

## **1 History and methods**

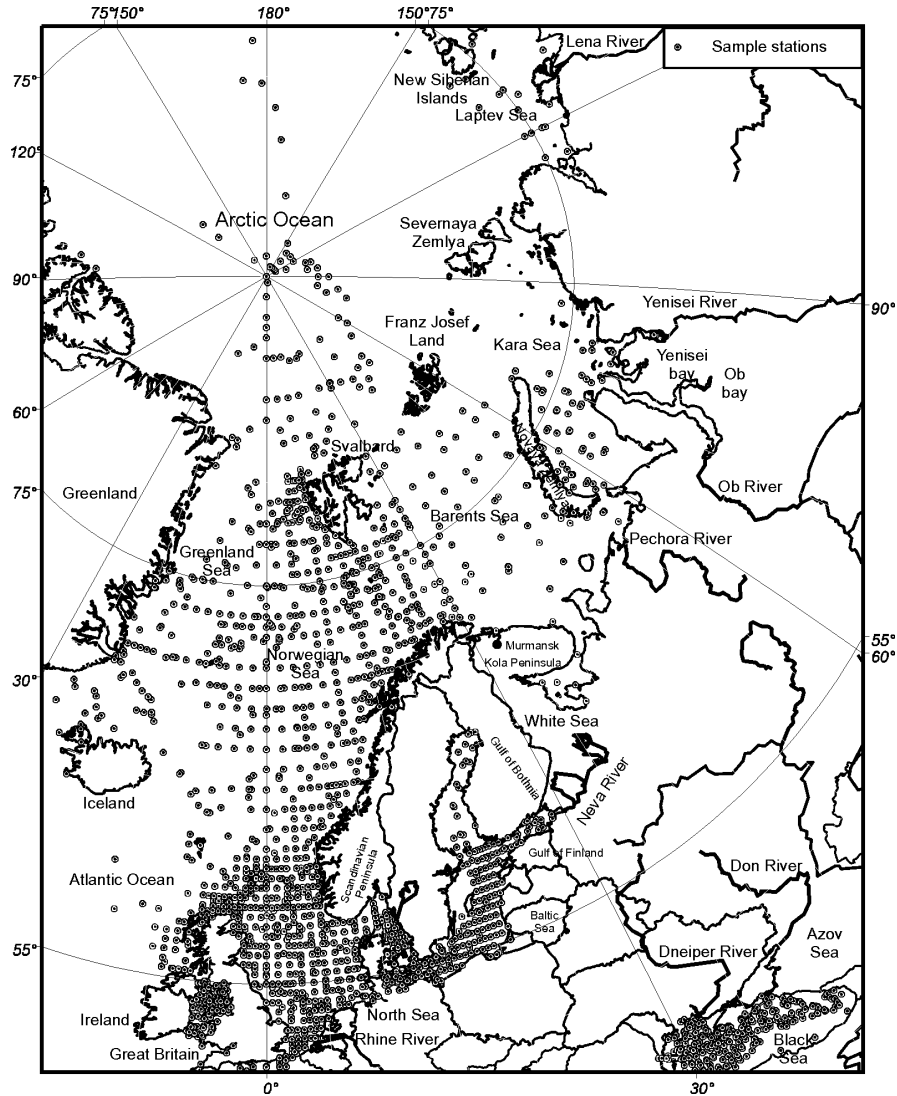
## 1.1 History of investigations

The first investigations of radioactivity in the marine environment were carried out by Russian and foreign institutes in the 1960-1980s. This time period coincided with the period of nuclear tests on the island of Novaya Zemlya and with the large-scale release of radioactivity by Western European nuclear facilities. Radioecological observations in the Barents and Kara Seas were conducted by PINRO, Murmansk Hydrometeorological Service, Typhoon enterprise (Obninsk), the Khlopin Radium Institute the Kurchatov Research Centre. German and British institutes began monitoring artificial radionuclide levels in water and bottom sediments of the Norwegian-Greenland Basin and the western part of the Barents Sea in order to trace the transport of radioactivity from Western European nuclear facilities (Kautsky 1980, 1986; Vakulovsky et al. 1985; Nies and Nielsen 1996; Nies et al. 1997, 1998) (Fig. 1.1). The Norwegian Institute of Marine Research investigated the contamination of marketable fish species in the zone of direct impact of the Novaya Zemlya nuclear tests.

Important results, characterizing the contamination of the fjord ecosystems of the eastern bays of Novaya Zemlya were obtained by a Russian-Norwegian expedition on the Victor Bujnitsky vessel (Strand and Nielsen 1995). Numerous containers, barges, and reactor compartments containing radioactive substances were located and investigated in the bays Tsivolki, Stepovogo and Abrosimova using side-view sonar, video camera and nuclear detectors. Similar works were carried out in the Novaya Zemlya bays and on the adjoining shelf by the Russian Institutes VNIIOceangeologia and Sevmorgeologia, together with the Bedford Oceanographic Institute (Ivanov et al. 1995 a, b; Ivanov 1999). Additional information was collected on the distribution of radionuclides in other bays of Novaya Zemlya including Chernaya and Pechorskaya Bays. In 1994 the Murmansk Hydrometeorological Service conducted a detailed investigation of radionuclide accumulation in bottom sediments in the area adjoining Atomflot, the nuclear ship repair and transport enterprise in Murmansk (Namyatov 1995, 1998).

In 1990-1995 a series of surveys were carried out in the Barents and Norwegian Seas by the British Ministry for Agriculture, Fisheries and Food (MAFF), the German Hydrographic Institute, the Norwegian Fishery Directorate, the Norwegian Institute of Marine Research and others (Dahlgaard 1993; Sickel et al. 1995; Foyen and Swaren 1995; Solberg et al. 1995). These investigations resulted in a large amount of environmental data on the radioactivity content of seawater, bottom sediments, fish and other organisms (Fig. 1.2).

In 1990-1996 a number of investigations were carried out in the Norwegian Sea area at the site of the sunken atomic submarine Komsomolets (Blindheim et al.



**Fig. 1.1.** Sampling locations for  $^{137}\text{Cs}$  in the surface layer of the European seas and Arctic Basin (1970-1990s)

1994; Strand et al. 1994; Kuznetsov et al. 1996, 1997 a; Strand 1998; Lisovsky 1999). Participating institutes included the Norwegian Institute of Marine Research together with the North Atlantic Treaty Organization (NATO), and the Institute of Oceanology of the Russian Academy of Sciences (IO RAS) together with the Russian Ministry of Emergency Situations and the Hydrographic Service

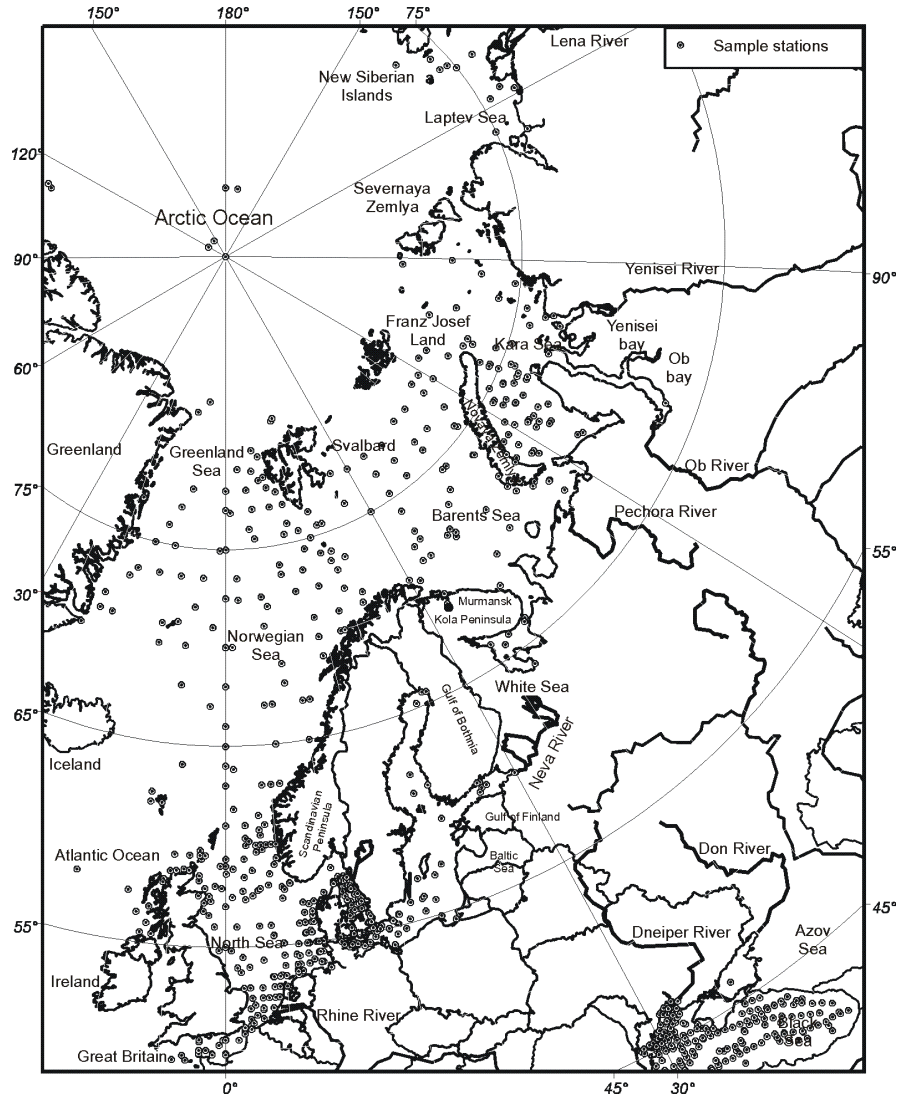


Fig. 1.2. Sampling locations for  $^{90}\text{Sr}$  in the European seas and Arctic Basin (1950-1990s)

of the Russian Northern Fleet. The general conclusion of these investigations was that radioactive contamination of sediments and bottom fauna at the site of the sunken submarine is insignificant.

A number of other investigations were aimed at assessing the impact of artificial radionuclide discharges to the Kara and Laptev Seas via the Ob and Yenisey Rivers. Investigations were carried out in 1992-1998 by the Typhoon enterprise, Texas University, IO RAS, Geochemical Institute of RAS, Khlopin Radium Insti-

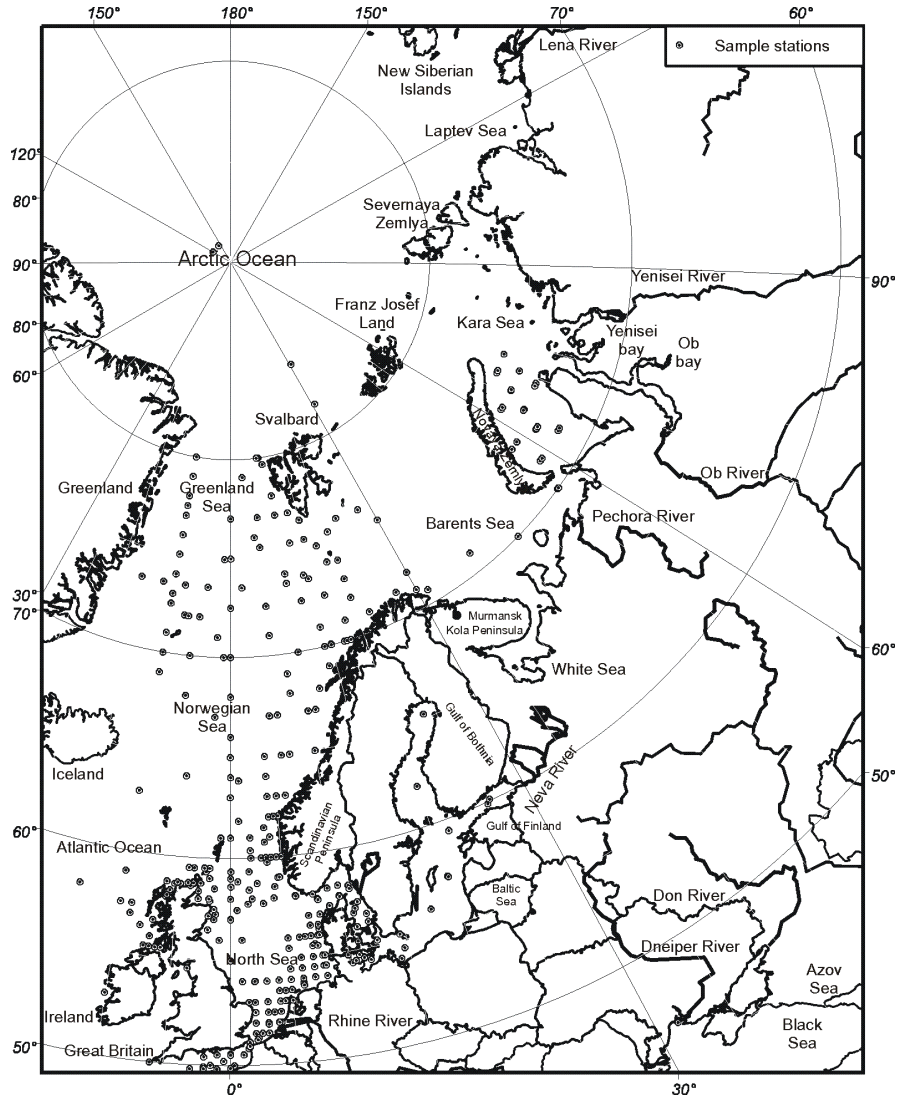
tute, and GEOMAR (Germany) (Vakulovsky et al. 1994; Sapozhnikov and Plishkin 1994; Kuznetsov et al. 1994; Champ et al. 1994; Brooks et al. 1995; Panteleev et al. 1995; Trapeznikov et al. 1997; Chumichev 1997; Nikitin et al. 1997). The role of the Siberian nuclear industries in the contamination of both living and non-living components of the marine environment became apparent during these investigations. Previous investigations in the area were focused almost entirely on investigating radioactivity levels in the non-living components of the marine ecosystem, e.g. seawater and bottom sediment (Fig. 1.3, 1.4).

In 1990-2000 the scientific staff of Murmansk Marine Biological Institute (MMBI) dedicated themselves to investigating marine radioactivity in numerous Arctic and Sub-Arctic marine ecosystems using a multidisciplinary oceanographic investigative approach. Several cruises were carried out in cooperation with M. Sklodowska-Kurie University (Lublin, Poland), the Norwegian Polar Institute, the Norwegian company Akvaplan-niva, the Finnish Institute of Marine Research, the Finnish Institute of Radiation and Nuclear Safety, Alfred Wegener Polar Institute, the Marine Research Institute (Germany), and others. For example, MMBI carried out investigations of the plutonium content in bottom sediments, fish and algae in the White Sea and adjacent areas (Rissanen et al. 1997 a, b).

The cooperation of specialists from different countries and institutes has greatly enriched methodological and scientific aspects of the MMBI investigations. As a result of these endeavours, advances have been achieved in the understanding of environmental radioactivity and a number of fundamental contributions have been made in the field of radioecology (Matishov et al. 1992 a, b, c, 1993 d, 1994 a, b, c, 1995 a, b, 1996 e, f, 1997 a, b, c, d, e, f, 1998 b, c, 1999 a, b, c, d, e, i, h, 2000 a, b, c; Matishov and Denisenko 1996; Matishov G. and Matishov D. 1997; Matishov D. 1992 b, 1993 b, 1994, 1997, 1998; Matishov D. 1993; Matishov D. and Rissanen 1993; Matishov D. et al. 1993, 1995 a, b, 1996, 1997; Rissanen et al. 1995; Matishov D. and Namyatov 1997; Matishov D. and Matishov G. 1998).

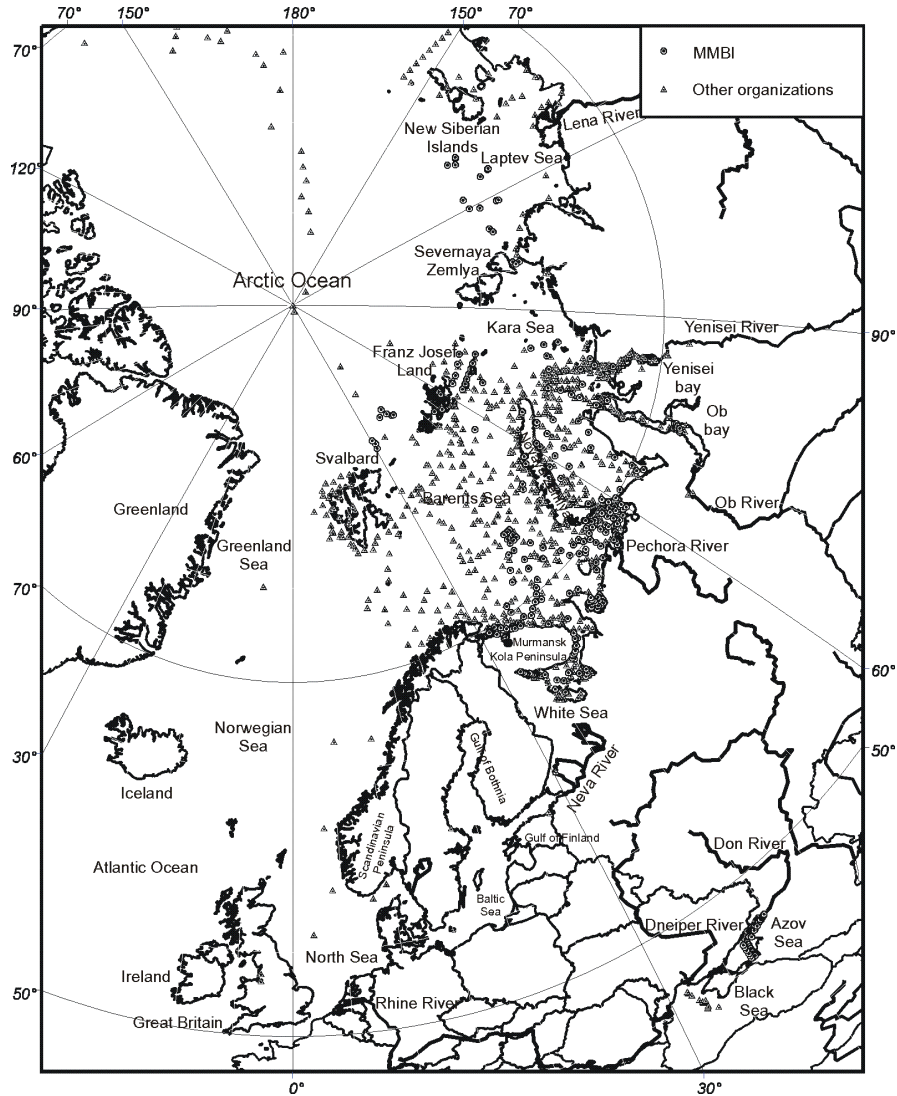
Radioactivity that is present in Arctic and sub-Arctic marine ecosystems as a result of human interventions is the focus of this book. The work is based on the analysis of an extensive collection of radioactivity measurements on marine organisms representing diverse taxonomic groups combined with supporting environmental data. Anthropogenic radionuclides ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{60}\text{Co}$ , and  $^{239, 240}\text{Pu}$ ) in the Barents-Kara region is emphasized in this work. Particular attention is given to the investigation of the bays of the Kola Peninsula and Novaya Zemlya, as well as the estuaries of the Yenisey, Ob, and Pechora Rivers. Attention is drawn to this region due to the existence of several important regional and local radionuclide emission sources: the radioactive waste burial grounds on the continental shelf, the bases for Russia's nuclear fleet, and the former nuclear test site at Chernaya Bay, Novaya Zemlya (Matishov et al. 1993 c). Conditions in these areas are compared with other areas where sources of radioactivity are present such as the Baltic Sea. It is equally important to study the spectrum of artificial radionuclides on shelves and coastlines where local sources of contamination are absent. In this connection we include a discussion of the radioecological situation in the Azov Sea based on studies conducted from 1995-1997 (Matishov et al. 1999 a, b, 2000 c).

For the first time, the levels of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239,240}\text{Pu}$  accumulation in different species and populations of marine organisms is shown, as well as the



**Fig. 1.3.** Sampling locations for  $^{239,240}\text{Pu}$  in the European seas and Arctic Basin (1950-1990s)

characteristics of radionuclide migration across trophic chains from macrophytes and plankton to zoobenthos, fish, birds, seals and whales. This work is more than a compilation and review of a unique radioactivity database. A synthesis of the data is carried out based on the principals of three scientific fields- radioactivity,



**Fig. 1.4.** Sampling of radionuclides in bottom sediments of the European seas and Arctic Basin (1950-1990s)

ecology, and oceanography. This work culminates in a new scientific perspective of how living organisms, in conducting their normal life-sustaining activities, sequester and transport anthropogenic radionuclides and thereby contribute to the assimilation capacity of marine ecosystems impacted by this form of environmental pollution.

## 1.2 Methods

The results of the Murmansk Marine Biological Institute expeditions conducted between 1990 and 2000 serve as the foundation for this work. These expeditions brought together MMBI scientists working in all fields of oceanography, as well as ecotoxicology. A special oceanographic and radioecological survey was carried out onboard the RV *Dalnie Zelentsy* in September 2000 in the area of the sunken atomic submarine *Kursk* on the Murmansk Bank (Fig. 1.5).

In addition to the expeditions carried out on the MMBI research vessel RV *Dalnie Zelentsy* and research and transport vessel *Pomor*, we have carried out oceanographic and radiological investigations on the German icebreaker *Polarstern*, atomic icebreakers *Arktika*, *Soviet Union*, *Tajmyr* of the Murmansk Shipping Company, the vessel GS-440 of the Hydrographic Service of the Northern Navy, RV *Academician Petrov* of Geohemical Institute of RAS and many others.

Our strategy of sampling was directed on the elucidation of the artificial radionuclide contamination levels of all components of the marine environment and the ecological pyramid of the Arctic seas. Such an approach is necessary to (1) determine the migration of radionuclides  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{239, 240}\text{Pu}$  over the food chain from macrophytes and plankton to sea mammals, (2) reconstruct the pathways of radionuclide transfer, and (3) identify radionuclide sources. MMBI studies were also carried out in the Azov Sea consisting of the sampling of bottom sediments and benthic organisms for the determination of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{239, 240}\text{Pu}$ .

The new approach applied in our radioecological investigations involved the efforts of MMBI experts in taxonomy (algologists, benthologists, ichthyologists, ornithologists) to determine species and classify flora and fauna specimens into groups. As a result,  $\gamma$ -spectrometric and radiochemical analyses have been carried out on more than 300 species of marine biota.

Samples were collected using routine methods and equipment in accordance with the methods adopted by the International Atomic Energy Agency (IAEA 1996).

Sea water samples were collected in 100 liter volumes (Fig. 1.6) and filtered through the sorbent  $\text{Cu}_2\text{Fe}(\text{CN})_6$  to extract cesium according to the methods of the German Federal Maritime and Hydrographic Agency (GFMHA 1997). Zooplankton samples were collected using towed plankton nets in the pelagic layer to a depth of 100 meters. Zoobenthos were sampled with a Van-Veen grab, and Ocean-50 box corer.

Bottom sediments were sampled by Van-Veen grab, Ocean-50 box corer, dredge, gravity corer Gemini and a 4 meter long gravity corer. (Fig. 1.7, 1.8). If the sediments in the corer were not mixed, the surface layer (0-2 cm) was removed using a plastic tube of 104 mm diameter. The sediment column retrieved by the



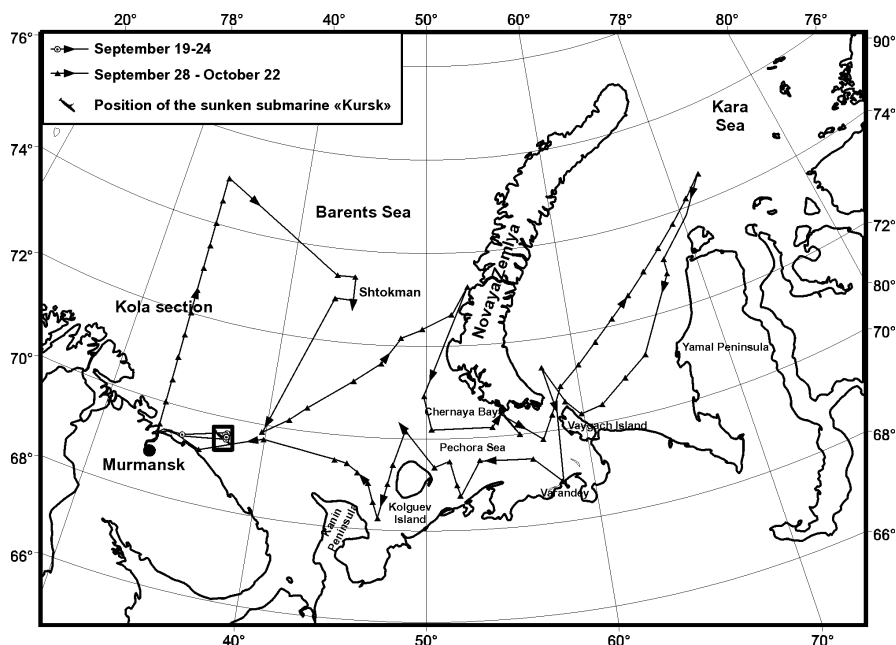


Fig. 1.5. MMBI expeditions on RV «Dalnie Zelentsy» in 2000

plastic tube was subsequently sliced into 10 mm layers. In some cases the first 10 cm of the sediment column was sliced into 2 cm interval. Below 2 cm sediments were sliced into 5 cm intervals.

Red and Phaeophyceae algae (*Laminaria Saccharina*, *Alaria* sp., *Fucus vesiculosus*, *Fucus inflatus*, *Ascophyllum nodosum* and others) were sampled in the littoral zone of the White, Barents, Pechora and Kara Seas (Fig. 1.8). Many samples of macrophytes were collected in the bays adjoining the different atomic bases. In addition, lichens of species *Cetraria nivalis*, *Cladonia mitis*, *Cladonia Rangiferina*, *Cladonia alpestris*, were collected on the Kola Peninsula coast and on the south-eastern islands of the Barents Sea and Novaya Zemlya.

Benthic organisms were collected in representative geomorphological zones of the Barents-Kara Sea continental shelf. The benthic collections were sorted by species, and the most abundant organisms were taken for radiochemical analyses. Specimens from more than ten groups were analyzed: Annelida, Bryozoa, Gastropoda, Echinodermata, Decapoda, Amphipoda, Ophiuroidea, Asteroidea and others.

Sea birds were taken for analyses in August 1996 on the eastern coast of the Kola Peninsula and in the adjoining bays Ivanoskaya and Drozdovka: two sea gulls (*Larus marinus*), two common gulls (*Larus carnus*), two great skuas (*Catharacta skua*), one goosander (*Mergus merganser*), one stint (*Tringa erythropus*) and six Temmink's stints (*Calidris minuta*). Sea birds were taken for analyses in

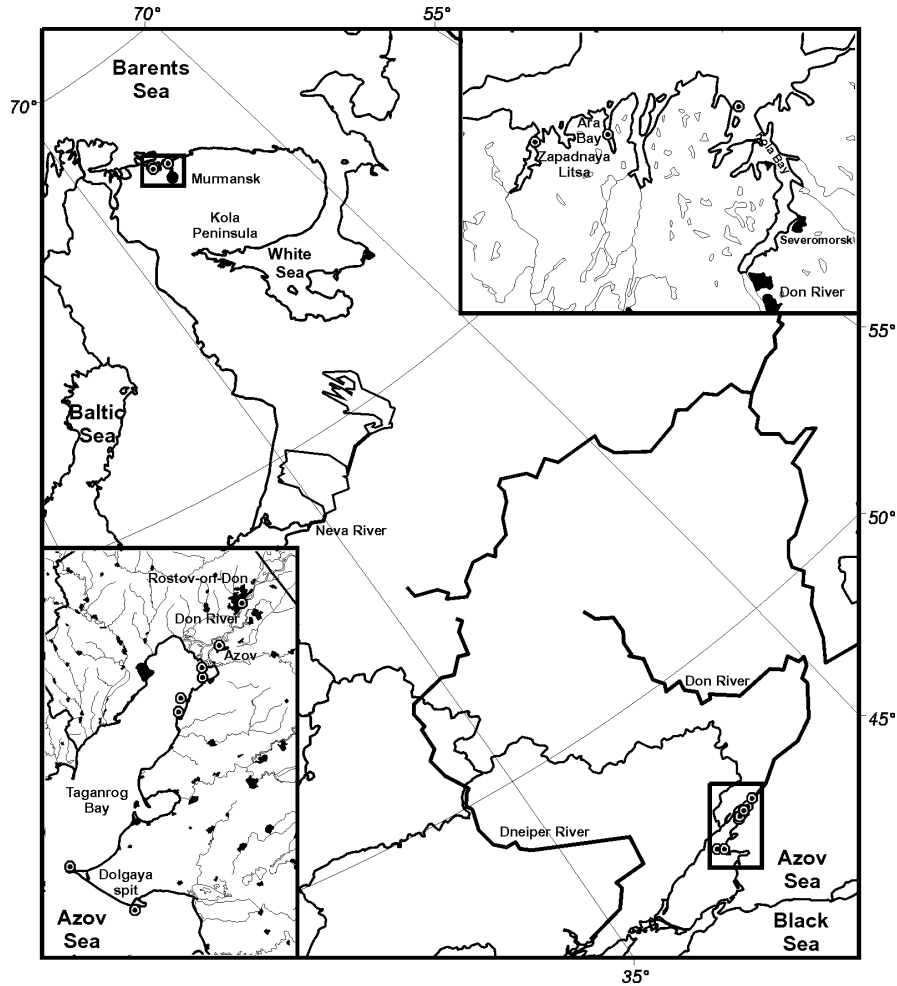
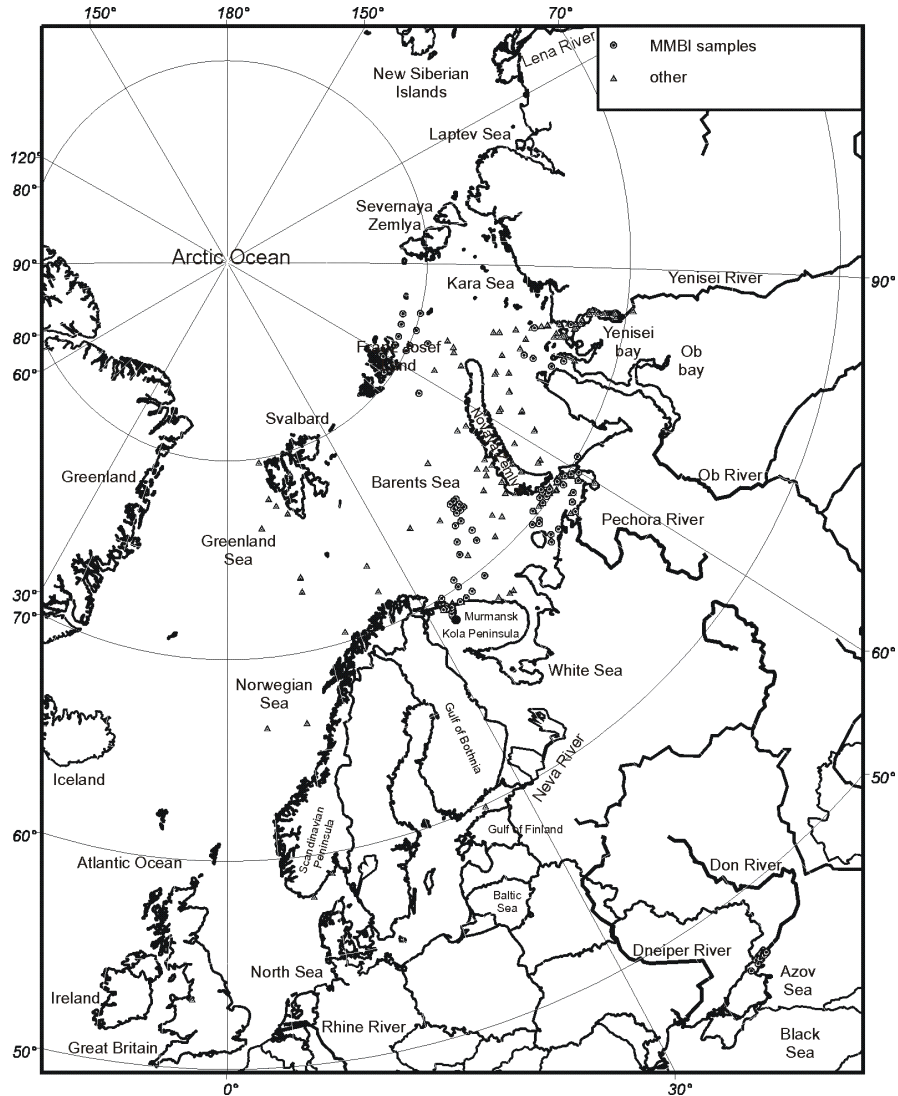


Fig. 1.6.  $^{137}\text{Cs}$  investigations in the European seas during 1990s

February 1996 in the Dalnie Zelentsy area (Fig. 1.10) and adjoining Yarnyshnaya Bay: four eiders (*Somateria mollissima*), one black guillemot (*Cephus grylle*) and four purple sandpipers (*Calidris maritima*).

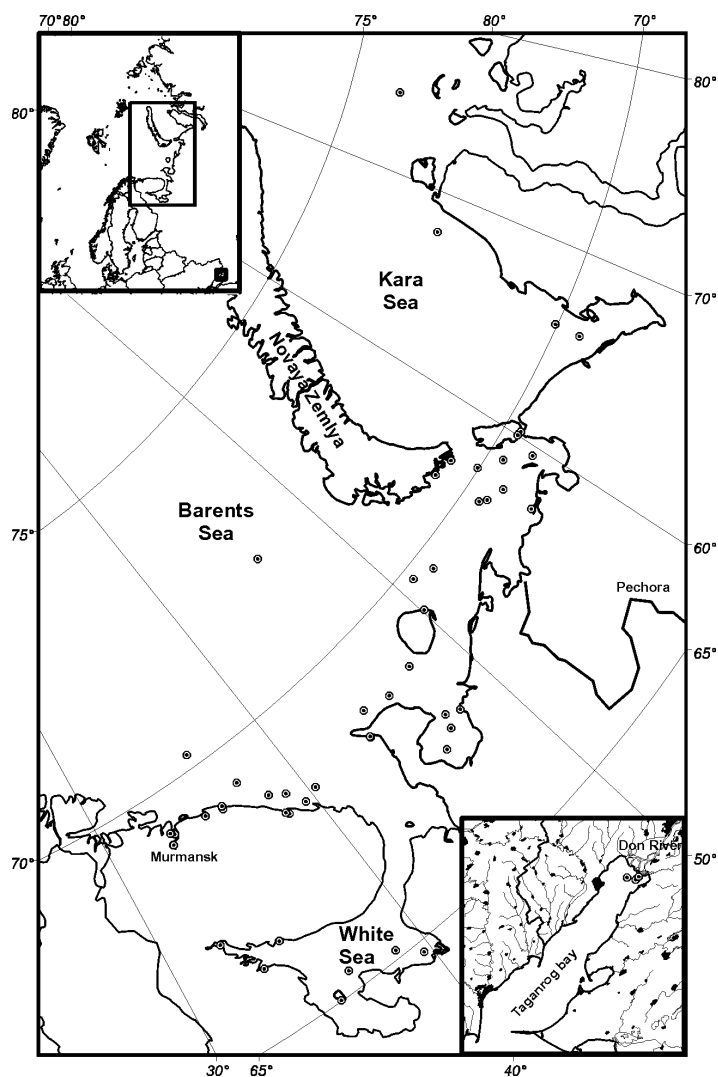
The preliminary processing of algae, benthos, fish, bird, and seal samples varied depending upon the organism type. For example, fish bones were separated from muscle and the liver and other internal organs were collected separately. Muscles and bones were dried (105 °C) and homogenized before analyses. Approximately 60 g of dried muscle and 40 g of bone material were processed for each analysis. Large bivalve and Gastropoda molluscs were separated from shells before drying. Starfish were cut and the piloric appendages, stomachs and bowels



**Fig. 1.7.** Bottom sediment samples for the analysis of  $^{239, 240}\text{Pu}$  in the European seas and Arctic Basin (1950-1990s)

were separated from the skin, dried and homogenized. Only the cod liver with high fat content was analyzed wet. Samples were placed in Marinella containers of 550 ml or plastic vessels of 35 or 100 ml for analysis by high-purity gamma spectrometry analysis. The minimum counting time on the detector was 1000 min.

The following laboratories provided  $\gamma$ -spectrometric and radiochemical analyses: the Chair of radiochemistry and colloidal chemistry of M. Skłodowska-Kurie University, (Lublin, Poland); Northern Branch of the Finnish Centre of Radiation



**Fig. 1.8.** Benthos and macrophyte sampling stations (MMBI unpublished)

and Nuclear Safety (STUK, Finland); Laboratory of Radiochemistry of the Bedford Oceanographic Institute (Canada); Department of Marine Radioactivity of German Hydrographic Institute (Hamburg); Typhoon enterprise (Obninsk, Rus-

sia); V.G. Khlopin Radium Institute (S.-Petersburg) and Murmansk Hydrometeorological Service.

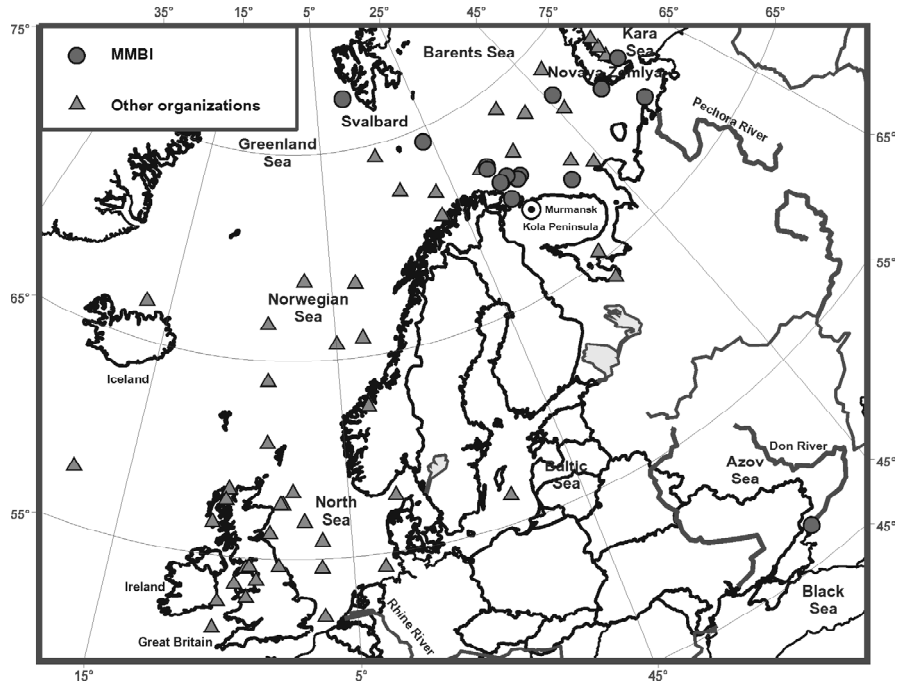
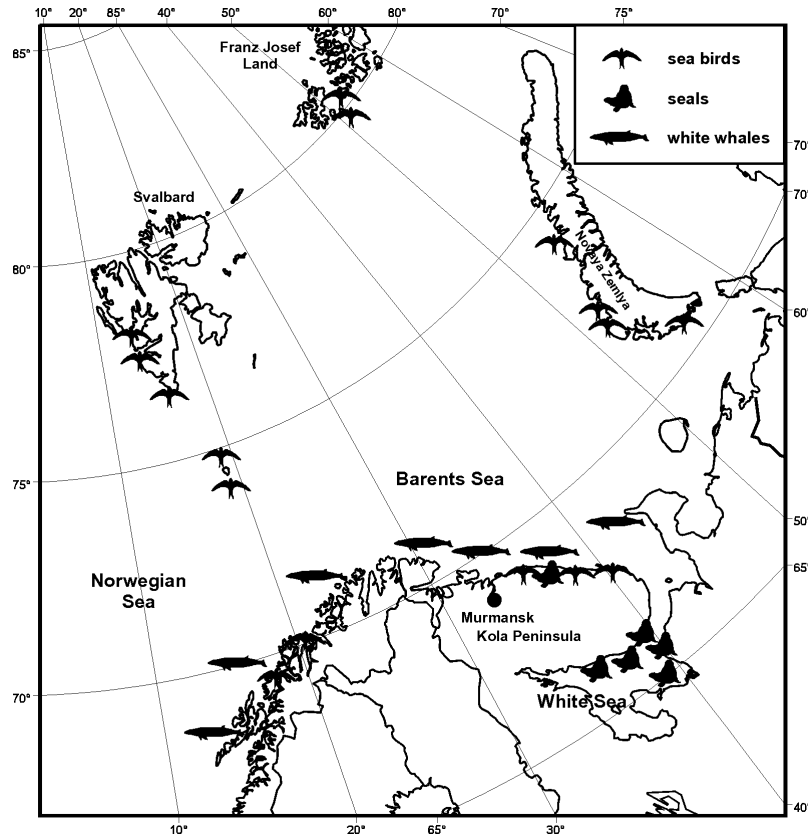


Fig. 1.9. Samples taken for the investigation of radioactivity in marine fish

Analyses were primarily carried out to determine the content of  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and other  $\gamma$ -radiating isotopes in environmental material and biota. The determination of  $\gamma$ -emitting radionuclides is performed using semi-conductor  $\gamma$ -spectrometers, containing Ge(Li) detectors of high resolution according to internationally established methods (IAEA 1996).

For the determination of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$ , weighed sediment, water, or marine biota samples are heated to temperatures not higher than 350–400 °C. The radionuclide  $^{242}\text{Pu}$  is used as a tracer (IAEA 1996). The radiochemical determination of  $^{239,240}\text{Pu}$  is then carried out on a sample (not more than 10–20 g) by the addition of acids, followed by plutonium separation using ion-exchange resins and plutonium electrolytic deposition onto stainless steel discs. Sample quantification is carried out by alpha particle spectrometry at the V.G. Khlopin Radium (Matishov et al. 1999 c, e) and at the Finnish Centre of Radiation and Nuclear Safety (Rissanen et al. 1995, 1997 a).  $^{90}\text{Sr}$  determinations are carried out by the conventional ‘fumic nitric acid’ method after the addition of  $^{85}\text{Sr}$  isotope as tracer. The  $\beta$ -activity of the prepared  $^{90}\text{Sr}$  sample is measured on a scintillation counter NaI(Tl).

Thus, during 1990-2000 more than 2000 analyses on the content of  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ , and  $^{239,240}\text{Pu}$  in environmental and biological samples were carried out (Table 1). As a result, new, more comprehensive, information is available on the content



**Fig. 1.10.** Sampling locations for sea birds, seals, and whales

and distribution of the above-mentioned radionuclides in different compartments of Russia's Arctic and sub-Arctic marine ecosystems. This radioecological database provides the foundation for presenting monitoring methodology and critical tasks for marine radioecology in the future - the monitoring of  $^{239,240}\text{Pu}$  will be a critical task. Plutonium is a main irradiator of marine biological resources and will require monitoring to assess its impact on ecosystems as a whole.

Systematizing and analyzing the large amount of radioecological, radiochemical, oceanographic and biological data on the Arctic Seas required the use of traditional cartographic methods as part of the research (Matishov et al. 1994 a; Ionov et al. 1996). In the course of this work, a series of special radioecological biogeographic maps were created to present information on the entire spectrum of ar-

tificial radionuclides in both living and non-living components of the environment. The cartographic basis for these maps was adopted from the Numerical Atlas GEBCO 97.

**Table 1.1.** Analyses ( $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{239,240}\text{Pu}$ ) performed on samples from different components of the marine ecosystem (MMBI data)

Ecosystem component	Azov Sea				White Sea				Barents Sea				Kara Sea				Laptev Sea				Total			
	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>239,240</sup> Pu	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>60</sup> Co	<sup>239,240</sup> Pu	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>60</sup> Co	<sup>239,240</sup> Pu	<sup>137</sup> Cs	<sup>60</sup> Co	<sup>239,240</sup> Pu	<sup>137</sup> Cs	<sup>60</sup> Co	<sup>239,240</sup> Pu	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>60</sup> Co	<sup>239,240</sup> Pu			
Water	7	5	-	-	-	-	-	3	-	-	-	-	-	-	-	-	-	10	5	-	-			
Sediment	121	47	21	51	4	1	18	440	21	60	96	95	8	22	19	726	72	99	157	157	157	157		
Macro-phytes	3	3	3	22	-	-	-	53	7	6	22	-	-	-	-	-	-	78	10	6	25			
Plankton	1	1	1	-	-	-	-	-	-	-	-	-	-	-	-	-	-	1	1	-	1			
Benthos	-	-	-	19	-	-	-	150	-	4	-	39	-	-	-	-	-	208	-	4	-			
Fish	5	3	-	1	-	-	-	28	-	-	14	-	-	-	-	-	-	34	3	-	14			
Birds	8	-	-	-	-	-	-	30	-	-	-	-	-	-	-	-	-	38	-	-	-			
Seals	-	-	-	133	5	-	5	-	-	-	-	-	-	-	-	-	-	133	5	-	5			
Total (all radionuclides)					Total												1228 96 109 202				1635			





<http://www.springer.com/978-3-540-20197-7>

Radioecology in Northern European Seas

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2004, XXI, 335 p., Hardcover

ISBN: 978-3-540-20197-7