Fishes as radionuclide bioindicators in the area of Leningrad nuclear power plant (Gulf of Finland, Baltic Sea)

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Introduction

The radioactivity of the aquatic environment comprises radionuclides of natural origin, global fallout from nuclear weapon tests, accidents at nuclear facilities and also everyday leakage of radioactive substances from nuclear power plants.

The area under investigation is characterized by a high concentration of nuclear facilities. Leningrad NPP (LNPP) with 4 RBMK-1000 reactors, North-west regional storage facility of low and intermediate level radioactive wastes (LSK "Radon") and the North-west science and industrial center of atomic energy (Figure 1).

Radioecological monitoring of the Koporskaya Bay coastal waters (LNPP cooling water body) as well as rivers and lakes of the bay drainage basin, is a part of the integral ecological monitoring of Sosnovy Bor Region. The work was carried out at the Regional environmental monitoring laboratory of the V.G. Khlopin Radium Institute.

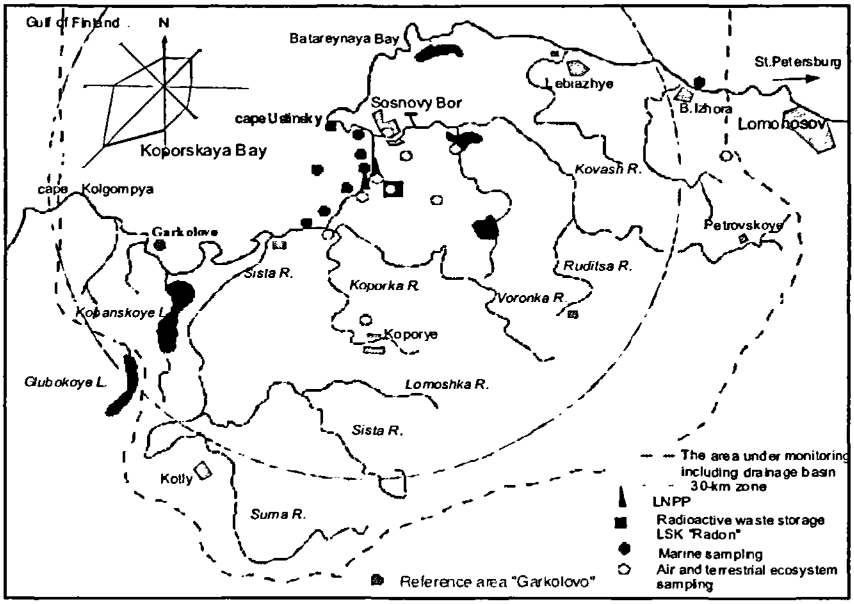

Figure 1
The area under monitoring (Sosnovy Bor Region), including Leningrad NPP 30-km zone.

Materials and methods

Most of the fish samples were collected in 1982-1999. In that period, 462 samples of 25 fish species were analyzed for radionuclide content. Fish were caught by our own efforts or collected from local fishery company catches. The locations of sampling sites are shown in Figure 1. A substantial portion of our catches comprised samples from the LNPP water input area. Fish samples were analyzed by routine biological methods before treatment for radioactivity measurement.

After biological analysis the samples were prepared to determine gamma-emitting radionuclides by means of generally accepted methods (Marey & Zykova, 1980), in particular: "wet" ashing of fish samples in the presence of nitric acid and hydrogen peroxide mixture in 3:1 proportion, with the subsequent ash calcination at 450°C to produce mineral compounds.

Semi-conductor spectrometers with DGDK detectors type and ÀI-1024-95 multi-channel analyzers as well as a Canberra HPGe-detector (20% efficiency) were used for the purpose of gamma-emitting radionuclides determination in the energy range from 50 keV up to 2000 keV. The low detection limit of measurements of gamma-emitting radionuclides volumetric activity in fish samples was 1.0 $Bq.kg^{-1}$ fresh weight. The activity determination error was equal to 10-30% at the 95% confidence level.

Results and discussion

The influence of nuclear power plants on radioactivity of water and aquatic ecosystem components of the cooling water bodies is evident mainly in heated water outlet canals and adjacent sea areas. Moreover, technogenic radionuclides content is substantially lower than the content of radionuclides of natural origin (e.g., ^{40}K, ^{232}Th, ^{7}Be), and also it is lower than levels of radioactivity allowed by the Radiation Safety Standards (1996).

Activation products (mainly: ^{51}Cr, ^{58}Co, ^{60}Co, ^{59}Fe, ^{54}Mn) account for most of the radionuclide contribution to the artificial radioactivity of the aquatic environment in NPP areas. Since the probability of detection of radionuclides in hydrobionts is significantly higher than in water samples, fish were used as indicators of technogenic radionuclides discharged into the aquatic environment. The main results of the investigations, conducted in Sosnovy Bor Region, are presented in the papers and reports of the Laboratory (Kryshev, 1992; Blinova, 1994; Blinova, 1996; Zimin *et al.*, 1997; Blinova *et al.*, 1998; Blinova *et al.*, 1999; Blinova *et al.*, 2000).

Biological transfer of radioactivity

Activation products were often detected in fish samples from the bay, but these elements were of infrequent occurrence in fish collected in rivers of the Koporskaya Bay drainage basin. The occurrence of activation products in few fish samples from the Kovash River due to fish migration (more often - during spawning). From Table 1 it is clear, that the most frequent occurrence of the above mentioned elements was in fish samples from the LNPP outlet canal.

The occurrence frequency of these radionuclides in fish samples from the sea sites, spaced at 3-5 km from the nuclear plant, was lower than in the LNPP outlet canal. In the Kovash River (1 km upstream of the river mouth) near 3 km downstream from the LNPP heated water outlet, activation products were detected only a few times. It should be kept in mind that very few fish species are real

Fish species and location	^{54}Mn	^{60}Co	^{65}Zn
LNPP outlet canal			
roach	18 (30 %)	11 (52 %)	70 (43 %)
threespined stickleback	1.4 (38 %)	5.6 (62 %)	30 (56 %)
Koporskaya Bay			
roach	1.5 (7 %)	10 (14 %)	60 (28 %)
threespined stickleback	<	0.4 (5 %)	0.4 (5 %)
Kovash River, down stream			
roach	3.7 (17 %)	1.9 (17 %)	34 (17 %)
Kovash River, upper stream			
dace	<	0.4 (10 %)	13 (40 %)

Note: 1) <- below the detection limit;
2) detection frequency is indicated in parentheses

Table 1
The mean specific activity of NPP originated radionuclides (Bq. kg^{-1} fresh wt.) in fish from the Leningrad NPP area between 1976-1984. The value in brackets indicates the proportion of fish in which these radionuclides were detected.

migrating species. Except for anadromous species, namely *Lampetra fluviatilis*, *Salmo salar*, *Salmo trutta*, most species migrate only to the down stream sites for spawning or feeding. Thus roach (*Rutilus rutilus*) dwelling in the Koporskaya Bay never migrate to the upper stream part of the river.

In the upper stream of the Kovash River radionuclides originated from the LNPP were registered in only one fish species, namely dace (*Leuciscus leuciscus*). This may be evidence of the radionuclide bio-transfer from the nuclear power plant to the river. Dace usually dwells at streams; its occurrence in the Koporskaya Bay is of low frequency; nevertheless this species, the activation products in the samples confirm the far migration of this species.

age	^{137}Cs	^{51}Cr	^{54}Mn	^{60}Co	^{58}Co	^{65}Zn	^{59}Fe
0+	1.0	32.0	42.3	30.9	13.4	16.6	45.9
2+	1.0	27.2	32.6	21.1	10.5	82.8	27.9
3+	0.8	9.0	19.6	14.1	5.3	<	20.8
4+	1.2	<	7.6	5.4	3.0	<	7.4

Note: 1) <- below the detection limit;

Table 2
Radionuclide concentrations (Bq.kg^{-1} fresh wt.) in roach (*Rutilus rutilus*) of different age groups collected in the NPP outlet canal on 12.08.1983.

Special features of radionuclide accumulation in connection with fish age

Table 2 shows the content of ^{54}Mn, ^{51}Cr, ^{60}Co and other activation products in roach collected at the LNPP heated water outlet in the summer of 1983. The unusual character of accumulation of radionuclides was registered for the roach of different age groups collected on 12.08.1983. All concentrations of the radionuclides, except for ^{65}Zn, clearly decreased in the series "young fish - old fish".

This phenomenon is presumed to be explained by the significant contribution of the radionuclides absorbed by mucous skin epithelium to the whole radionuclide content. In view of the fact, that small-sized fish have a relatively larger skin surface areas than big-sized ones, young fish individuals can absorb more radionuclides than old fish individuals of the same total body weight. It is known, that the mean concentration factor of radionuclides and the mean body length (or age) of fish correlate positively (Rowan & Rasmussen, 1994; Kasamatsu & Ishikawa, 1997), which is associated mainly with food habits and trophic level. Since, as a rule, an opposite situation takes place.

The fact that the roach was immediately exposed to heated water with ^{54}Mn, ^{51}Cr, ^{60}Co and other radionuclides, discharged from the LNPP, at the outlet canal, plays, of course, the important part in the process of radionuclide accumulation.

Long time dynamics of ^{137}Cs content in fish

The radiocesium dynamics in populations of some fish species from the Koporskaya Bay, as well as from rivers and lakes of the bay drainage basin, was reviewed. The data available, partly presented in Table 3, made it possible to characterize radionuclide accumulation by fish of different ecological groups and trophic levels and to trace its dynamics. Since 1986 (after the accident at Chernobyl NPP) up to nowadays, the major contribution to cesium content in fish was via radionuclides from the Koporskaya Bay drainage basin, which was subjected to Chernobyl fallout mainly in the west part.

At the present time, considerable radiocesium concentrations were registered only in fish caught in Lake Glubokoye (the Koporskaya Bay drainage basin); both ^{137}Cs and ^{134}Cs were registered there (Figure 2). The isotope ratio ^{137}Cs: ^{134}Cs in fish samples from Lake Glubokoye indicated the Chernobyl origin of these radionuclides. In 1997 and 1998 ^{137}Cs concentrations in perch and roach from Lake Glubokoye were roughly two orders of magnitude greater than concentrations in the same species from the Koporskaya Bay coastal waters.

Koporskaya Bay has a wide water exchange with the open part of the Gulf of Finland, that is why radiocesium contents in coastal water and hydrobionts, including fish, were significant only in the first years after the accident at Chernobyl NPP, with their subsequent fast reduction.

The processes of intake and elimination of ^{137}Cs by fish of different food habits and trophic levels were different in their character. The maximum ^{137}Cs concentrations in prey fish species were registered in 1986-1987. In comparison, predatory species maxima occurred a

year	Baltic herring				perch				threespined stickleback			
	n	min	max	ave	n	min	max	ave	n	min	max	ave
1982	6	1,5	5,6	2,9	2	0,4	2,2	1,3	13	0,8	11,6	4.2
1883	7	0,8	4,3	1,7	4	1,6	4,0	2,8	15	0,3	2,9	1.4
1984	8	1,3	2,1	1,6	3	2,3	4,8	3,1	11	0,7	2,9	1.6
1985	7	0,5	3,7	1,7	-	-	-	-	9	1,3	3,0	2.0
1986	2	21,1	33,2	27,1	1	21,7	21,7	21,7	5	95,6	233,5	134.9
1987	4	21,0	50,7	32,6	2	83,3	163,4	123,3	6	27,5	255,2	99.2
1988	2	14,1	24,6	19,4	7	29,2	196,8	102,4	-	-	-	
1989	-	-	-	-	2	75,9	149,5	112,7	20	1,6	45,9	30.7
1990	3	13,3	24,1	19,2	2	54,0	178,3	116,2	24	10,9	54,0	30.3
1991	3	14,1	23,9	17,9	1	15,7	15,7	15,7	14	12,6	34,0	20.9
1992	2	9,2	13,7	11,4	-	-	-	-	7	15,3	21,8	18.4
1993	-	-	-	-	-	-	-	-	1	14,0	14,0	14.0
1994	1	9,3	9,3	9,3	-	-	-	-	1	10,9	10,9	10.9
1995	1	7,4	7,4	7,4	-	-	-	-	-	-	-	
1996	1	6,7	6,7	6,7	-	-	-	-	4	1,2	22,2	12.6
1997	1	10,8	10,8	10,8	5	17,3	23,4	21,2	2	12,7	13,4	13.1
1998	-	-	-	-	1	16,3	16,3	21,2	3	8,5	11,5	10.1
1999	1	6,1	6,1	6,1	1	7,4	7,4	7,4	3	8,7	13,4	11.3

Table 3
^{137}Cs concentrations (Bq.kg^{-1} fresh wt.) in Baltic herring (*Clupea harengus membras*), perch (*Perca fluviatilis*) and threespined stickleback (*Gasterosteus aculeatus*) collected in the Koporskaya Bay during the investigation period.

year later (Figures 3-6). In 1987-1990, average ^{137}Cs concentrations in perch (predatory species) varied from 100-120 Bq.kg^{-1}. Whereas, the planktivore Baltic herring, the most important commercial fish species in the Eastern Baltic, registered 20-30 Bq.kg^{-1} ^{137}Cs. 7-50 Bq.kg^{-1} it was registered in benthivore roach, a popular sport fishing and also a commercial species.

Substantially higher ^{137}Cs concentrations in samples of threespined stickleback in comparison with Baltic herring, species with similar feeding habits, are worthy of attention (Figure 3). Threespined stickleback is a resident omnivore fish species, feeding predominantly on plankton and meiobenthos. Most of this species samples

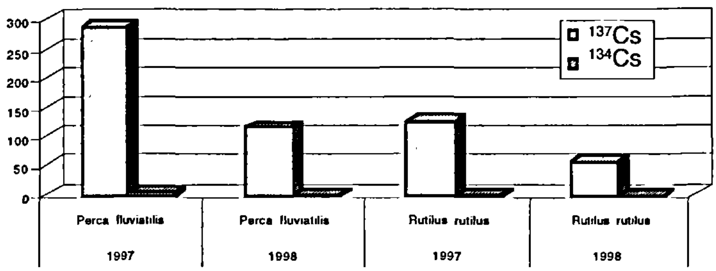

Figure 2
^{137}Cs and ^{134}Cs in prey fish *Rutilus rutilus* and predator fish *Perca fluviatilis* samples (Bq.kg^{-1} fresh wt.) from Lake Glubokoye contaminated with Chernobyl fallout.

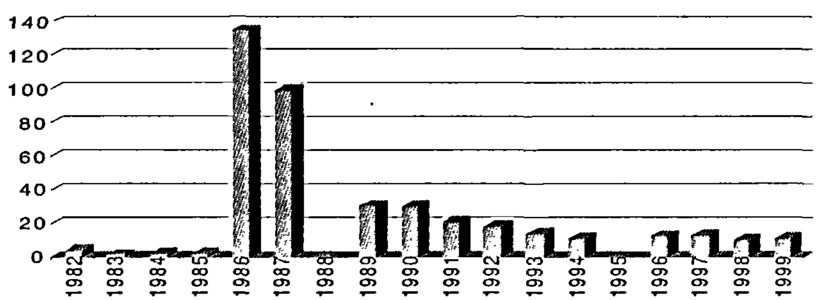

Figure 3
Long time dynamics of ^{137}Cs in omnivore fish *Gasterosteus aculeatus* samples (Bq.kg^{-1} fresh wt.) from the Koporskaya Bay (Leningrad NPP environs).

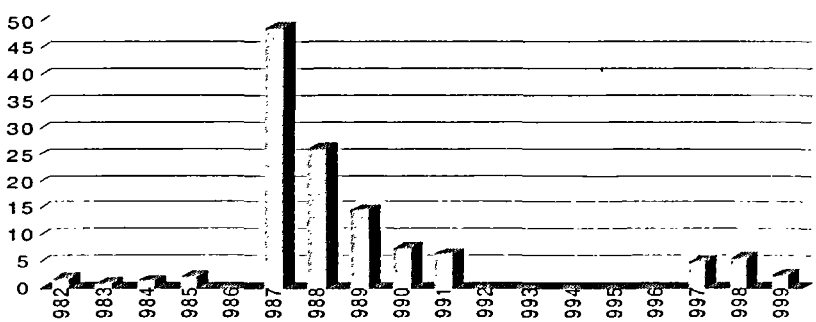

Figure 4
Long time dynamics of ^{137}Cs in benthivore fish *Rutilus rutilus* samples (Bq.kg^{-1} fresh wt.) from the Koporskaya Bay (Leningrad NPP environs).

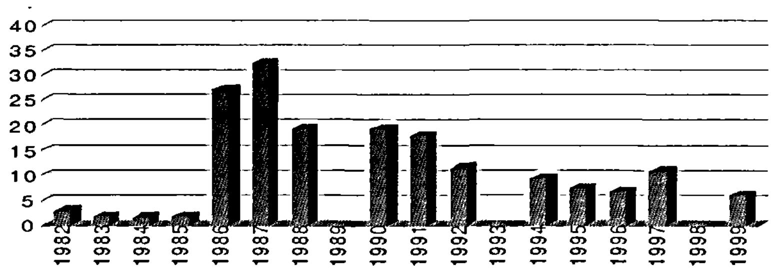

Figure 5
Long time dynamics of ^{137}Cs in planktivore fish *Clupea harengus membras* samples ($Bq.kg^{-1}$ fresh wt.) from the Koporskaya Bay (Leningrad NPP environs).

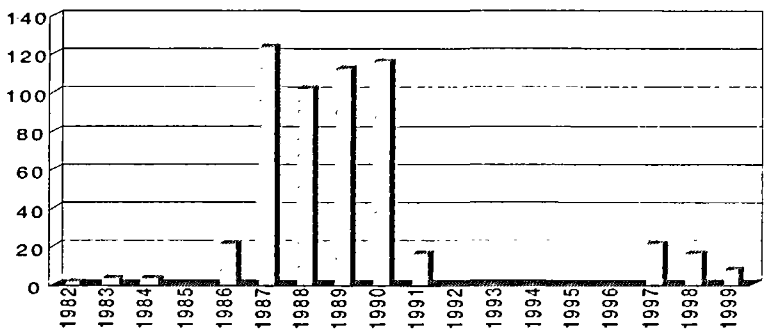

Figure 6
Long time dynamics of ^{137}Cs in piscivore fish *Perca fluviatilis* samples ($Bq.kg^{-1}$ fresh wt.) from the Koporskaya Bay (Leningrad NPP environs).

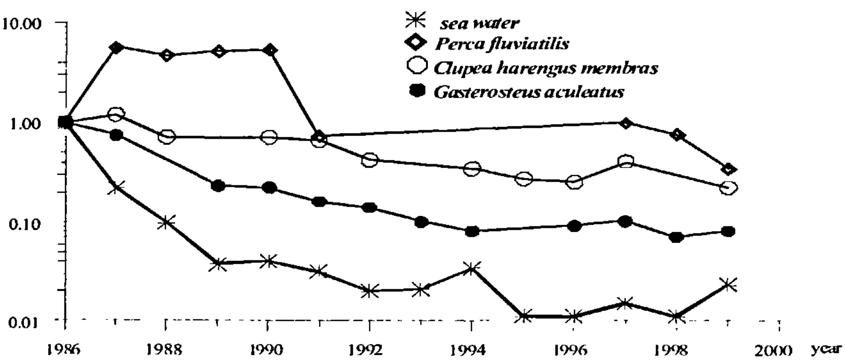

Figure 7
Relative ^{137} Cs concentrations in marine ecosystem components of the Koporskaya Bay, normalized to the data of 1986.

were collected at the water-input area, near the LNPP pump station, where a significant quantity of detritus and silt is accumulated. These are an appropriate substrata for radiocesium retention. The diet of the threespined sticklebacks from that site seems to contain a marked share of suspended organic matter as well as hydrobionts feeding on detritus. Hence, the high ^{137}Cs concentrations in this fish seems to owe their origin to diet.

Figure 7 demonstrates dynamics of ^{137}Cs content in prey and predatory fish after Chernobyl accident in 1986-1999. The steady reduction of ^{137}Cs concentrations in Baltic herring, planktivore species, and also in threespined stickleback, species with wide feeding spectrum (predominantly - planktivore), correlated positively with dynamics of ^{137}Cs content in the sea water. In contrast, most high concentrations of ^{137}Cs in predatory species, namely perch, were registered in 1987, a one year after the accident fallout. Then ^{137}Cs concentrations in perch decreased only after 1991, after a long period of retention.

Internal exposure doses for the population from consumption of local fish

It is important to estimate the internal exposure doses for the human population from consumption of $^{134,137}Cs$ with local fish. A preliminary evaluation of internal exposure doses for the human population, inhabiting the Sosnovy Bor Region, was made in 1998. The consumption of local fish contaminated with $^{134,137}Cs$ in consequence of the Chernobyl NPP accident now accounts for most of the expected doses of internal exposure for the population living on the southern coast of the Eastern Gulf of Finland. These doses vary over the range from 0.16 μSv from consumption of 1 kg of sea fish up to 20 μSv from consumption 1 kg of freshwater predatory fish (averaged for freshwater fish 3.0 μSv), (ISTC Project, 1999).

The annual per capita fish consumption is proposed to be 5 kg according "The Methods of Calculation of Distribution of Radioactive Substances from NPP and Exposure of the Local Population" (Normative Technical Document, 1984), but such a rate seems to be an underestimate, especially for the sea coast areas. However, calculations were made in accordance with the above cited guidelines; and taking into account this fish consumption rate, the expected doses were evaluated as 0.8 μSv from consumption of sea fish or 15.0 μSv from the freshwater fish.

According to "Sanitary regulations in the design and exploitation of nuclear power plants (1993)" adopted in the Russian Federation, the annual limit for population exposure to radioactive substances, originated from all kinds of water usage equals 50 μSv. Thus the consumption of freshwater fish, caught in the contaminated water bodies of the Koporskaya Bay drainage basin, may be considered the most significant radioactive source in the region.

Bibliography

BLINOVA L. D., 1994 — St. Petersburg (Leningrad) NPP's region radioecological state estimation. ENC' 94 International nuclear congress-Atoms for Energy. *Transactions.*, 2: 37-39.

BLINOVA L. D., 1996 — "Littoral ecosystem radioecological monitoring in the region of atomic energy enterprises". *In: Protection of the Natural Environment,* Proceedings of International Symposium on Ionizing Radiation, Stockholm, 20-24 May 1996, 2: 497-503.

BLINOVA L. D., DUSHIN V. N., GOLUBEVA L. V., MICHURINA T. G., 1998 — "Radioecological Monitoring and Radiological Control in the Region of Sosnovy Bor Nuclear Facilities as Baltic Sea Region Safety Basis. - Radiation protection No110". *In: The radiological exposure of the population of the European Community to radioactivity in the Baltic Sea,* EUR 19200, Proceedings of Marina Balt Seminar. Stockholm, Sweden, 1998: 365-372.

BLINOVA L. D., DUSHIN V. N., GOLUBEVA L. V., MICHURINA T. G., 1999 — The development of environmental protection approaches in the region of nuclear facilities at the Russian coast of the Baltic Sea. *In: Radiation. Environmental Protection Approaches for Nuclear Facilities.* Proceedings of the Second Int. Symposium on Ionizing, Canada, Ottawa, 1999, section 6.

BLINOVA L. D., DUSHIN V. N., ZIMINA L. M., 2000 — Methodological approaches to the environmental protection in the area of nuclear power plant on the Russian coast of the Baltic Sea. *In: Problems of development of atomic energy in the Don Region.* Materials of science conference. Rostov-on-Don, 29 February 1 March, 2000. 2: 169-175.

ISTC Project, 1999 — "Ecological assessment of the region, where radioactivity dangerous enterprises are located. North-west region of Russia". *In: Methodological approaches to the organization of ecological monitoring* *system in the vicinity of nuclear facilities.* (1999) Final report, #535-97, 149 p.

Kasamatsu F., Ishikawa Y., 1997 — Natural variations of radionuclide ^{137}Cs concentration in marine organisms with special reference to the effect of food habits and trophic level. *Mar. Ecol. Progr. Ser.*, 160: 109-120.

Kryshev I. I., (ed.), 1992 — *Radiological consequences of the Chernobyl accident.* Moscow, Nuclear Society International: 142 p.

Marey A., Zykova A., (eds), 1980 — *Methodical recommendations for sanitary monitoring of radioactive substances content in the environment.* Moscow.

Normative-Technical Document 38.220.56-84, 1984 — *Safety in Atomic Energy. General principles of NPP safety. The Methods of Calculation of Distribution of Radioactive Substances from NPP and Exposure of the Local Population.* Moscow, Energoatomizdat.

Radiation Safety Standards, 1996 — NRB-96, Hygienic standards GN 2.6.1.054-96, Moscow, 127 p.

Regulations and Standards in Atomic Energy, 1993 — Sanitary regulations in the design and exploitation of nuclear power plants. SP-AS-88/93, Moscow.

Rowan D. J., Rasmussen J. B., 1994 — Bioaccumulation of radiocesium by fish : the influence of physicochemical factors and trophic structure. *Can. J. Fish. Aquat. Sci.*, 51: 2388-2410.

Zimin V. L., Blinova L. D., Zimina L. M., 1997 — "Ecological monitoring in the vicinity of nuclear facilities for risk assessment and decision making". *In: New risk frontiers. Proceedings of the Annual meeting of the Society for Risk Analysis-Europe.*, 15-18 June 1997, Stockholm, Sweden: 746-754.