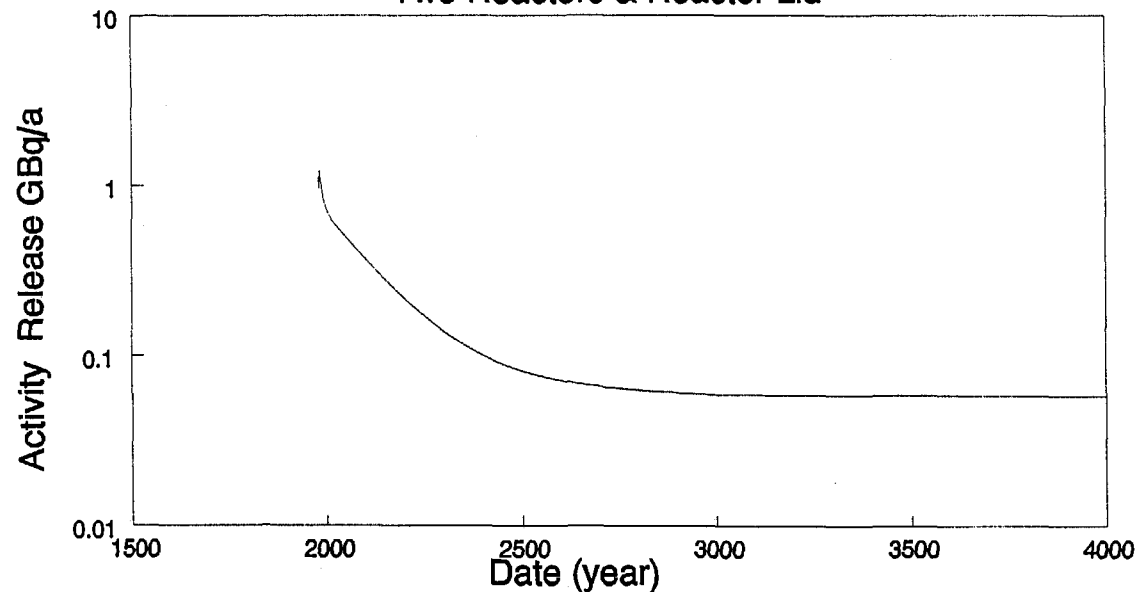
Total Radionuclide Release Rate (GBq yr⁻¹) from Reactor-related Unenclosed Solid Radioactive Waste



Source: Two submarine reactors and one reactor lid

Sea of Japan

Two Reactors & Reactor Lid



— Total - all activation products

Ver: 2.0 (1000 yr sediment cover on lid)
28/5/96

Release Rate Conclusions



- Pacific Ocean, east coast of Kamchatka (Area 8).
 - Total release rates start below 0.01 GBq yr^{-1} and over 1,000 years, fall to 100 Bq yr^{-1} .

- Sea of Japan (Area 10).
 - Total release rate starts just above 1 GBq yr^{-1} , dropping off to a level less than 0.1 GBq yr^{-1} , extending past the year 4,000.