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Radioactive Aerosols within Conditions of the New Safe Confinement in 2017–2020

Ключові слова:

New Safe Confinement, Shelter object, aerosols, volumetric activity, density of radionuclide deposition, activity median aerodynamic diameter

The results of monitored behavior of radioactive aerosols within the conditions of the New Safe Confinement (NSC) in 2017–2020 are presented. Maximum “unorganized” flux of beta-emitting products of the Chornobyl accident from the Shelter object through the process openings and leakages of light roofing in the NSC basic volume observed in 2017 made 7.9 kBq/(m²·day). Mean annual density of radionuclide deposition onto the light roofing during four years has been changing within the range of 1.7–2.2 kBq/(m²·day). Over these years, mean annual volumetric activity of sum of long-lived beta-emitting nuclides coming in the “Bypass” system dropped from 0.84 to 0.17 Bq/m³. Their carriers were aerosol particles with activity median aerodynamic diameter, mainly, from 4.3 to 10 μm. The data are given on radioactive contamination of near-surface air layer in the NSC basic volume.

Introduction

On November 27, 2016, the Arch of New Safe Confinement (NSC) was installed over the Shelter object. The termination of construction and sealing of the NSC enclosing perimeters essentially reduced in 2018 the impact of environmental meteorological conditions (atmosphere precipitation, temperature and wind) in the Shelter object. As a result of changed temperature and humidity mode inside the object, gradual dryout is occurring of water clusters of radioactively contaminated water in the rooms and drop in air humidity, that in its turn contributes in resuspension growth, including from the surface of fuel-containing materials (FCM).

Radioactive aerosols, being released from the Shelter object by airflows through the process openings and leakages on object’s upper marks and its light roofing, became restricted in the space under the NSC. As a result of precipitated radioactive particles being released from

the Shelter object, and the dust producible during the NSC operation, layer of surface radioactive contamination is generated on the underlying surface under the NSC.

In 2017–2020, systematic survey of “unorganized” flux of radioactive aerosols from the Shelter object [1–3] was continued. This survey initiated in 1992 remains an important source of an experimental data needed to assess radiation safety and to understand the processes occurring inside the Shelter object, and its impact on air radioactive contamination in the NSC basic volume.

This work is aimed at surveying the dynamics of radioactive aerosol situation within the conditions of the NSC — Shelter object complex in 2017–2020.

Methods and gears for sampling and measuring radioactive aerosols

To evaluate the “unorganized” flux of radioactive aerosols through the process openings and leakages

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on light roofing of the Shelter object and radionuclide deposition density, gauze accumulating plachets were used. Over the process hatches no. 7 (row $И^{+1400}$, axis 46^{+1300}), no. 10 (row $Л^{+700}$, axis 46^{+1300}), no. 13 (row $К^{+700}$, axis 48_{-700}) and no. 15 (row $Л^{+1300}$, axis 48_{-500}), four plachet holders were placed on roofing, each of them was equipped by two horizontal plachets. Among the plachets, metal sheet was located. The bottom plachet collects the particles coming from the hatch. The upper horizontal plachet is destined to monitor the density of radionuclide depositions. To retain aerosol particles, the gauze was preliminary impregnated by oil products (lythol-24 and oil-dilutant). The plachets are replaced each month.

To monitor the disperse content, aerosols were sampled from “Bypass” system through the hatch located in the room 2016/2 (Unit 3 of the Chornobyl NPP, elevation mark +45.00) (Fig. 1). Sampling of aerosol particles and their in-size classification was made with using 5-cascade impactor IBF-5K. This device makes particle gradation on five ranges of aerodynamic diameter (AD): $<0.5 \mu\text{m}$; $0.5\text{--}1.2 \mu\text{m}$; $1.2\text{--}3.7 \mu\text{m}$; $3.7\text{--}8.5 \mu\text{m}$; $8.5\text{--}17.0 \mu\text{m}$. As the fifth stage ($<0.5 \mu\text{m}$), finely dispersed filter was used, which allowed more completely catching submicron-sized aerosols. The sampling was held during 1–2 weeks.

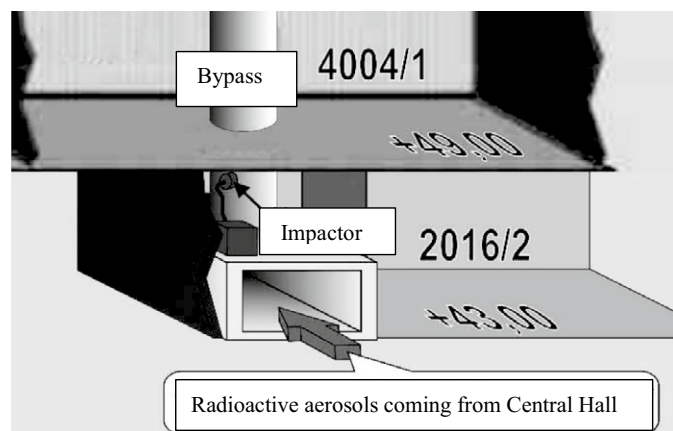


Fig. 1. Aerosol sampling by IBF-5K impactor in “Bypass” system

To determine the volumetric activity and radionuclide contents, air sampling under the NSC was carried out with using filtering and ventilation facility (FVF) “GRAD-1.8” (Fig. 2) equipped by filtering material FPP-15–1.5 with 0.56 m^2 filter square and mean daily volume of pumped air around $9 \times 10^3 \text{ m}^3/\text{day}$. The facility is located near the southern wall of the Unit 4 Turbine Hall on mark +10.00 at intersection of axis 42 and row A_{-6000} . Filter exposure duration made one week.

To determine the density of radionuclide depositions on the underlying surface, horizontal gauze plachets (HGP) were used. An exposure duration of HGP made a month. The first HGP was installed near the southern wall of the Turbine Hall at intersection of axis 42 and row A_{-6000} (see Fig. 2).

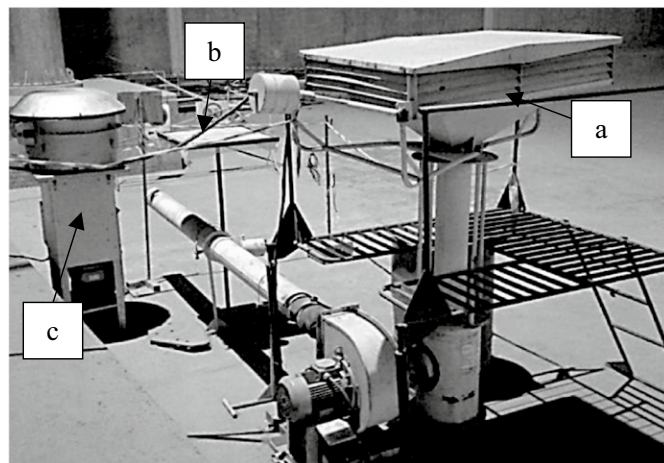


Fig. 2. Layout of sampling gear on mark +10,00: a — FVF “GRAD-1.8”; b — horizontal gauze plachet; c — “Andersen” impactor

The second HGP was placed in August 2018 near the western wall of Unit 4 (intersection of axis 59 and row III).

The disperse content of ^{137}Cs -bearing aerosol in the near-surface air layer under the NSC near the southern wall of Unit 4 Turbine Hall was sampled by way of its pumping through the impactor facility “Andersen” (model RM-10) with $1650 \text{ m}^3/\text{day}$ performance, installed in close vicinity to FVF “GRAD-1.8” (see Fig. 2). The facility makes particle sorting for five AD: $<0.8 \mu\text{m}$; $0.8\text{--}1.4 \mu\text{m}$; $1.4\text{--}2.3 \mu\text{m}$; $2.3\text{--}4.9 \mu\text{m}$; $4.9\text{--}10.0 \mu\text{m}$. As the fifth level ($<0.8 \mu\text{m}$), 600 cm^2 — square filtering material Whatman-41 is used. The sampling was carried continuously during 2 weeks.

Radionuclide activities in samples were measured at the gamma-spectrometry complex consisting of semiconductor detector GL2020R of ultrapure germanium with 500 mkm-thick beryllium window and 8192-channel amplitude pulse analyzer. The measuring range covers the energies from 10 to 1400 keV. The detector has 0.57 and 1.2 keV resolution for 122 keV gamma-quanta energies (gamma-line of ^{57}Co) and 661.6 keV (gamma-line of ^{137}Cs), accordingly. On the results of gamma-spectrometry measurements, total volumetric activity of long-lived beta-emitting nuclides ($\Sigma\beta$) — products of the Chornobyl accident was calculated, based on the ratios of radionuclides with ^{241}Am in base content of the fourth Unit fuel at measurement moment [4, 5].

Monitoring results of radioactive aerosol flux through the process openings and leakages on light roofing of the Shelter object

Fig. 3 shows the dynamics of the “unorganized” flux of aerosols bearing beta-emitting nuclides ($\Sigma\beta$) through the leakages on upper marks of the Shelter object and process openings of its light roofing in 2017–2020. The $\Sigma\beta$ includes ^{137}Cs , $^{90}\text{Sr} + ^{90}\text{Y}$, and ^{241}Pu isotopes. Activity of $^{90}\text{Sr} + ^{90}\text{Y}$ and ^{241}Pu isotopes was determined by calculation method. Some generalized survey results are presented in Tab. 1.

Based on Tab. 1 and publications [2, 3], histogram of $\Sigma\beta$ releases over 13-year period (Fig. 4) was built. As the

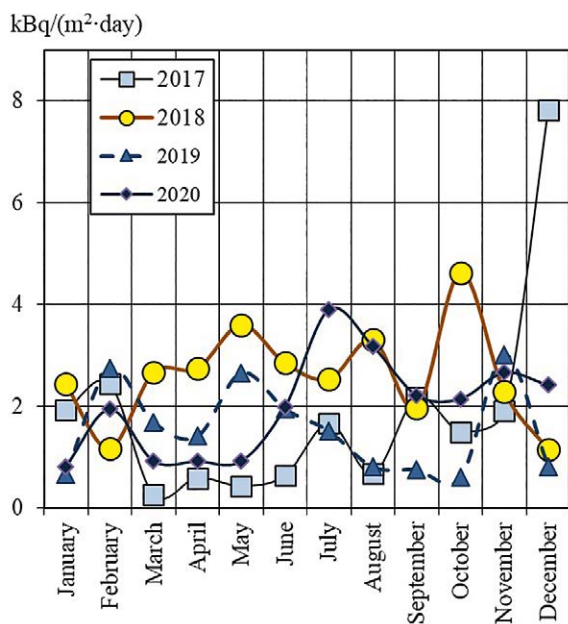


Fig. 3. Dynamics of “unorganized” flux of $\Sigma\beta$ through the leakages on upper marks of the Shelter object in 2017–2020

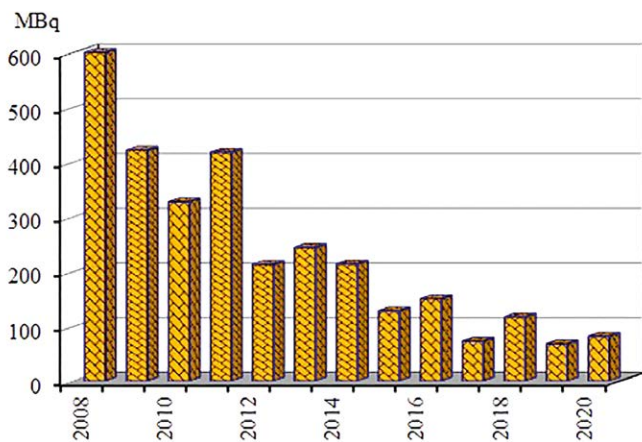


Fig. 4. Dynamics of “unorganized” release of $\Sigma\beta$ from the Shelter object in 2008–2020

Table 1. Estimates of parameters of “unorganized” flux of radioactive aerosols through the leakages on upper marks of the Shelter object in 2017–2020

Parameter	2017	2018	2019	2020
Mean annual of $\Sigma\beta$ flux, kBq/(m ² · day)	1.8	2.6	1.6	2.0
Speed variation coefficients of $\Sigma\beta$ “unorganized” flux, %	110	37	56	49
Maximum of $\Sigma\beta$ flux, kBq/(m ² · day)	7.9	4.6	3.0	3.9
“Unorganized” release of $\Sigma\beta$, MBq	73	117	68	82

figure shows, after the NSC installation, maximum $\Sigma\beta$ value 117 MBq was registered in 2018. It is provided by that year dismantling of roofing of Unit 4 Turbine Hall and measures to commission the NSC, which were accompanied by a significant dust rise and aerosol origination within the limits of space under the NSC. Minimum release value was registered in 2019.

Radionuclide depositions on the light roofing of the Shelter object are produced from settled radioactive particles being released from the object through its process openings and leakages, and deposit of dust due to resuspension during the technological activities in the space under the NSC. The dynamics of radionuclide deposition density on light roofing of the Shelter object is shown in Fig. 5. Generalized survey results are shown in Tab. 2.

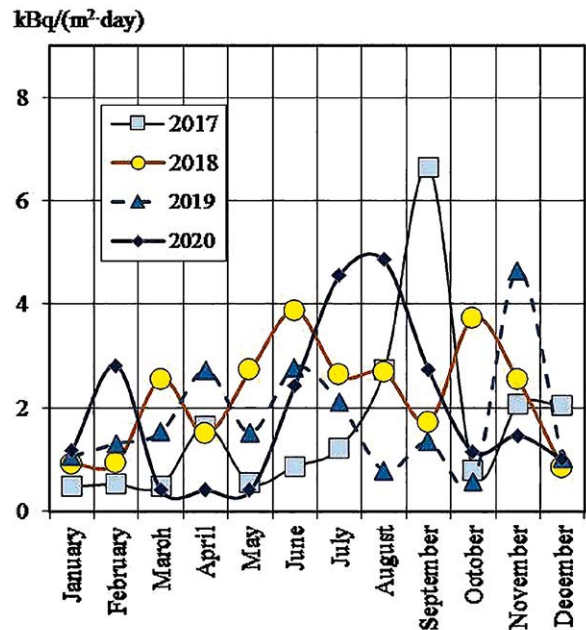


Fig. 5. Dynamics of Sb fallout density onto light roofing of the Shelter object in 2017–2020

Table 2. Estimates of parameters of density of radionuclide depositions on the light roofing of the Shelter object in 2017–2020

Parameter	2017	2018	2019	2020
Mean annual density of deposition $\Sigma\beta$, kBq/(m ² · day)	1.7	2.2	1.8	2.0
Variation coefficients of $\Sigma\beta$ deposition density, %	370	140	220	220
Maximum $\Sigma\beta$ deposition density, kBq/(m ² · day)	6.6	3.9	4.6	4.9

In 2017–2020, linear correlation coefficient between the “unorganized” flux from the Shelter object and radionuclide deposition density on its light roofing was within the range of 0.28–0.70. Maximum values of correlation coefficient were observed in 2018 and is linked to mentioned above building and assembly works in the space under the NSC.

Dispersivity of radioactive aerosols in flux from the Shelter object through the “Bypass” system

In 2017–2020, initiated in 2003 systematic survey of radioactive aerosols coming from the former Unit 4 Central Hall through the “Bypass” system into ventilation stack of the Chernobyl NPP Stage 2 [1–3, 6] was continued. The choice of “Bypass” system as a sampling place for investigation of disperse and radionuclide content of aerosol releases from the Shelter object is provided by the fact that here micro- and macro flows are coming from many rooms and corridors of the Shelter object, united in the Central Hall of emergency Unit including from those, where lava-like FCM and residues of nuclear fuel are located.

Volumetric activity of aerosols bearing the products of accident at the fourth unit Chernobyl NPP

The Fig. 6 presents the results of surveyed dynamics of $\Sigma\beta$ volumetric activity in the “Bypass” system over 2003–2020 period. As the figure demonstrates, the trend line having exponential character shows the drop in $\Sigma\beta$ volumetric activity of the releases through the “Bypass” system.

During 2017–2020, to determine volumetric activity and disperse content of radioactive aerosols in the “Bypass” system, 56 samples were taken, in which ¹³⁷Cs

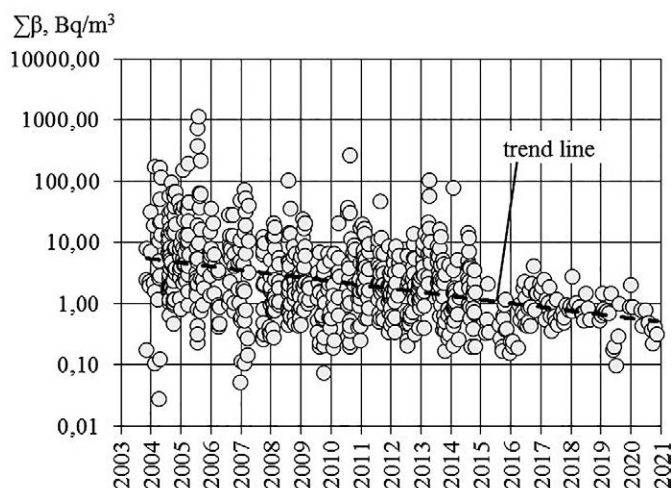


Fig. 6. Dynamics of $\Sigma\beta$ volumetric activity in the “Bypass” system of the Shelter object

volumetric activity has been varying within the range of 0.016–2.3 Bq/m³, $\Sigma\beta$ — 0.043–2.7 Bq/m³, and ²⁴¹Am — (0.58–15) · 10⁻³ Bq/m³.

The Tab. 3 shows mean annual values of ¹³⁷Cs, ²⁴¹Am and $\Sigma\beta$ volumetric activities in the “Bypass” system. This table demonstrates that radionuclide concentrations lowered during four years in 4–5 times.

Table 3. Mean annual values of ¹³⁷Cs, ²⁴¹Am and $\Sigma\beta$ volumetric activities, Bq/m³ in 2017–2020

Parameter	2017	2018	2019	2020
¹³⁷ Cs	0.51	0.53	0.33	0.10
²⁴¹ Am	6.1 · 10 ⁻³	5.8 · 10 ⁻³	3.7 · 10 ⁻³	1.6 · 10 ⁻³
$\Sigma\beta$	0.84	0.71	0.52	0.17

The dynamics of $\Sigma\beta$ volumetric activity in the “Bypass” system over 2015–2020 period is shown in Fig. 7.

In Tab. 4, mean annual values of ratios of ¹³⁷Cs/²⁴¹Am and ¹³⁷Cs/ $\Sigma\beta$ volumetric activities in the “Bypass” system, are presented.

Table 4. Mean annual values of ratios of ¹³⁷Cs/²⁴¹Am and ¹³⁷Cs/ $\Sigma\beta$ volumetric activities in 2017–2020

Parameter	2017	2018	2019	2020
¹³⁷ Cs/ ²⁴¹ Am	73	78	91	64
¹³⁷ Cs/ $\Sigma\beta$	0.59	0.57	0.63	0.58

As the table demonstrates, in 2017–2020, ¹³⁷Cs share within $\Sigma\beta$ content, being released through the “Bypass” system into atmosphere, has been remaining, practically, unchanged.

Dispersivity of radioactive aerosols. The outcomes of surveyed disperse content and concentration of radio-

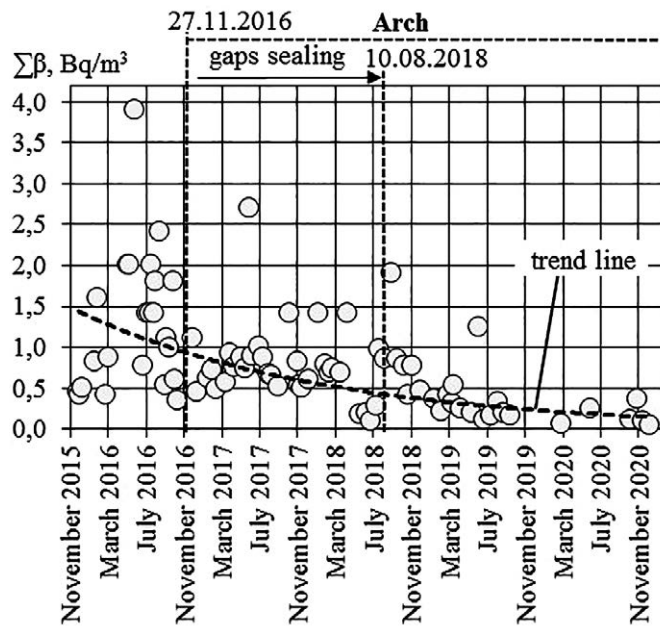


Fig. 7. Dynamics of $\Sigma\beta$ volumetric activity in the “Bypass” system in 2015–2020

active aerosols in the “Bypass” system have shown that in 2017–2020, the activity median aerodynamic diameter (AMAD) of ^{137}Cs -bearing particles had been varying from 1.6 to 14 μm (main mass of values — within the range from 5 to 10 μm), at the same time, the mean value made 7.7 μm . For comparison, in 2016 the AMAD was changing within the range of 5.7–11 μm , and mean value made 9.8 μm [6]. The analysis of all obtained data demonstrates that after the NSC Arch installation on November 27, 2016, in its design position, in the next four years, the input of large ^{137}Cs -bearing particles in volumetric activity of airflow through the “Bypass” system, decreased.

In 2017–2020, AMAD value of ^{241}Am -bearing particles in the “Bypass” system had been varying from 1.9 to 10 μm , and mean value made 7.6 μm .

Survey of airflow direction in the “Bypass” system.

In 2017–2018, survey initiated in 2015 over airflow direction in the “Bypass” system, was continued. The monitoring of airflow direction is carried out weekly, including the days of impactor IBF-5K recharge. The 2015–2016 survey demonstrated that under a wide range of meteorological conditions, air movement in the “Bypass” system not only stopped, but was headed inside the Object [6]. Airflow direction “inside” was observed also in previous years, in addition, prevailing in spring period, when a significant temperature difference occurred inside and outside the Shelter object.

In period of 2017–2020, 127 cases of airflow direction “inside” and only 40 cases “outside” were registered.

As it was highlighted in work [6], in spite of the fact that monitoring of airflow direction in the “Bypass” system had episodic character, dynamics of $\Sigma\beta$ volumetric activity in the “Bypass” system correlates with the results of surveyed airflow direction.

Investigation of radiation contamination of the near-surface air layer under the NSC

Volumetric activity of radionuclides in the near-surface air layer. In 2017–2020, within the space under the NSC near southern wall of Unit 4 Turbine Hall, 150 aerosol samples were taken in the near-surface air layer. Volumetric activity of ^{137}Cs during a year was changing within the range from 14 mBq/m^3 to 5 Bq/m^3 . At the same time, mean annual value of nuclide volumetric activity during four years has been remaining at the level of about 0.2 Bq/m^3 . Volumetric activity of ^{241}Am during a year was changing within the range from 0.3 mBq/m^3 to 0.1 Bq/m^3 . Mean annual value of ^{241}Am volumetric activity was changing within the range of 1.7–4.4 mBq/m^3 .

To identify the sources of radioactive aerosol origination, it is very important to know radionuclide ratio. In Fig. 8, ratio of volumetric activities of ^{137}Cs and ^{241}Am in the near-surface air layer under the NSC is shown in period from 2017 to 2020. The Figure demonstrates that the ratio is within the range of 30–440. Besides, for more than 80% of cases the ratio value was not above the range of 30–100. The value 30 (with considering measurement errors) is typ-

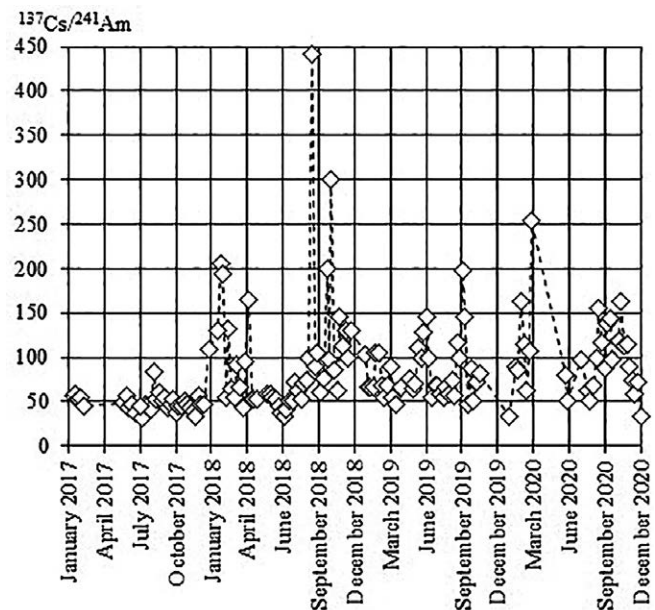


Fig. 8. Ratio of volumetric activities of $^{137}\text{Cs}/^{241}\text{Am}$ in air near Turbine Hall wall under the NSC in 2017–2020

ical for base content of the fourth Unit fuel in 2017–2020. The higher values of $^{137}\text{Cs}/^{241}\text{Am}$ (40–440) ratio appeared due to presence in the air of fuel dust and aerosol particle-carriers of “condensation” origin radio cesium [7].

Density and velocity of radionuclide depositions on the underlying surface. In 2017–2020, 39 samples of radionuclide depositions were taken within the space under the NSC near southern wall of the Unit 4 Turbine Hall. Deposition density of ^{137}Cs during a year had been changing within the range of 0.30–54 Bq/(m² · day), and mean annual value was reducing during four years from 31 Bq/(m² · day) to 1.8 Bq/(m² · day). Deposition density of ^{241}Am during a year was changing within the range of 6.8–1900 mBq/(m² · day). Mean annual density of americium deposition was reducing during four years from 1.2 Bq/(m² · day) to 0.042 Bq/(m² · day). The ratio of $^{137}\text{Cs}/^{241}\text{Am}$ in fall-outs was within the limits of values 24–126. Mean value of ratio magnitude over 2017–2020 period made 44.

Near the fourth Unit western wall, starting from 2018 to 2020, 25 deposition samples were taken. Deposition density of ^{137}Cs was changing within the range of 1.0–40 Bq/(m² · day) under mean value over survey period 9.9 Bq/(m² · day). Deposition density of ^{241}Am was changing within the range of 0.02–1.2 Bq/(m² · day) under mean value — 0.23 Bq/(m² · day). The ratio of $^{137}\text{Cs}/^{241}\text{Am}$ in deposition was within the limits of values 26–94, and mean value made 50.

Deposition velocity of ^{137}Cs near southern wall of Unit 4 Turbine Hall was changing during four years within the range of $1.4 \cdot 10^{-5}$ – $1.6 \cdot 10^{-2}$ m/s under mean value $2.1 \cdot 10^{-3}$ m/s. Deposition velocity of ^{241}Am was changing within the range of $2.9 \cdot 10^{-5}$ – $2.3 \cdot 10^{-2}$ m/s under mean value of $3.7 \cdot 10^{-3}$ m/s.

Dispersivity of ^{137}Cs and ^{241}Am -bearing aerosols in the surface air layer. In 2017–2020, the survey of dynamics of disperse content of radioactive aerosols in the near-surface air layer near the southern wall of Unit 4 Turbine Hall was continued, which were initiated in 1997 [6]. Totally during four years, 58 samples were taken. The calculated distribution of radionuclides on sizes of ^{137}Cs -bearing particles has shown that AMAD value, from one sample to another, during four years had been varying from 1.2 to 5.0 μm. AMAD of ^{241}Am -bearing particle was also changing within analogous range of 1.3–5.0 μm. In most cases, ^{137}Cs -bearing particles had wide polydispersity, since standard geometry deviation (σ) exceeded 3, in addition, in a half of cases, σ were higher 4 that indicated bimodal aerosol distribution. Under low AMAD (≤ 1.7 μm), ^{137}Cs share related to particles with AD less 0.8 μm, reached 36% of its total activity in a sample.

Conclusions

With the NSC commission, the “unorganized” release of radioactive aerosols from the Shelter object in the NSC basic volume in 2020 dropped almost in one and a half times as compared to 2016. Mean annual value of activity flux in the NSC basic volume has also dropped at the same amount.

Mean annual density of radionuclide depositions onto the light roofing of the Shelter object in 2017–2020 increased almost twice as compared to 2016 [2, 3].

The AMAD of ^{137}Cs -bearing aerosols in the flux from the Shelter object through the “Bypass” system in 2017–2020 has been changing, in most cases, from 5 to 10 μm, on top of that, the mean value made 7.7 μm. With the NSC commission, the contribution of large ^{137}Cs -bearing particles in volumetric activity of airflow through the “Bypass” system dropped. At the same time, the dynamics of $\Sigma\beta$ volumetric activity in the “Bypass” system correlates with the results of surveyed airflow direction in the system. As a result of changed temperature and humidity mode in the Shelter object under NSC, in 2017–2020 an increase was observed in the number of cases of air movement inside the object through the “Bypass” system.

As of 2020, a drop in contamination level of the near-surface air layer under the NSC is observed, as follows: for ^{137}Cs — to 0.2 Bq/m³, and for ^{241}Am — up to the values within the range of 1.7–4.4 mBq/m³. In 2017–2020, AMAD of ^{137}Cs -bearing particles in the near-surface air layer under the NSC has been varying from 1.2 to 5 μm, and for ^{241}Am — from 1.3 to 5 μm.

Mean annual density of ^{137}Cs deposition on the underlying surface under the NSC has been dropping from 2017 during four years — from 31 Bq/(m² · day) to 1.8 Bq/(m² · day). Mean annual density of ^{241}Am deposition over the same period has been dropping from 1.2 Bq/(m² · day) to 0.042 Bq/(m² · day).

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Радіоактивні аерозолі в умовах нового безпечного конфайнмента у 2017–2020 рр.

Наведено результати контролю поведінки радіоактивного аерозолу в умовах нового безпечного конфайнмента (НБК) у 2017–2020 рр. З введенням в експлуатацію НБК «неорганізований» викид радіоактивного аерозолу з об’єкта «Укриття» в основний об’єм НБК в 2020 р. знизився майже в півтора рази в порівнянні з 2016 р. На стільки ж знизилася й питома швидкість викиду аерозолу в основний об’єм НБК. Максимальна швидкість «неорганізованого» викиду продуктів Чорнобильської аварії у простір під накриттям НБК спостерігалася в 2017 р. — 7,9 кБк/(м²·доба). Середньорічна щільність випадань радіонуклідів на легку покрівлю протягом чотирьох років змінювалася в діапазоні 1,7–2,2 кБк/(м²·доба), тобто зросла майже в два рази в порівнянні з 2017 р. Середньорічна об’ємна активність суми довгоживучих бета-випромінюючих нуклідів, що надходили в систему «Байпас», знизилася за чотири роки від 0,84 до 0,17 Бк/м³. Їхніми носіями були частинки з медіанним за активністю аеродинамічним діаметром 4,3–10 мкм. Станом на 2020 р. спостерігається зниження рівня забруднення приземного шару повітря під НБК по ¹³⁷Cs до 0,2 Бк/м³. У 2017–2020 рр. медіанний за активністю аеродинамічний діаметр (АМАД) частинок-носіїв ¹³⁷Cs в приземному шарі повітря під НБК варіював від 1,2 до 5 мкм. Середньорічна щільність випадань ¹³⁷Cs на підстильну поверхню під НБК знижувалася протягом чотирьох років з 31 Бк/(м²·доба) до 1,8 Бк/(м²·доба).

Ключові слова: новий безпечний конфайнмент, об’єкт «Укриття», аерозоль, об’ємна активність, щільність випадань радіонуклідів, медіанний за активністю аеродинамічний діаметр.

Надійшла 05.07.2021

Received 05.07.2021