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IDENTIFICATION OF THE MAIN DOSE-FORMING RADIONUCLIDES IN NPP EMISSIONS

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The results of an analysis of gas-aerosol emissions from the ventilation tubes of European NPP are presented. An assessment is made of the contribution of the controlled radionuclides in the exposure of the public to the radiation from NPP emissions of five types of reactor facilities: AGR, BWR, LWGR, PWR, and CANDU. The radionuclides forming the 95% dose in the critical group of the population are determined for each type of NPP.

The main dose-forming radionuclides must be taken into account in order to determine the impact of radiation on man and the environment [1]. They are not known at present. One reason for the impossibility of picking them is that only a limited number of radionuclides are controlled in NPP emissions. In Russia, the emissions of seven radionuclides – ^{134,137}Cs, ^{58,60}Co, ^{51}Cr, ^{54}Mn, and ^{131}I – and the total β-activity and total emissions of inert radioactive gases [2] are controlled.

An electronic database on the radioactive emissions and discharges from NPP with different types of reactors has been developed in European countries [3]. The content of 101 radionuclides and four total indices are monitored: β-, α-activity, inert radioactive gases, and iodine isotopes (Table 1). Analysis of these data permits determining the contribution of each radionuclide to dose formation and formulating a list of radionuclides for each type of reactor that determine the 95% contribution the effective dose for the public. The results of ranking the radionuclides in the emissions from European NPP according to their contribution to the radiation exposure of the public are presented below.

The contribution of the controlled radionuclides was determined for the five most common types of reactors used in NPP: water moderated and cooled power reactors PWR, boiling water BWR, advanced gas-cooled AGR, high-power channel reactors LWGR, and heavy-water CANDU. The conditions for the formation of the radiation exposure – the relief, half-life structure, and food ration in the general population in the action zone of the emissions and the meteorological conditions – are assumed to be the same. The effective emission height in the calculation was 120 m. The activity of the radionuclides entering the atmosphere was averaged over 10 years of observations at NPP and led to the number of reactors in the sample. Thus, the average yearly activity of the emissions of each radionuclide for one reactor was obtained.

Five samples for the emissions of five types of reactors over the last 10 years of operation were formed in order to determine the dose loads on the general population: 19 NPP with PWR in France, three NPP with BWR in Sweden, eight NPP with AGR in Great Britain, one NPP with LWGR in Lithuania, and one NPP with CANDU in Rumania. Each sample included radionuclides which are monitored in at least 50% of the NPP (Table 2).

The CAP-88 PC 4.0.1.17 software package was used to determine the effective dose for the general population [4]. Three metrological files, which characterize the weather conditions with different combinations of stability categories in terms of Pasquill's classification, were picked in a random fashion for the calculation [5]. The results of the dose calculations

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TABLE 1. Controlled Parameters in the Emissions from European NPP

Country	Number of NPP	Number of controlled parameters (activity of individual nuclides or total activity of a group of nuclides)
Belgium	2	4
Bulgaria	1	49
Great Britain	9	11
Hungary	1	42
Germany	8	41
Spain	5	54
Netherlands	1	16
Rumania	1	31
Slovakia	2	48
Slovenia	1	34
Finland	2	42
France	20	43
Czech Republic	2	40
Sweden	4	93

TABLE 2. Monitored Radionuclides in Emissions from at Least Half of the European NPP

Radionuclide	Fraction of NPP, %
^{3}H	100
^{14}C, ^{131}I	93
^{41}Ar, ^{60}Co, ^{137}Cs, ^{133}Xe	86
^{58}Co, ^{51}Cr, ^{134}Cs, ^{59}Fe, ^{85}Kr, ^{88}Kr, ^{54}Mn. ^{95}Nb, ^{135}Xe, ^{95}Zr	79
^{85m} Kr, ^{87}Kr, ^{124}Sb, ^{90}Sr	71
^{110m}Ag, ^{141}Ce, ^{144}Ce, ^{133}I, ^{103}Ru, ^{125}Sb, ^{133m}Xe, ^{135m}Xe	64
^{140}Ba, ^{89}Sr, ^{131m} Xe, ^{138}Xe, ^{65}Zn	57
^{57}Co, ^{132}I	50

were averaged over three meteorological data sets. The dose in the critical population group was determined for locations at distances 3, 10, and 30 km from the emission source. The effective dose at the indicated distances was averaged over 16 rhumbs of the propagation direction of the emissions (Table 3).

The main radionuclides contributing >95% to the early effective dose from atmospheric gas-aerosol emissions were determined for each type of reactor:

- advanced gas-cooled AGR – of the seven controlled, %: ^{14}C 61, ^{35}S 24, ^{41}Ar 12;
- boiling water BWR from 29 controlled, %: ^{88}Kr+^{88}Rb 40,^{14}C 32,^{87}Kr 13,^{135}Xe 8, ^{60}Co 2, ^{131}I 1;
- high capacity channel LWGR from 21 controlled, %: ^{41}Ar 30, ^{90}Sr+^{90}Y 18, ^{137}Cs + ^{137m}Ba 17, ^{60}Co 12, ^{88}Kr + ^{88}Rb 6, ^{14}C 5, ^{131}I 1.5;
- water moderated and cooled power PWR from 27 controlled, %: ^{14}C 95, ^{3}H 3; and
- heavy-water CANDU from 27 controlled, %: ^{3}H 86,^{14}C 12.

TABLE 3. Average Yearly Effective Dose of Gas-Aerosol Emission from NPP with Different Types of Reactors into the Atmosphere, μSv/yr

Reactor type	Distance from emission source, km		
	3	10	30
AGR	0.14	$8.1 \cdot 10^{-2}$	$5.4 \cdot 10^{-2}$
BWR	0.36	0.14	$5.2 \cdot 10^{-2}$
LWGR	$5.2 \cdot 10^{-2}$	$2.1 \cdot 10^{-2}$	$9.6 \cdot 10^{-3}$
PWR	$4.1 \cdot 10^{-2}$	$2.4 \cdot 10^{-2}$	$1.6 \cdot 10^{-2}$
CANDU	0.27	0.15	$9.4 \cdot 10^{-2}$

TABLE 4. Contribution of Radionuclides Controlled in Domestic NPP to the Radiation Exposure Dose, %

Radionuclide	RBMK	VVER
^{137}Cs	10.6	0.35
^{134}Cs	0.45	0.19
^{60}Co	12.4	0.26
^{58}Co	0.008	0.01
^{51}Cr	0.001	0.0001
^{54}Mn	0.31	0.001
^{131}I	1.67	0.004
Inert radioactive gases	50.07	1.49
Total	75.51	2.32

The radionuclides forming >95% of the effective radiation exposure dose were recommended by the European commission for inclusion in the radiation monitoring program [6]. The calculations showed that for equal conditions the NPP with PWR and LWGR have the lowest radiation impact on the population.

The computational results for the main dose forming radionuclides in the emissions from NPP with PWR and LWGR were used to determine the contribution of gas-aerosol emissions from domestic NPP with VVER and RBMK to the radiation exposure of the critical population group (Table 4). On the basis of the data obtained, it can be supposed that the controlled radionuclides in NPP with RBMK reflect the radiation exposure of the critical population group and for NPP with VVER they determine only a very small fraction.

The determined contribution of radionuclides to the radiation exposure of the critical population group from gas-aerosol emissions from NPP characterizes the modeled situation for the formation of the radiation impact. The real conditions for the propagation of radionuclides into the environment and the radiation exposure of the population can differ strongly for each particular plant location. Taking account of the meteorological data, landscape features of the region of the NPP, the main characteristics of the emission sources, the yearly emissions of radionuclides, half-life structure, and food ration of the population in the region of a particular NPP can strongly affect the quantitative contribution of each radionuclide to the radiation exposure of the critical population group.

For equal conditions of radiation impact of gas-aerosol emissions from different reactors of NPP, the dose-forming radionuclides differ considerably. For all NPP, the emissions of ^{14}C into the atmosphere make a significant contribution to the radiation exposure: 5% for RBMK and up to 95% for VVER. The radioactive inert gases dominate in the emissions from advanced gas-cooled reactors, boiling water reactors, and channel reactors of high capacity. Only the atmospheric emissions of ^{35}S from the advances gas-cooled reactors make a large contribution (24%) to the radiation exposure of the population. The tritium emissions from heavy-water nuclear reactors form the main dose for the critical population group.

The ranking by contribution to the radiation exposure for the population makes it possible to define the main dose-forming radionuclides determining >95% of the effective dose for each type of reactor, including the RBMK and VVER operating in our country.

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