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Technical Note

Special monitoring results for determination of radionuclide composition of Russian NPP atmospheric releases

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ABSTRACT

Measurements of activity concentrations of radionuclides in atmospheric releases were performed in 2017–2018 at vent stacks of seven Russian nuclear power plants. The selected instruments and research methods, with detection limits significantly lower than the existing detection limit of Russian NPPs routine control, allowed to reliably determine up to 26 radionuclides. Analysis of experimental data allows to determine the list of radionuclides for calculation the effective dose rates to public and the permissible annual discharge levels for each Russian NPP. Radiocarbon is determined as major contributor for the dose from the atmospheric releases of LWGR reactors – up to 98% for EGP-6 and RBMK-1000 (Smolensk NPP) reactors. For PWR reactors (VVER) radionuclides contribution to the annual dose from atmospheric releases is more complicated, but, in general, dose is formed by tritium, ^{14}C and noble gases. The special monitoring results with ranking of measured radionuclides according to their contribution to the effective dose makes it possible to optimize the list of controlled radionuclides in airborne releases of Russian NPPs from 94 to 8–16 for different NPPs.

1. Introduction

Long-term monitoring of atmospheric releases from nuclear power plants allows to accumulate a large amount of information about the radionuclide structure of such releases during normal operation of NPP.

The IAEA recommends to consider 29 radionuclides for assessment of the environmental impact during the normal operation of nuclear power plants [1]. According to analysis of radioactive effluents there are more than 35 radionuclides were found in releases of NPP in Korea [2]. The approach of the European Commission suggests the organization of monitoring of 11 key radionuclides, providing control over the list of 33 radionuclides [3]. The analysis of list of detected radionuclides in European NPPs discharges during last 5 years demonstrated a common nomenclature of controlled radionuclides: 101 radionuclides and 5 integral indicators. Tritium (^{3}H) and three integral indicators (Σ iodine, $\Sigma\beta + \gamma$, Σ radioactive noble gas) are monitored in atmospheric releases at all European NPPs. Another 35 radionuclides are monitored in atmospheric releases at half or more of the European NPPs. Less than 50% of the European NPPs control the additional 67 radionuclides in their releases [4].

In the period from 2003 to 2017, Russian regulatory authorities require to control only 4 radionuclides (^{60}Co, ^{131}I, ^{134}Cs, ^{137}Cs) and 1 integral indicator (Σ radioactive noble gas) in atmospheric releases from Russian NPPs, declaring that "they form 98% of the annual effective dose to public" [5]. However, model estimates for European NPPs with different types of reactor installations showed that the main contributors to the effective dose from atmospheric releases are ^{14}C and ^{3}H [6].

The absence of tritium and carbon-14 control in the atmospheric releases of Russian nuclear power plants led to underestimation of the effective dose up to 25% for LWGR and 98% for PWR [6]. The existed methods and approaches to radioactive airborne releases control of Russian NPPs did not fit the new regulatory requirements of Russian Federation [7].

Moreover, if controlled radionuclide is absent in the atmospheric releases, the concentration corresponding to a half of the

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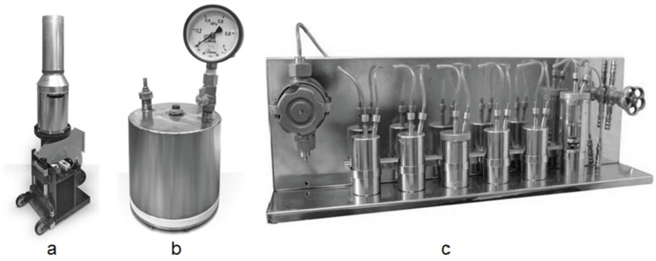

Fig. 1. Sampling methods: a) air filtering installation for radionuclides activity assessment; b) metal vessel for radioactive noble gases activity assessment; c) system of bubblers for H-13 and C-14 collection.

detection limit should be taken into account for regulation purposes [3].

The objectives of the present study in accordance with IAEA recommendations [8] are the following:

- to provide data on the discharge of radionuclides to the environment, as a basis for the estimation by predictive modelling of environmental radiation levels and activity concentrations and exposure of the public;
- to justify annual permissible release limits for atmospheric releases of radionuclides.

To achieve these goals it is necessary:

- to perform on-site measurements of the radionuclides (technologically caused by the normal operation of NPP) in released air (special monitoring with detection limits significantly lower than the existing detection limit of Russian NPPs routine control);
- to rank measured radionuclides according to the effective dose and select those, which totally contribute 99% to annual effective dose to the public.

2. Materials and methods

Assessment of the radionuclides activity concentrations in vent stacks of Russian NPPs was performed with different approaches:

- radioactive aerosols were captured with powerful air filtering installation (Fig. 1a):
  - allow to pump about 12 000 m^3 per day;
  - air flow more than 400 $m^3 \cdot h^{-1}$ through one layer of sorption-filtering material;
  - linear air flow velocity for the aerosol package up to 150 cm/s;
  - weight <45 kg;
  - dimensions, mm - 400×400×1900.
- radioactive noble gases were collected into special metal vessel under pressure of 500 kPa (Fig. 1b);
- tritium and radiocarbon were collected with special system of bubblers (air consumption ~42 dm^3/h) (Fig. 1c).

In comparison with routine control systems of radionuclides activity concentrations in Russia NPPs airborne releases, the applied special monitoring approach allow to reduce detection limit (Table 1):

- on 2–3 orders for radioactive aerosols;
- in 3–4 times for radioactive noble gases;
- in 20 times for tritium.

Achieved detection limits are significantly lower in comparison with methods established for USA NPPs in standard radiological effluent controls for PWR and BWR reactors [9,10].

Aerosol filter consisting of micro-fibrous materials was used for filtering radioactive aerosols of fission and corrosion products [11]. Special sorption and filtering material based on activated carbon was used for radioactive iodine analysis. The analysis of radionuclide concentration after sampling was performed using gamma- and beta-spectrometry.

On-site measurements of radionuclides content in NPPs airborne releases were performed for Russian NPPs with different types of reactors (Table 2). The measured values of the radionuclide activity concentrations were used to estimate contribution to the effective dose from atmospheric releases to local population (critical group). The radionuclides forming 99% of the annual effective dose were identified. Hypothetical annual dose to the public with consideration of all possible exposure pathways was estimated using recommended by regulator dose coefficients [12]. For each radionuclide, all possible ways of internal and external exposure were considered and dose functionals of the transition from radionuclides activity concentration to the effective dose from atmospheric releases to critical group were formed [8]. The main external exposure pathways considered: direct exposure from a source of ionizing radiation; exposure due to the plume of radionuclides in the atmosphere; contact exposure from radionuclides on the skin; exposure from the radionuclides deposited on the ground or on sediments. The main internal exposure pathways considered: inhalation of radionuclides in the plume; ingestion of radionuclides in food or beverages; for tritium oxide in the plume, absorption through the skin; inhalation of resuspended radionuclides.

3. Results and discussion

The radionuclides forming the annual effective dose, which were not possible to be determined by routine radiation monitoring system, have been measured by a special measuring system with a low detection level. Measurements of activity concentrations of radionuclides in atmospheric releases were performed in 2017–2018 at vent stacks of seven Russian nuclear power plants (Table 2). The sources of air releases at investigated nuclear power plants are the vent pipes of power units, radioactive waste storage facilities, spent fuel storage facilities, turbine buildings, special buildings, etc. The release of radioactive gases and aerosols, removed by ventilation systems, depends on compliance with

Table 1
Detection limits for routine NPPs and proposed control systems of radionuclides assessement.

Control indicator	Existing detection limit, Bq/m^3	Detection limit of proposed method, Bq/m^3
Radioactive aerosols (661.7 keV)	$5 \cdot 10^{-2}$	$2.5 \cdot 10^{-5}$
Radioactive iodine (364.5 keV)	$3 \cdot 10^{-2}$	$3.5 \cdot 10^{-5}$
Inert radioactive gases in the energy range from 65 keV to 3000 keV	$3 \cdot 10^{3}$	$8.0 \cdot 10^{1}$
Tritium and radiocarbon	37.0 (H-3)	3.5 (H-3)
	0.4 (C-14)	7.0 (C-14)
	sample volume 1 m^3 during 7 days	sample volume 1 m^3 during one day

Table 2
Objects of survey.

№	Nuclear power plant	Type of reactor
Water-Water Energetic Reactor (VVER)		
1	Rostov	VVER-1000
2	Kola	VVER-440
3	Kalinin	VVER-1000
4	Novovoronezh	VVER-440,-1000
Light Water Graphite Reactor (LWGR)		
5	Bilibino	EGP-6
6	Smolensk	RBMK-1000
7	Kursk	RBMK-1000

ventilation modes, the efficiency of filters, the tightness of equipment and operating mode. The measurements were carried out under the standard operating conditions of the NPPs.

Maximum values of radionuclides concentration in Russian NPPs airborne releases are shown in Table 3. Empty boxes in Table 3 mean that the activity concentration of radionuclide was below the detection limit. According to the study, from seven to 26 radionuclides were determined in discharges to atmospheric air. Contribution of activity of selected radionuclides to total activity has similarities to each group of NPP. Common characteristic of the releases is the predominance of noble gases in the total activity (Fig. 2). Another common characteristic is the fact, that activity of radionuclides except noble gases is formed by two radionuclides (^{3}H and ^{14}C) for 99.9%.

This list of controlled radionuclides could be optimized in terms of effective dose according to Russian Radiation Safety Standards. Discharge limits are set for each source, which total emission creates an individual annual effective dose more than 10 μSv without taking into account the atmospheric dispersion. At the same time, the total contribution of controlled radionuclides to the annual effective dose, created by the discharges of this source, is at least 99%. This approach makes it possible to optimize the list of controlled radionuclides in atmospheric releases. On the other hand, the approach of the European Commission suggests the organization of monitoring of 11 key radionuclides as mentioned

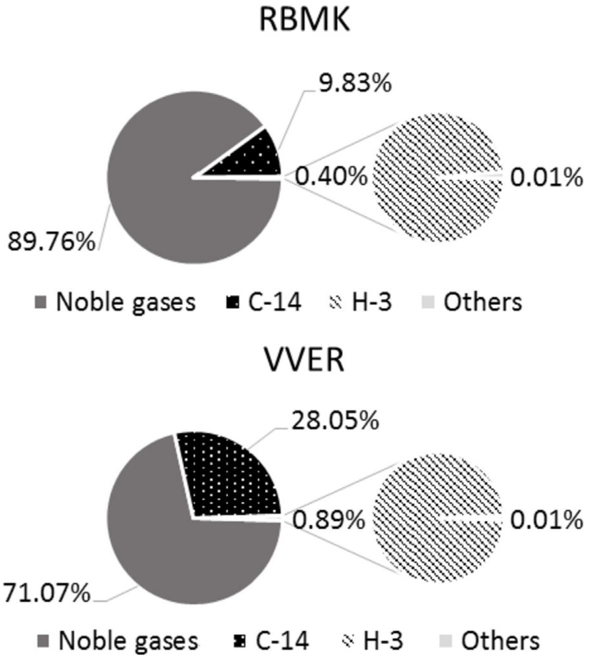

Fig. 2. The contribution of noble gases in the total normalized discharge activity.

Table 3
Maximum value of radionuclides concentration in Russian NPPs airborne releases (Bq/m^{3}).

R/n	Nuclear power plant						
	Rostov	Kola	Kalinin	Novovoronezh	Bilibino	Smolensk	Kursk
H-3	$5{,}3 \cdot 10^{3}$	$9{,}5 \cdot 10^{2}$	$2{,}8 \cdot 10^{3}$	$1{,}2 \cdot 10^{3}$	$4{,}3 \cdot 10^{3}$	$1{,}6 \cdot 10^{2}$	$3{,}8 \cdot 10^{2}$
C-14	$1{,}0 \cdot 10^{2}$	$1{,}9 \cdot 10^{1}$	$1{,}8 \cdot 10^{2}$	$4{,}0 \cdot 10^{1}$	$4{,}2 \cdot 10^{3}$	$7{,}5 \cdot 10^{2}$	$1{,}5 \cdot 10^{4}$
Ar-41	$7{,}7 \cdot 10^{3}$	$5{,}0 \cdot 10^{3}$	$2{,}1 \cdot 10^{3}$	$6{,}1 \cdot 10^{2}$	$7{,}0 \cdot 10^{2}$	$6{,}5 \cdot 10^{3}$	$9{,}9 \cdot 10^{3}$
Kr-85 m						$2{,}1 \cdot 10^{3}$	$7{,}5 \cdot 10^{3}$
Kr-87						$7{,}1 \cdot 10^{3}$	$6{,}1 \cdot 10^{3}$
Kr-88						$6{,}9 \cdot 10^{3}$	$9{,}5 \cdot 10^{3}$
Xe-133		$7{,}2 \cdot 10^{2}$	$5{,}9 \cdot 10^{2}$	$3{,}2 \cdot 10^{2}$		$5{,}3 \cdot 10^{3}$	$2{,}3 \cdot 10^{4}$
Xe-135		$1{,}1 \cdot 10^{3}$		$5{,}6 \cdot 10^{1}$		$7{,}8 \cdot 10^{3}$	$5{,}6 \cdot 10^{4}$
Xe-138							$1{,}2 \cdot 10^{3}$
Na-24						$4{,}2 \cdot 10^{-2}$	$3{,}0 \cdot 10^{0}$
Cr-51			$3{,}0 \cdot 10^{-2}$			$8{,}9 \cdot 10^{-3}$	$3{,}6 \cdot 10^{-1}$
Mn-54		$1{,}2 \cdot 10^{-2}$	$1{,}8 \cdot 10^{-3}$	$8{,}4 \cdot 10^{-2}$	$3{,}0 \cdot 10^{-3}$	$3{,}9 \cdot 10^{-3}$	$1{,}6 \cdot 10^{-1}$
Co-58		$7{,}4 \cdot 10^{-3}$	$3{,}0 \cdot 10^{-3}$	$2{,}9 \cdot 10^{-1}$			$7{,}3 \cdot 10^{-3}$
Fe-59						$5{,}5 \cdot 10^{-4}$	$2{,}3 \cdot 10^{-2}$
Co-60	$1{,}5 \cdot 10^{-3}$	$1{,}4 \cdot 10^{-2}$	$5{,}7 \cdot 10^{-3}$	$8{,}5 \cdot 10^{-3}$	$12{,}5 \cdot 10^{-2}$	$5{,}9 \cdot 10^{-3}$	$9{,}8 \cdot 10^{-1}$
Ag-110 m		$4{,}3 \cdot 10^{-2}$					
Sb-124		$5{,}5 \cdot 10^{-3}$					
Sr-90	$0{,}8 \cdot 10^{-5}$			$1{,}1 \cdot 10^{-3}$	$5{,}0 \cdot 10^{-5}$		$2{,}8 \cdot 10^{-3}$
Nb-95		$8{,}4 \cdot 10^{-3}$				$2{,}2 \cdot 10^{-3}$	$1{,}2 \cdot 10^{-2}$
Zr-95						$1{,}8 \cdot 10^{-3}$	$3{,}6 \cdot 10^{-3}$
Tc-99 m						$5{,}1 \cdot 10^{-3}$	$3{,}2 \cdot 10^{-2}$
Ru-103		$2{,}9 \cdot 10^{-3}$					
I-131				$3{,}5 \cdot 10^{-4}$		$3{,}8 \cdot 10^{-3}$	$4{,}9 \cdot 10^{-1}$
I-133				$9{,}8 \cdot 10^{-4}$		$2{,}3 \cdot 10^{-2}$	$9{,}7 \cdot 10^{-1}$
Cs-134	$2{,}9 \cdot 10^{-3}$	$2{,}1 \cdot 10^{-3}$	$7{,}1 \cdot 10^{-3}$	$8{,}9 \cdot 10^{-3}$			$9{,}7 \cdot 10^{-3}$
Cs-137	$7{,}8 \cdot 10^{-3}$	$2{,}7 \cdot 10^{-2}$	$5{,}0 \cdot 10^{-2}$	$1{,}5 \cdot 10^{-2}$	$2{,}5 \cdot 10^{-4}$	$7{,}0 \cdot 10^{-4}$	$1{,}7 \cdot 10^{-1}$
Cs-138					$2{,}5 \cdot 10^{2}$		
Ba-140							$9{,}9 \cdot 10^{-1}$
La-140							$6{,}7 \cdot 10^{-1}$
Ce-144		$5{,}6 \cdot 10^{-3}$					
Eu-152				$2{,}5 \cdot 10^{-4}$			

Table 4
Dose forming radionuclides structure in releases (%) (values are given for radionuclides with a contribution > 0.1%).

№	R/n	Nuclear power plant, release source (if specified)															
		Rostov			Kola		Kalinin			Novovoronezh		Bilibino		Smolensk		Kursk	
		VT-1	VT-2	VT-3	VT-1	VT-2	VT-1	VT-2	VT-3	VT-3,4	VT-5	VT-1	VT-2	VT-1	VT-2	VT-1	VT-2
1	H-3	52.2	19.2	28.0	34.2	29.0	21.3	1.9	15.1	11.1	39.9	2.1	1.3	0.2	0.2	0.4	1.6
2	C-14	15.3	76.2	17.5	39.6	2.0	68.0	89.9	82.7	10.1	42.0	97.0	97.5	77.7	98.0	60.2	8.9
3	Ar-41	29.4	1.9	48.9	1.9	40.3	5.3	0.2	0.2	4.1	6.3	0.2	0.7	5.9	1.1	7.2	11.3
4	Kr-85 m			0.1	0.1	1.1								0.2		0.9	0.3
5	Kr-87	0.6	0.8	1.6	2.8	2.5	0.7	0.3		3.0	1.5			3.9	0.1	3.8	0.3
6	Kr-88	2.1	1.7	3.5	6.9	9.9	2.2	1.1	1.0	9.9	5.9	0.1		10.6	0.4	15.7	5.1
7	Xe-133					0.3				0.7						0.5	
8	Xe-135			0.1	0.2	5.1				0.7				1.1		11.2	
9	Xe-138						0.4	0.2	0.2	1.7	1.2						
10	Mn-54				0.2					6.8							0.9
11	Co-58									6.2							
12	Co-60				6.9	0.8	0.9	0.5	0.1	8.4	0.7	0.6	0.4	0.2			48.3
13	Ag-110 m				3.0	2.4				10.9							
14	Sr-90									0.6							
15	Cs-134					0.2		0.5		6.7							2.0
16	Cs-137			0.1	3.6	6.2	0.7	5.1	0.2	16.6	1.3					0.1	20.9

above. It can be assumed that the key radionuclides for European NPP built after Soviet design are applicable for VVER nuclear power operated in Russia [13]. However, the practical confirmation of this assumption is required.

The technogenic radionuclides were ranked according to their contribution in the annual dose (Table 4). According to Table 4 dose-forming radionuclides structure in discharges is specific to each of the groups, NPPs and even discharge sources. Radiocarbon is major contributor for the dose from the atmospheric releases of LWGR reactors – up to 98% for EGP-6 and RBMK-1000 (Smolensk NPP) reactors. For PWR reactors (VVER) radionuclides contribution to the annual dose from atmospheric releases is more complicated, but, in general, dose is formed by tritium, ^{14}C and noble gases. For all Russian NPPs, except Kursk NPP (RBMK-1000 reactor), releases of ^{60}Co, ^{134}Cs, ^{137}Cs and other aerosols do not provide any considerable contribution for the annual dose.

As can be seen from Table 4, ranking of measured radionuclides according to their contribution to the effective dose makes it possible to optimize the list of controlled radionuclides in airborne releases of Russian NPPs from 94 to 8–16 for different nuclear power plants (forming 99% of the annual effective dose). In addition, this list should include radionuclides, which should be controlled in the atmospheric releases of Russian NPPs according to Radiation Safety Norms of Russia, 2003 (^{60}Co, ^{131}I, ^{134}Cs, ^{137}Cs and radioactive noble gases). The list of radionuclides, which was obtained as a result of the study, is in a good agreement with main radionuclides contributing >95% to effective dose from atmospheric gas-aerosol releases of NPPs with different types of reactors in European countries [6].

4. Conclusion

Measurements of activity concentrations of radionuclides in atmospheric releases were performed in 2017–2018 at vent stacks of most Russian nuclear power plants. Radionuclide composition of radionuclides in atmospheric releases from Russian NPPs was studied to determine the main radionuclides forming 99% of annual individual dose for the critical group of population specific for each NPP. The selected instruments and research methods, with detection limits significantly lower than the existing detection limit of Russian NPPs allowed to reliably determine up to 26 radionuclides. Less than 16 of the detected radionuclides form at least 99% of annual individual dose for the critical group of population.

About 99.9% of activity of airborne releases is formed by noble gases and two radionuclides (^{3}H and ^{14}C). Radiocarbon is major contributor for the dose from the atmospheric releases of LWGR reactors. For PWR reactors (VVER) radionuclides contribution to the annual dose from atmospheric releases is more complicated, but, in general, dose is formed by tritium, ^{14}C and noble gases.

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