FORMATION OF ATMOSPHERIC AIR CONTAMINATION WITH TRITIUM ABOVE THE WATER AREAS OF INDUSTRIAL WATER BODIES

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The article considers the possible pathways for tritium release from the water area of industrial reservoirs into the atmosphere under normal operating conditions at the facilities holding special (non-removable) radioactive waste generated due to the implementation of the state nuclear weapons program and the state defense order. The paper provides a list ranking each potential pathway of atmospheric air contamination with tritium above the water area of industrial reservoirs and evaluates the degree of its significance. The study demonstrates that it is the water evaporation from the surface of the reservoirs that most significantly contributes to the contamination (at least 99.99% of the totally released tritium), while the total contribution of water aerosol droplet entrainment and tritium-containing methane emission is negligible (does not exceed 0.006%).

Keywords: *activity, tritium, evaporation, droplet entrainment, emission, methane, effective dose, radioactive waste.*

Introduction

Industrial reservoirs, including the Techa Cascade of water reservoirs (hereinafter referred to as TCR), were established to accumulate liquid radioactive waste (LRW) resulting from the national armament program of 1949—1956 [1]. Today, it is a unique technogenic phenomenon being considered an aerial source of radionuclide release into the atmosphere not isolated from the environment. The published analytical and field studies show that radioactive substances can release both abiotically and biotically into the atmosphere from the surface of these industrial reservoirs.

A few papers [2]—[7] explore the main pathways for the radioactive substances, including tritium (T or ^{3}H): these are released as aqueous aerosols due to droplet entrainment and water evaporation. As regards the water area of the Karachay reservoir, the calculated average specific water entrainment from its surface in the warm season was found to be equal to $1.95 \cdot 10^{-2}$ $dm^3 \cdot m^{-2} \cdot yr^{-1}$ [2]. Radioactive releases into the atmosphere with the droplet entrainment appear to be uneven in time and largely depend on various radionuclides accumulated in the surface water microlayer [3]. As regards the two key radionuclides, namely, ^{137}Cs and ^{90}Sr, the specific activity of a water aerosol was assumed as equal to its specific activity in the water of relevant industrial reservoirs [4]. A few studies showed that most of the water aerosol has been precipitating fast, thus, complicating radionuclide transfer away from the water area [5], [6]. At the same time, in the warm season it is seen as a continuous process providing

constant presence of radioactive substances in the air above the industrial reservoir surface.

The published research demonstrates that monthly water evaporation from 1 m^2 of a water area averages to some 30—113 liters [7]. Given such evaporation intensity, tritium activity contained in ~0.5 m^3 of water in a given reservoir gets released annually into the atmosphere.

Some biotic mechanisms contributing to radionuclide releases from industrial reservoirs into the atmosphere were considered as well. Thus, the estimated radionuclide release from the water bodies V-10 and V-17 with bell mosquitoes amounted to several gigabecquerels for ^{90}Sr and hundreds of megabecquerels in case of ^{137}Cs [8]. Such a specific pathway for activity release causes no direct atmospheric contamination and cannot be considered a fugitive source of radioactive emissions. Comprehensive studies focused on the ecosystems of the industrial reservoirs and their state revealed certain features characteristic for the biocenoses in each of these water bodies in particular. Many ecosystems are characterized by a reduced state: ichthyofauna is absent, phytoplankton and zooplankton species are not abundant, which is not common for the region [9]. At the same time, under anaerobic conditions, vital activity of some microorganisms results in biogenic generation of methane that may involve the radioactive isotope of hydrogen, i. e., tritium, in its composition. Methane release from the water surface may cause higher tritium concentrations in the surface air. This effect has not been estimated before, nevertheless, these estimates can be made based on the published studies exploring the intensity of greenhouse gas emissions from the surface of natural water bodies in similar climatic zones [10], [11].

In case if officially an industrial water body gets the status of a nuclear facility, it shall be subject to a systematic and continuous management [1]. Monitoring measures planned and implemented in relation to any radiation exposure source depend on the current and the predicted risk levels to the public and the environment under normal and abnormal operational conditions and emergency situations. During normal operation, the radiation impact on the population and the environment is limited based on the emission and discharge standards established for the radioactive substances.

As it comes to industrial reservoirs, one may decide if the application of standards regulating atmospheric radionuclide releases is required based on a conservative assessment of potential human exposure driven by the contaminated air above the water areas. If, given that the radionuclide dilution in the air is not accounted for, the expected annual effective dose of 10 μSv is exceeded, the release standards set for all radionuclides contributing to 99 % of the dose should be validated [12], [13]. The induced annual exposure dose should be evaluated with an account taken of all radionuclides contained in the water of the industrial reservoirs and listed as contaminants subject to the state regulation in the field of the environmental protection [14].

This paper provides a conservative estimate of potential atmospheric air contamination with tritium above the water areas of industrial reservoirs due to droplet entrainment (water aerosol formation), water evaporation and methane release. The studies performed can be viewed as the preliminary stage supporting the decision-making on the preferred calculation options that can be followed to inventory the areal sources of atmospheric contamination with tritium.

Materials and methods

This study implements a universal approach to the assessment of the environmental impact produced by tritium releases from the water areas of industrial reservoirs into the atmosphere, which includes:

- characterization of industrial reservoirs as areal fugitive sources of tritium releases into the atmosphere: water area, tritium content in water;
- an algorithm to be followed to calculate and rank most probable pathways of tritium releases into the atmosphere from a unit area of water bodies by the importance level: evaporation, methane emission, water aerosol formation;
- an algorithm that can be used to calculate annual effective doses of human exposure with no account taken of dispersion and to rank them according to their potential dose contribution given the considered mechanisms of air contamination with tritium over a unit water area of the industrial reservoirs.

Fugitive aerial sources of atmospheric tritium releases from the water areas of the industrial reservoirs and their characteristics

Tritium releases into the atmosphere with water aerosol and during evaporation are governed both by the droplet entrainment and vaporization intensities and by the specific activity of water in a particular reservoir. Basically, approximately the same values are provided on the surface areas of industrial reservoirs V-2, V-3, V-4, V-10, V-11 and V-17 in the literature sources [9], [15]—[17]. Data presented by different authors on the content of tritium in these water bodies differ significantly and are contingent upon the research period [9], [16], [17]. The

following data on the surface area and tritium concentrations in the water of each considered water body were taken for the screening assessment of radiation exposure (see Table 1).

Table 1. Water area and specific activity of tritium in the water of the industrial reservoirs

Reservoir	Surface area of the water area, km²	Specific activity of tritium in the water, Bq/g	Tritium content in the water, TU
V-2	18.6	15.9	$1.35 \cdot 10^5$
V-3	0.79	56.0	$4.75 \cdot 10^5$
V-4	1.47	8.5	$7.20 \cdot 10^4$
V-10	18.6	3.7	$3.14 \cdot 10^4$
V-11	44.2	0.78	$6.61 \cdot 10^3$
V-17	0.13	3,900	$3.31 \cdot 10^7$

Tritium units (TU) are often used to describe tritium concentrations in water: 1 TU corresponds to the isotope ratio $^3H/^1H = 10^{-18}$. The relationship between the TU and the concentration or radioactivity units is as follows: one TU is equal to 0.118 Bq/l ($1.18 \cdot 10^{-4}$ Bq/g) or $3.3 \cdot 10^{-16}$ g/l [18, 19].

Estimated tritium releases into the atmosphere driven by evaporation from the reservoir water surfaces

Previously published study [7] estimates water evaporation from the water area of industrial water bodies based on two methods considering each month in the time interval between April and October:

- an empirical formula expressing the relationship between the average monthly evapotranspiration (mm), the average temperature (°C) and relative air humidity (%) for this period [20];
- based on the estimated total amount of heat absorbed by water, the maximum possible evaporation for each month has been calculated [21].

Table 2 presents the published calculated evaporation rates adopted under this study [7]. Obviously, 1 mm of evaporation corresponds to 1 liter of water lost from a surface area of 1 m².

Table 2. Average and maximum water evaporation from industrial reservoirs in the warm season

Value	Evaporation per month, mm/month						
	April	May	June	July	August	September	October
Average	43.9	98.0	113.6	98.9	77.9	55.1	31.5
Maximum	74.8	142.3	185.6	189.0	168.4	133.3	78.0

In general, monthly (m) tritium release into the atmosphere (${}^{elem}Q_i^m$) from a surface area element S^{elem} of the i-th reservoir was calculated based on the following equation:

$$ {}^{elem}Q_i^m = S^{elem} \cdot E_i^m \cdot 10^{-3} \cdot A_i \cdot \rho, \text{ Bq/month}, \quad (1) $$

where E_i^m is the water evaporation rate in the water reservoir (i) per month (m), mm/month;
S^{elem} is the unit surface area (assumed as 10^6 mm²);
10^{-3} is the proportionality factor, ml/mm³;
A_i is the specific activity of tritium in the water reservoir (i), Bq/g;
ρ is the water density (assumed as constant and equal to 1 g/ml).

Annual tritium release (${}^{elem}Q_i^y$) into the atmosphere per unit area of the i-th water reservoir was calculated as a sum of tritium release evaporation for each month:

$$ {}^{elem}Q_i^y = \sum_m {}^{elem}Q_i^m, \text{ Bq/year}. \quad (2) $$

Based on Table 2, one can calculate annual tritium releases into the atmosphere due to water evaporation in a range between the average (${}^{elem}_{mean}Q_i^y$) and the maximum possible values (${}^{elem}_{max}Q_i^y$).

Estimated atmospheric tritium releases with water aerosol due to drop entrainment from the reservoir water surfaces

This study did not calculate water aerosol formation intensity above the water areas of the industrial reservoirs. Considering this pathway, tritium releases were estimated based on data available on the droplet entrainment from the surface of the Karachay Lake, which averaged 0.02 dm³·m⁻²·year⁻¹ [2}—[4]. Since the calculated atmospheric air contamination by tritium during droplet entrainment was of a screening nature, the following conservative conditions could be reasonably assumed:

- in any reservoir, tritium was assumed as being evenly distributed over the entire water volume, including the surface layer contributing to the water aerosol formation;
- within the same year interval, the intensity of water aerosol formation over the water area of any industrial reservoir was assumed equal corresponding to the one calculated for the Karachay Lake.

Thus, annual tritium releases into the atmosphere due to droplet entrainment (${}^{elem}p_i^y$) from a unit area of the i-th water reservoir could be calculated as follows:

$$ {}^{elem}p_i^y = S^{elem} \cdot 0{,}02 \cdot A_i \cdot \rho \cdot 10^3, \text{ Bq/year}, \quad (3) $$

where ^{Selem} is the unit surface area (1 m²);
0.02 stands for the droplet entrainment intensity from the surface of any reservoir, dm³·m⁻²·year⁻¹;
A_i is the specific activity of tritium in the water reservoir (i), Bq/g;

ρ is the water density (1 g/ml);
10^{-3} is the proportionality factor, ml/mm³.

Estimated atmospheric tritium release with methane emissions from the reservoir water surfaces

Lakes seen as essential natural sources of methane release into the atmosphere constitute to (24—27)% of the global methane emissions from the natural sources [10]. In water bodies, methane is generated all year round. Nevertheless, its release into the atmosphere may occur only in the warm season. In the Western Siberia, methane emissions into the atmosphere from lakes with a surface area of less than 100 ha amount to 8.6 mgCH_4m^{-2}year^{-1} [11]. In the winter season, due to a stable ice cover, methane gets accumulated in the water of the reservoirs partially freezing into ice. In the spring, during a short ice melting and lake water mixing period, methane gets intensively released into the atmosphere. In this study, the methane emission period was assumed equal to the length of the growing season typical for the considered natural zone with an average daily surface air temperature of at least 5 °C. Methane emissions into the atmosphere occur due to the following mechanisms, namely bubble emission from the shallow parts of water reservoirs and diffusion from the water reservoir surfaces.

A review [10] notes that the highest methane release rate per unit area of a water surface (7.2 gCH_4m^{-2}year^{-1}) is considered typical for the lakes of the broad-leaved forests, while the lowest rates are observed in the northern and middle taiga lakes (1.7—2.2 gCH_4m^{-2}year^{-1}).

This rate differs for the lakes depending on the size of their water surface areas [10]:

- for water reservoirs with an area of up to 0.1 km² — 8.9 ± 2.9 gCH_4m^{-2}year^{-1};
- for water reservoirs with an area of 0.1 to 10 km², which corresponds to the area of V-3, V-4, V-17 reservoirs, — 3.6 ± 0.3 gCH_4m^{-2}year^{-1};
- for water reservoirs with an area of over 10 km², which corresponds to the area of V-2, V-10, V-11 reservoirs — 1.5 ± 0.1 gCH_4m^{-2}year^{-1}.

To estimate tritium releases into the atmosphere associated with methane emissions from the water area of a reservoir (*i*), it was conservatively assumed that:

- a single methane molecule may contain no more than one tritium atom (CH_3T);
- the ratio between protium and tritium isotopes in all objects of local biocenosis in a reservoir (*i*) was assumed equal to the one typical for the water from the considered reservoir.

The ratio of the number of CH_3T and CH_4 molecules in a gas mixture of pure and tritiated methane can be calculated as a fourfold ratio between the number of tritium and protium atoms in the considered gas mixture (i. e., accepted isotope ratio in TU, see Table 1). The mass ratio between CH_3T and pure methane (CH_4) at a given $^3H/^1H$ will be equal to the division of the molecule number in these gases multiplied by the quotient of their molar masses (18/16). If for both gases the sum of the molecule masses is equated to 1 gram, then, taking into account the mass ratio of CH_3T/CH_4 at a given $^3H/^1H$, one can calculate the masses of CH_3T and tritium per 1 gram of such a mixture. The activity of the tritium mass resulting in 1 gram of reservoir methane (*i*) can be calculated as follows:

$$A_i = \frac{N_A \cdot \ln 2 \cdot m_i}{T_{1/2} \cdot \mu}, \text{ Bq}, \quad (4)$$

were μ is the molar mass (for $^3H = 3$ g/mol);
$T_{1/2}$ is the half-life ($3.89 \cdot 10^8$ sec or 12.32 years);
m_i is the tritium mass in 1 gram of methane in the reservoir (*i*), g;
N_A is the Avogadro's number ($6.022 \cdot 10^{23}$).

General dependence representing annual tritium releases in the methane form ($^{elem}M_i^y$) from a unit surface area (1 m²) of a reservoir (i) can be expressed as follows:

$$^{elem}M_i^y = S^{elem} \cdot J_i \cdot L_i, \text{ Bq/year}, \quad (5)$$

where S^{elem} is the unit surface area (1 m²);
$J_i = A_i/(1\text{g})$ is the specific activity of tritium in the methane of the considered water reservoir (*i*), Bq/g;
L_i is methane emission from the surface area of a reservoir (*i*), gCH_4 m^{-2}year^{-1}.

Estimated annual effective dose due to tritium releases from the water areas of the industrial reservoirs

If tritium release rates from the water areas of the industrial reservoirs into the atmosphere due to water evaporation ($^{elem}Q_i^y$, Bq/year), water aerosol droplet entrainment ($^{elem}p_i^y$, Bq/year) and methane emission ($^{elem}M_i^y$, Bq/year) are considered separately, one can calculate the annual effective exposure dose for a person with respect to each atmospheric air contamination pathway individually from any unit surface area (1 m²) of the *i*-th reservoir as follows:

$$\text{for evaporation: } E_i^Q = \Psi_i^Q \cdot {}^{elem}Q_i^y, \text{ Sv/year}, \quad (6)$$

$$\text{for droplet entrainment: } E_i^p = \Psi_i^p \cdot {}^{elem}p_i^y, \text{ Sv/year}, \quad (7)$$

$$\text{for methane emission: } E_i^M = \Psi_i^M \cdot {}^{elem}M_i^y, \text{ Sv/year}, \quad (8)$$

where Ψ_i^Q is the transition function that relates the annually released tritium activity during water

evaporation from the *i*-th reservoir with the annual effective dose for the population, Sv/Bq;
Ψ_i^p is the transition function that relates the annually released tritium activity due to water aerosol's droplet entrainment from the *i*-th reservoir with the annual effective dose, Sv/Bq;
Ψ_i^M is the transition function that relates the annually released tritium activity due to methane emissions from the *i*-th reservoir with the annual effective dose, Sv/Bq;
${}^{elem}Q_i^y$ is the intensity of tritium release during water evaporation from 1 m² of the *i*-th reservoir, Bq/year;
${}^{elem}p_i^y$ is the intensity of tritium release during water aerosol's droplet entrainment from 1 m² of the *i*-th reservoir, Bq/year;
${}^{elem}M_i^y$ is the intensity of tritium release during methane emission from 1 m² of the *i*-th reservoir, Bq/year.

To estimate the expected doses from tritium in the HTO form (tritium water) contained in the atmospheric air, one should assume that its specific activity in the water found within the tissues of biological objects and in the atmospheric moisture is equal. The ratio used to calculate the transition function relating the annually released tritium activity from the *i*-th reservoir with the annual effective public exposure dose, the evaporation processes (Ψ_i^Q, Sv/Bq) and droplet entrainment (Ψ_i^p, Sv/Bq) takes into account tritium intake through inhalation, by ingestion and through the skin and can be calculated based on the following expressions:

for evaporation:

$$\Psi_i^Q(x) = \frac{1}{3{,}15 \cdot 10^7} \cdot \frac{G_i^Q(x)}{H} \cdot k_T, \text{ Sv/Bq}, \quad (9)$$

$$\Psi_i^p(x) = \frac{1}{3{,}15 \cdot 10^7} \cdot \frac{G_i^p(x)}{H} \cdot k_T, \text{ Sv/Bq}, \quad (10)$$

where $3.15 \cdot 10^7$ is the number of seconds in a calendar year, s;
$G_i^Q(x)$ is the average annual meteorological dilution factor in the ground layer of the atmosphere for tritium released as vapor during its evaporation from the water area of the *i*-th reservoir, s/m³;
$G_i^p(x)$ is the average annual meteorological dilution factor in the ground layer of the atmosphere for tritium emitted during droplet entrainment from the water area of the *i*-th reservoir in a water aerosol form, s/m³;
H is the absolute air humidity. If no data from local field studies are available, *H* is recommended to be taken equal to $6 \cdot 10^{-3}$ l/m³ [13];
k_T is the dose coefficient for tritium in the HTO form, the value of which is recommended to be taken equal to $2.6 \cdot 10^{-8}$ (Sv·l)/(Bq·yr) [13].

The transition function relating the tritium activity annually released from the i-th reservoir due to methane emission with the annual effective dose of public exposure takes into account only the inhalation tritium intake:

$$\Psi_i^M(x) = \frac{1}{3{,}15 \cdot 10^7} \cdot G_i^M(x) \cdot V_{\text{cr. g}} \cdot \varepsilon_{\text{pop}}^{\text{air}}, \text{ Sv/Bq}, \quad (11)$$

where $3.15 \cdot 10^7$ is the number of seconds in a calendar year, s;
$G_i^M(x)$ is the average annual meteorological dilution factor in the surface layer of the atmosphere for tritium released from the water area of the *i*-th reservoir due to methane emission, s/m³;
$V_{\text{cr. g}}$ is the annual volume of air inhaled by a critical group of population, in the considered case, for the 1—2-year-old children $V_{\text{cr. g}} = 1{,}900$ m³/year [23];
$\varepsilon_{\text{pop}}^{\text{air}}$ is the dose coefficient due to inhalation-driven tritium intake by a critical group of population, in the considered case for the 1—2-year-old children $\varepsilon_{\text{pop}}^{\text{air}}$ is equal to $2.7 \cdot 10^{-10}$ Sv/Bq [23].

Hypothetical annual effective dose was estimated with no account taken of tritium compound dispersion in the atmosphere with the dilution factors from formulas (9)—(11) assumed as equal to 1 s/m³.

Estimated radiation impact of tritium releases from the water areas of the industrial reservoirs

When it comes to the operational safety of nuclear facilities intended for non-removable RW storage, one should provide evidence on whether the radioactive releases are to be categorized either as regulated by standards or not with relevant demonstration provided through measurements or calculations focused on the atmospheric air contamination with radionuclides, which is viewed as an essential stage of this process. These data can be obtained based on the preliminary estimates characterizing the considered radioactive substance, namely, its form, specific activity and the intensity of its release into the atmosphere.

Tritium release from industrial reservoir surfaces

The atmospheric air above the industrial reservoirs gets contaminated by tritium at a rate proportional to the activity of the tritium-containing substances and the rate of its release into the atmosphere from a unit surface area of the reservoir. Tritium activity in aqueous aerosols and water vapor was calculated based on the published initial data (see the Materials and Methods section), whereas its activity in methane was calculated based on the equality of tritium units in water and in methane within a same reservoir. Table 3 presents the expected specific activities calculated for the tritium contained in the methane of the industrial reservoirs.

Table 3. Expected specific activities and tritium releases with methane over the surface of the industrial reservoirs

Reservoir	Specific activity of methane, Bq/g	Tritium release due to methane emission	
		Specific, $Bq \cdot m^{-2} \cdot year^{-1}$	From the entire surface area, $Bq \cdot year^{-1}$
V-2	$3.61 \cdot 10^{1}$	54.1	$1.01 \cdot 10^{9}$
V-3	$1.27 \cdot 10^{2}$	457	$3.61 \cdot 10^{8}$
V-4	$1.93 \cdot 10^{1}$	69.3	$1.02 \cdot 10^{8}$
V-10	$8.40 \cdot 10^{0}$	12.6	$2.34 \cdot 10^{8}$
V-11	$1.77 \cdot 10^{0}$	2.65	$1.17 \cdot 10^{8}$
V-17	$8.85 \cdot 10^{3}$	31900	$4.14 \cdot 10^{9}$

For any industrial reservoir, the calculated specific activities of tritium in methane turned out to be 2.27 times higher than the specific activity of the water itself. In terms of the specific tritium release during methane emission from a unit surface area, the industrial reservoirs can be ranged in the following way: V-17 > V-3 > V-4 > V-2 > V-10 > V-11. According to the total tritium release during methane emission from the entire water area, these can be ranged differently: V-17 > V-2 > V-3 > V-10 > V-11 > V-4.

Available initial data for evaporation, droplet entrainment and methane emission intensity calculations, as well as the considered empirical regularities and accepted conservative conditions can be used for the screening assessment of annual tritium releases into the atmosphere from each square meter of the industrial reservoirs (Table 4).

Table 4. Intensity of tritium releases from the water area of each industrial reservoir

Reservoir	Total tritium release		Contribution of a single pathway to the total release, %		
	Specific, $Bq \cdot m^{-2} \cdot year^{-1}$	From the entire surface area, $Bq \cdot year^{-1}$	Water evaporation	Water aerosol	Methane emission
V-2	$8.25 \cdot 10^{6}$	$1.53 \cdot 10^{14}$	99.995	0.004	0.001
V-3	$2.91 \cdot 10^{7}$	$2.30 \cdot 10^{13}$	99.995	0.003	0.002
V-4	$4.41 \cdot 10^{6}$	$6.48 \cdot 10^{12}$	99.995	0.003	0.002
V-10	$1.92 \cdot 10^{6}$	$3.57 \cdot 10^{13}$	99.995	0.004	0.001
V-11	$4.05 \cdot 10^{5}$	$1.79 \cdot 10^{13}$	99.995	0.004	0.001
V-17	$2.02 \cdot 10^{9}$	$2.63 \cdot 10^{14}$	99.995	0.003	0.002

Data from Table 4 indicate that it's water evaporation that mainly contributes to air contamination by tritium all across the industrial reservoirs and their water areas, whereas methane emission provides only some negligible contribution. For this reason, this pathway has not been considered in case of artificial (industrial) water bodies and its impact appears to be limited to the evaporation driven releases [7], [24], [25].

The estimated contribution of methane to this process can be judged as true, false or requiring some adjustment only based on the long-term monitoring of its emissions from the water areas of the considered water reservoirs. Experiments seeking to identify the ratio between the specific activities of tritium in water and in methane within a single reservoir may turn to be quite helpful under other studies focused on other surface water reservoir contamination with tritium.

Dose across the water areas of the industrial reservoirs with no account taken of the dilution processes

Hypothetical public exposure dose due to air contamination should be calculated to evaluate the contribution of various factors on the annual effective exposure of a critical group. These can be most easily compared based on the calculated specific methane emission from a unit surface area of a considered reservoir. Table 5 presents the calculated annual effective dose with no dilution processes taken into account.

The data obtained provide no quantitative estimates regarding the radiation impact produced on the population by tritium releases from the water areas of the industrial reservoirs: based on them one can only estimate the contribution of the considered biotic and abiotic tritium releases into the atmosphere to the annual effective dose. It was demonstrated that water evaporation from the water areas should be considered as the main exposure-contributing factor. The contribution of methane emissions to the exposure dose being disproportional to the atmospheric air contamination intensity (being approximately an order of magnitude lower) is explained by the fact that the exposure of the critical group of population is limited only to the inhalation pathway. The expected contribution of methane emissions to the annual effective dose accounting for about 10^{-4} % is considered negligible and, therefore, this tritium release pathway into the atmosphere can be neglected.

Table 5. Annual effective dose of human exposure due to tritium released from a unit water area (1 m^2) of an industrial reservoir with no account taken of dilution processes occurring in the atmosphere

Reservoir	Annual effective dose, $\mu Sv \cdot year^{-1}$	Contribution to the effective dose, %		
		Water evaporation	Water aerosol	Methane emission
V-2	1.14	99.996	0.004	0.0001
V-3	4.00	99.996	0.004	0.0002
V-4	0.61	99.996	0.004	0.0002
V-10	0.26	99.996	0.004	0.0001
V-11	0.06	99.996	0.004	0.0001
V-17	278.41	99.996	0.004	0.0002

Conclusions and discussion

Water areas of reservoirs containing tritiated water are commonly considered as fugitive areal sources of tritium releases into the atmosphere given a single possible pathway — through water evaporation. Studies focused on water aerosol formation, experimental data on methane emission from the water areas of lakes and water bodies suggest the existence of two previously unconsidered ways of atmospheric air contamination by tritium — droplet entrainment of water aerosol (tritium in the HTO form) and methane emission (tritium in the CH_3T form).

The rates of tritium releases into the air due to droplet entrainment can be calculated based on the data published on water aerosol formation across the Karachay Lake, whereas those associated with methane emissions can be estimated conservatively based on the experiments implemented in the water areas of some Siberian lakes located approximately at the same latitude as the TCR.

The rates of tritium releases from the water areas of industrial reservoirs into the atmosphere calculated with an account taken of various pathways allowed to rank them in the following order: water evaporation from the surface of a reservoir > droplet entrainment > methane emission. Water evaporation accounts for 99.99 % of atmospheric air contamination by tritium, whereas, the total droplet entrainment of water aerosol and methane emission contribute to less than 0.006 %.

To validate the conservatively estimated tritium release rates from the water areas of the industrial reservoirs and to identify whether these might possibly need some adjustment or should be rejected, long-term experimental observations (studies) are required, including:

- integral activity of tritium in the air above the water area of the industrial reservoirs should be evaluated at various heights;
- contribution provided by evaporation and droplet entrainment to the atmospheric air contamination over the water areas of the industrial reservoirs should be measured instrumentally;
- methane emission intensities should be measured instrumentally;
- tritium activities in methane and water should be measured in each reservoir and within the same period of time;
- tritium/protium ratio should be calculated in each methane sample taken.

The proposed areas of research are expected to provide an unambiguous answer to the question on whether it is required to assign either all of the industrial reservoirs to the category of regulated fugitive tritium release sources or only some individual ones. Such studies are also required to demonstrate if the safe operation of long-term storage facilities for non-removable RW is possible.

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