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Assessment of regularities of ground-level air radionuclide contamination at the Russian coast of the Baltic Sea in the course of the long-term Leningrad Nuclear Power Plant (1983–1999) operation

Lidia D. Blinova,
Lidia V. Goloubeva,
Victor N. Dushin,
Tatiana G. Michurina

Abstract Long-term results of ground-level air radionuclide contamination at the Russian coast of the Baltic Sea (nuclear facilities in the vicinity of the town of Sosnovy Bor) are under consideration. As compared with 1992–1997 [1], the Leningrad Nuclear Power Plant (NPP) equipment hermetic sealing (in 1998–1999) caused both the reduction of detection frequency of ^{131}I aerosol and the maximum concentrations of ^{137}Cs in the ground-level air at the NPP site. Long-term weekly nuclides' concentration time series were examined by the correlation analysis. Results of the analysis are presented. The seasonal fluctuations of ^{7}Be concentrations in the ground-level air were examined by the harmonic (Fourier) analysis. The seasonal fluctuations of its concentrations with the maximum in spring-summer period are shown.

Key words air contamination • artificial radionuclides • ^{7}Be • nuclear power plant

L. D. Blinova✉, L. V. Goloubeva, T. G. Michurina
Regional Environmental Monitoring Laboratory,
V.G. Khlopin Radium Institute,
Ministry of Russian Federation on Atomic Energy,
Box 170/5, Sosnovy Bor, Leningrad region, 188 540 Russia,
Tel.: 7 812 69/ 232 68, Fax: 7 812 69/ 242 38
E-mail: head@atc.sbor.spb.su

V. N. Dushin
Regional Environmental Monitoring Laboratory,
V.G. Khlopin Radium Institute,
Ministry of Russian Federation on Atomic Energy,
2-nd Murinsky pr. 28, St.-Petersburg, 194 021 Russia

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Introduction

In the process of nuclear power plant operation, the gas-aerosol discharges to the environment should proceed within permissible limits. The objective of this study is to analyze long-term data concerning radionuclide contamination of the ground-level air and to study its regularities. A thorough knowledge of long-term radionuclide regularities in the ground-level air near NPP are of principle importance for the countermeasure planning and decision making, for environmental impact assessment and for optimization of radiological monitoring structure.

Methods

The present investigations are concerned with the catchment basin of the rivers falling into the Koporskaya Bay – a marine cooling water body of the Leningrad NPP, at a distance of 85 km from Saint-Petersburg. The 30-km zone around the Leningrad NPP, near Sosnovy Bor, is part of the area being monitored (Fig. 1). There is a set of nuclear facilities within the area under monitoring. They are: the Leningrad NPP with four RBMK-type reactors (4 GW total electric power), LSK "Radon" – the north-western regional storage facility for low- and intermediate level radioactive wastes, and North-Western Scientific and Industrial Center of Nuclear Energy, now developed on the basis of Scientific and Research Technological Institute (Fig. 1). The LNPP gas-aerosol discharges inflow into the atmosphere via two ventilating stacks, 150 and 100 m heigh. There are 10 sampling stations of the ground-level air, snow, fallout, soil, and other terrestrial natural objects within a 35-km radius from NPP. The high-volume air samplers weekly collect ground-level air aerosols onto

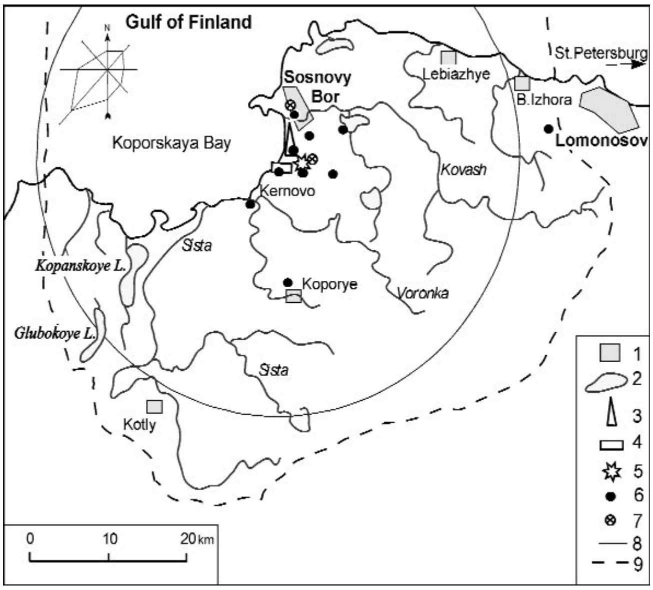

Fig. 1. The scheme of radiological monitoring in the area of Sosnovy Bor nuclear facilities at the Russian coast of the Baltic Sea; 1 – settlements, 2 – lakes, 3 – LNPP, 4 – Scientific-Research Technological Institute, 5 – Radioactive waste storage LSK “Radon”, 6 – air and terrestrial ecosystem sampling, 7 – air samplers “the Hunter” JL-150, 8 – 30-km zone, 9 – area under monitoring.

Petrianov filters at a height of 2.5 m over the ground surface. Two of the air samplers are JL-150 “The Hunter” with Whatman particle filters. These air samplers were received from Finland as a technical assistance at the end of 1999. The others are air samplers FVU (with Petrianov’s filters) manufactured in Russia. In the energy range 100–2000 keV, the detection limit of radionuclides in the air was 1.0×10^{-6} Bq/m^3, the radionuclide concentrations error varied from 5 to 30% at a confidence level of $P=0.95$.

There are defined procedures of inside and outside control of the results for quality assurance. Correctness of measurement of samples is checked regularly by using reference sources. Parallel samples are analyzed, if necessary. The measurements of background are carrying out at least monthly. One of the most important aspects of quality assurance is the participation in international and interlaboratory calibrations.

In this paper, the regularities of artificial radionuclides (^{137,134}Cs, ^{131}I, ^{60}Co, ^{54}Mn, ^{51}Cr) detection frequency in ground-level air at the Leningrad NPP industrial site were added to those received earlier [1]. The detection frequency is the ratio of the number of samples, in which a specific radionuclide has been detected, to the total number of the samples of the same sort (i.e. aerosols) collected during one year at a specific sampling station. Long-term weekly nuclide concentration time series were examined by the correlation analysis. The seasonal fluctuations of ^{7}Be concentrations in the ground-level air were examined by harmonic (Fourier) analysis.

Results and discussion

We consider the dynamic characteristics of ground-level air radionuclide contamination, such as the detection frequency of radionuclides and their concentrations in the air at the NPP site, to be an important criterion of stability characteristics of NPP equipment under its long-term operation. It was shown that the activated corrosion products (^{54}Mn, ^{60}Co) and aerosols of ^{131}I are present almost constantly in the air within a 3-km radius from the Leningrad NPP (in 60–90% of the samples) at routine NPP operation. Their averaged multiyear concentrations, as well as those of ^{137}Cs, in the air of the area under monitoring are extremely low (hundreds of thousands times lower than the permissible ones) and vary from microbecquerels to tens of microbecquerels in cubic meter during the LNPP normal operation.

The results of long-term monitoring of ground-level air showed that the concentrations of radionuclides in the air of the town Sosnovy Bor, which is located 5 km from LNPP, under the prevailing wind direction amounted to 10–30% of their concentrations in the air of the LNPP site at the same time period. This result is of particular importance and can be used in the case of countermeasure decision making. Steady levels of ^{131}I (aerosols) and corrosion products (^{54}Mn, ^{60}Co) in the air of the Sosnovy Bor town were in the range of $(2.0\div20.0)\times10^{-6}$ Bq/m^3.

In our previous publication [1] it was shown that from the beginning of the first LNPP unit large-scale overhaul and its reconstruction (from 1989 till 1991), the detection frequency of ^{131}I in the air of the Leningrad NPP industrial site sharply decreased. On the contrary, an increase of the detection frequency of fission (^{131}I) and neutron activation (^{60}Co, ^{54}Mn) products in the air occurred when LNPP-1 was put into operation on the overhaul completion (since 1992) (Fig. 2). The most probable reasons for this fact were the ageing of main technological equipment and the use of spent elements of the first NPP units (1973, 1975). Air monitoring data, received after publication [1], showed that the detection frequency of ^{131}I aerosol in the ground-level air at the Leningrad NPP site decreased from 60% in 1997 up to 40% in 1998, and up to 13% in 1999 (Fig. 2). Simultaneously, maximum concentrations of ^{137}Cs decreased tenfold (from 920×10^{-6} Bq/m^3 in 1998 up to 85×10^{-6} Bq/m^3 in 1999). The detection frequency of ^{60}Co in the ground-level air in 1997–1999 aggregate about 75% (Fig. 2). These results suggest high effectiveness of the actions taken to improve hermetic sealing of equipment at the LNPP in 1998–1999. These results could be considered as a preliminary information about the qualitative changes in NPP functioning under its long-term normal operation.

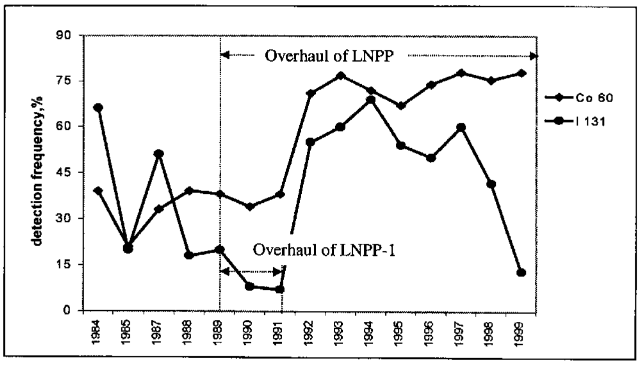

Fig. 2. Detection frequency of ^{60}Co and ^{131}I in the ground-level air at the Leningrad NPP industrial site.

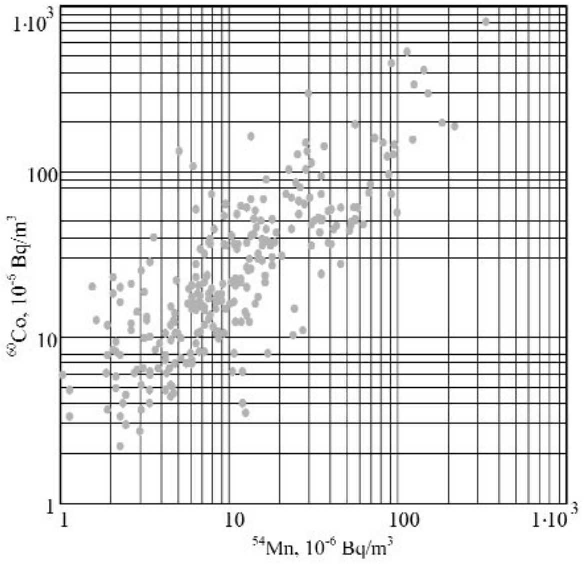

Fig. 3. 1983–1985, 1987–1999. Correlation of ^{60}Co (Y-coordinate) and ^{54}Mn (X-coordinate). R_{60-54}=0.79.

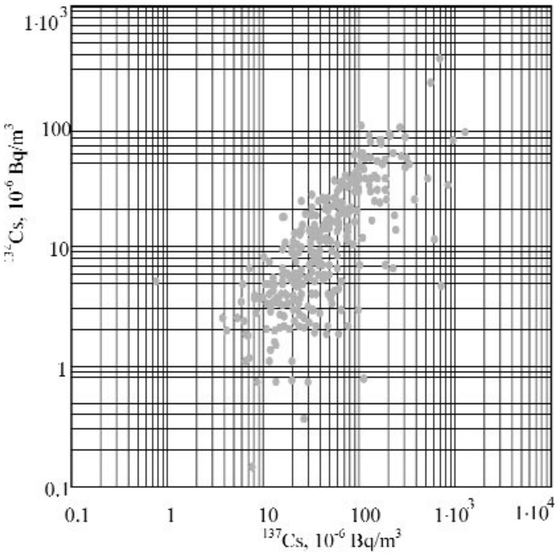

Fig. 4. 1983–1985, 1987–1999. Correlation of ^{134}Cs (Y-coordinate) and ^{137}Cs (X-coordinate). $R_{134-137}$=0.36.

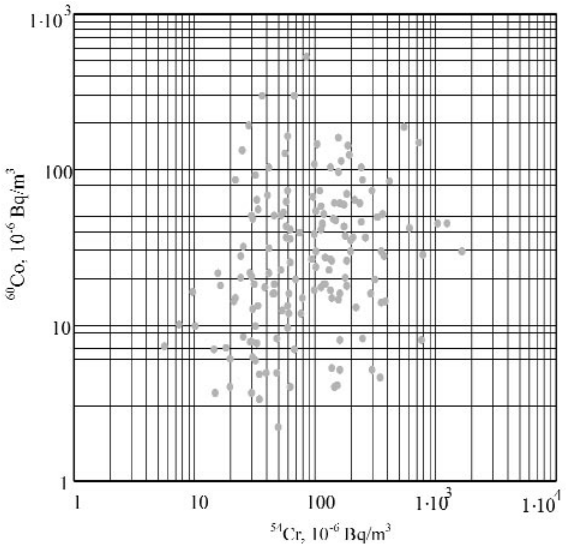

Fig. 5. 1983–1985, 1987–1999. Correlation of ^{60}Co (Y-coordinate) and ^{51}Cr (X-coordinate). R_{60-51}=0.14.

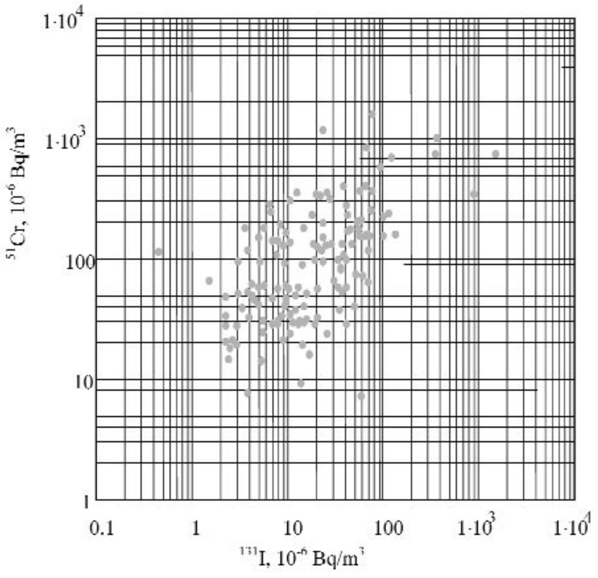

Fig. 6. 1983–1985, 1987–1999. Correlation of ^{51}Cr (Y-coordinate) and ^{131}I (X-coordinate). R_{51-131}=0.30.

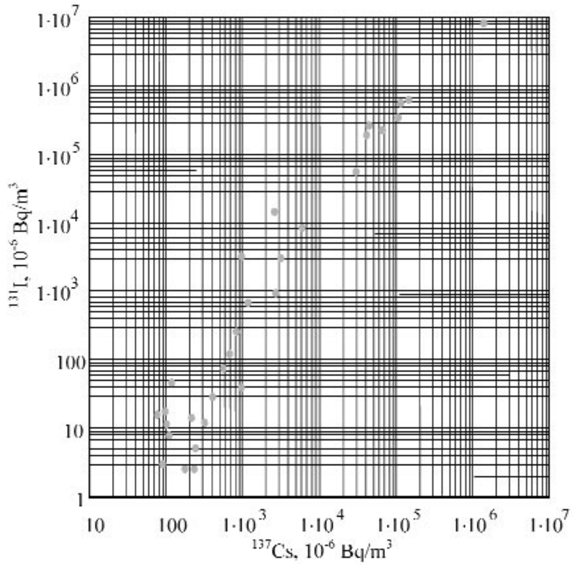

Fig. 7. 26.04–31.12.1986. Correlation of ^{131}I (Y-coordinate) and ^{137}Cs (X-coordinate). $R_{131-137}$=0.999.

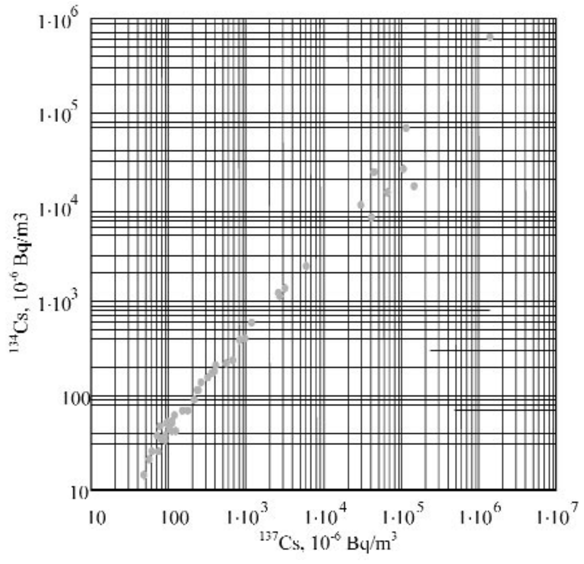

Fig. 8. 26.04–31.12.1986. Correlation of ^{134}Cs (Y-coordinate) and ^{137}Cs (X-coordinate). $R_{134-137}$=0.995.

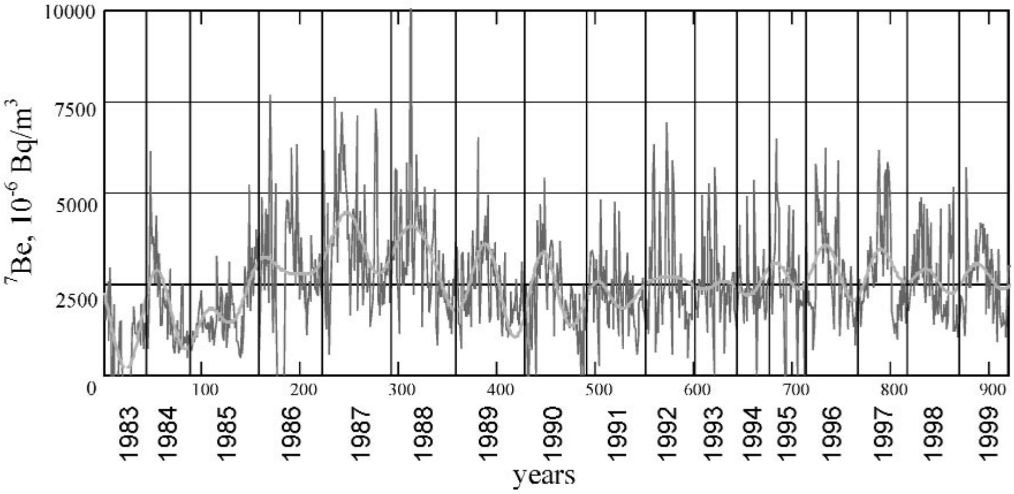

Fig. 9. 1983–1999. Fourier analysis of cosmogenous ^{7}Be in ground-level air at LNPP site. Seasonal cycle.

The correlation analysis was carried out for pairs of nuclides in ground-level air separately for two periods, one of which was (1983–1985, 1987–1999aa), another period started on April, 26, 1986, and finalized in December, 31, 1986. The first one is the period of minimum global fallout after nuclear testing, and is the starting point for LNPP operation at project capacity (1982). The second one (1986) is the period of heavy fallout due to transboundary radionuclide air transportation from the region of the Chernobyl accident.

For the first period the results showed a tight correlation between ^{60}Co and ^{54}Mn with coefficient R_{60-54}=0.79 (Fig. 3). Both of them, evidently, are from the same source, which could be pipelines of the power installation. ^{134}Cs and ^{137}Cs have a weak correlation coefficient $R_{134-137}$=0.36 (Fig. 4). So, a small portion of ^{137}Cs, about 30%, seems to have nuclear fuel nature, otherwise it ought to be strongly combined with ^{134}Cs. The rest of ^{137}Cs in the ground-level air at NPP site probably is due to discharges from the radioactive waste treatment and storage facility LSK “Radon” (Fig. 1), and to resuspention from upper layer of the ground after the Chernobyl accident and nuclear testing.

Such pairs as ^{131}I–^{137}Cs, ^{131}I–^{134}Cs have a weak and too close to each other correlation coefficient varying from 0.22 to 0.32. May be, it is connected with the different aerosol chemistry of iodine and ceasium and, thus, with different ways of activity in the atmosphere. It appears that ^{51}Cr, being the corrosion product, is initiated under the processes other than those for ^{60}Co and ^{54}Mn. The correlation coefficient for ^{51}Cr and ^{60}Co is too low and amounts only to 0.14 (Fig. 5). On the other hand, the correlation coefficient for ^{51}Cr and ^{131}I is close to the pairs ^{131}I–^{137}Cs and ^{131}I–^{134}Cs and amounts to 0.30 (Fig. 6). So, ^{51}Cr in the ground-level air at the NPP site appears to have nuclear fuel as the source. ^{131}I and ^{60}Co, as expected, have no correlation (R_{60-131}=0.086).

For the second period, i.e. the year of Chernobyl accident, we received for the Leningrad NPP site ground-level air the strongest correlation coefficient, estimated as 0.999 between ^{131}I and ^{137}Cs (Fig. 7), and estimated as 0.995 between ^{134}Cs and ^{137}Cs (Fig. 8). Analysis for other pairs of nuclides for this period is forcasted. The results received are not unexpected. But one can see (Fig. 7), that the curve consists of two lines. Probably, at the range of high nuclide concentrations ($>3\times10^3$) we see the data of the first initial stage of the Chernobyl accident. In the range below ($<3\times10^3$), the data are either of later time or transported via another atmospheric trajectory. Fig. 8 shows a straight line for ^{134}Cs and ^{137}Cs in the ground-level air close to the Leningrad NPP site for the accidental release from the Chernobyl reactor.

The analysis showed a close correlation of cosmogenous ^{7}Be in the air of different sampling stations within an LNPP 30-km radius. The seasonal fluctuations of its concentrations with a maximum in the spring-summer period are shown in Fig. 9. These results indirectly support the accuracy of the methods used for sampling and measurements of radioactive aerosol.

References

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