Original Article

Gas ebullition associated with biological processes in radioactively contaminated reservoirs could lead to airborne radioactive contamination

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ABSTRACT

Background: Storage reservoirs of radioactive waste could be the source of atmospheric pollution due to the efflux of aqueous aerosol from their water areas. The main mechanism of formation of aqueous aerosols is the collapse of gas bubbles at the water surface. In this paper, we discuss the potential influence of biological factors on gas ebullition in the water areas of the radioactively contaminated industrial reservoirs R-9 (Lake Karachay) and R-4 (Metlinsky pond) of the Mayak PA. The emission of the released non-dissolved gases captured with gas traps in reservoir R-9 was (88–290) ml/m^2 per day (2015) and in reservoir R-4 (270–460) ml/m^2 per day (2016). The analysis of gas composition in reservoir R-4 (60% methane, 35% nitrogen, 2.4% oxygen, 1.5% carbon dioxide) confirms their biological origin. It is associated with the processes of organic matter destruction in bottom sediments. The major source of organic matter in bottom sediments is the dying phytoplankton developing in these reservoirs.
Conclusion: The obtained results form the basis to set a task to quantify the relationship between the phytoplankton development, gases ebullition and radioactive atmosphere contamination.

1. Introduction

Air transport of radionuclides with aqueous aerosol has been studied in the nuclear industry enterprises: from the sea surface in Sellafield (Great Britain) [1,2] and water area of a small freshwater reservoir at the territory of the industrial site of the Mayak Production Association (Chelyabinsk Oblast, Russia) [3].

Mayak Production Association (Mayak PA) was founded in 1948. It is a multipurpose enterprise of the nuclear fuel cycle. It comprises reactor, radiochemical, isotopic and other production facilities targeted at manufacturing civil and military goods [4].

In the first years of its operation when there was no knowledge and technologies of safe radioactive waste management, several man-made natural facilities to store liquid radioactive waste of various activity concentration were created [4–7]. The heaviest radioactive contamination of the water and bottom sediments was observed in a small (~0.3 km^2) reservoir R-9 (Lake Karachay). For many years it has been used to store intermediate liquid radioactive waste [8].

Air pollution in the area adjacent to the enterprise in different years was determined by: emissions from the stacks of the enterprise, wind transport of radioactive dust from previously contaminated territory (blowing erosion) and transfer of radionuclides as parts of the aqueous aerosol from the water area of reservoir R-9. In November 2015, the water area of reservoir R-9 was closed down using a special technique. The reservoir was covered with concrete structures and rocky soil. It allowed completely excluding the formation of aqueous aerosol and air pollution. Based on the data analysis of the long-term air pollution monitoring (1968–2020) as well as on the results of the monitoring of natural and anthropogenic (manufacturing) environments, it was shown that up until 2015, the process of formation and transport of aqueous aerosol from the water area of Lake Karachay was the main source of air pollution in the area of the enterprise [3,9–15].

Aqueous aerosol above the open reservoir surface is formed due to various physical and chemical biological processes: when bubbles collapse under breaking waves [16], during rain [17], in case of gas ebullition from bottom sediments [18]. Bubble bursting is a

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fundamentally important physical process in nature, which has crucial environmental and climate ramifications. Our current understanding of surface bubble bursting aerosol formation includes three major pathways, namely film drop, jet drop production, and recently, film flapping [19].

Film drops are formed when the film of a bubble cap bursts, whereas jet drops form when a vertical water capillary collapses because of gravity. It is known that the parent bubble size determines the number of produced film and jet drops: large bubbles produce mainly film drops, while small bubbles produce mostly jet drops. Film drops are responsible for the major proportion (~ 60 %–80 %) of submicrometer particles, whereas jet drops mostly contribute to the production of supermicron particles. A flapping shear instability mechanism leads to a significant fraction of submicron aerosols produced by jet drops from very small (~ 1 μm radius) bubbles [20].

A bursting bubble ejects water droplets known as water spray. When bubbles collapse tiny drops of water are formed that play a crucial role in the formation of aerosol as well as in the transport of non-organic, organic matter, bacteria, pathogenic microorganisms from water and bottom sediments into the air. These spray aerosols mediate the transfer of moisture, salts, organics, and microorganisms through the water–air interface and exert significant effects on atmospheric chemistry, human health, and climate [19,21–23]. In particular, nano/microsized particulates and organisms present in the bulk water can be highly enriched in the jet drops ejected during bubble bursting [24].

The number of drops formed when bubbles collapse depends on the chemical composition of water, temperature, and number of microalgae [16]. The dependencies of the formed drop number on the size of a bubble, ebullition rate were calculated [25].

Aqueous aerosol above the surface of radioactively contaminated reservoirs could be formed under the influence of physical factors, such as waves, rain drops, pressure, and temperature changes. Perhaps, water radiolysis that leads to the formation of hydrogen could contribute to the formation of aerosol [26]. To control and predict radioactive atmospheric contamination due to the formation of aerosol it is also necessary to understand and evaluate the contribution of biological factors (photosynthesis, gas ebullition because of bottom bacteria activities) to this process. The following biological processes are known to result in bubble formation: formation of methane [27], carbon dioxide [28], nitrogen and nitrogen oxide [29] in bottom sediments in the process of organic matter destruction; emission of oxygen at the surface of aquatic plants and periphyton during photosynthesis [30,31].

Methane comprises the major part of the gas in the bubbles rising from the bottom sediments [27,32,33]. Findings from other pieces of literature have shown that methane emission depends on the depth of a waterbody and water level (methane release increases with the decrease in the level) [33,34], water temperature and phytoplankton production [35,36]. In the study [37], it has been demonstrated that for freshwater reservoirs, methane emission, including methane ebullition, depends on water temperature and cyanobacteria biomass. Several studies point to the importance of assessing the trophic status, including mass development (bloom) of phytoplankton to predict methane ebullition [38–42].

Environmental monitoring of radioactively contaminated reservoirs does not consider the role of phytoplankton in the formation of aqueous aerosol. Hence, radioactive contamination of the air is not considered either.

In this paper we put forward the problem of the relationships between phytoplankton development and emission of methane and other gases that determine the formation of the aerosol above the radioactively contaminated reservoirs. During the study, we measured the radionuclide content in water and bottom sediments and the volume of the gases rising from the bottom as bubbles, determined their composition, and assessed the phytoplankton biomass of the studied radioactively contaminated reservoirs of the Mayak PA (Chelyabinsk region, Russia).

2. Experimental

2.1. Materials and methods

2.1.1. Study area

Two special industrial reservoirs of the Mayak PA were the object of the current study: reservoir R-9 (in 2015 up until its open water area was closed down) and reservoir R-4 (in 2016).

Reservoir R-9 (Lake Karachay) until its water area was closed down in 2015 had been the reservoir with the highest level of radioactive contamination among the special industrial reservoirs of the Mayak PA. As of 1970 data, the content of ^{90}Sr in the upper layer of the bottom sediments reached 1.3×10^{11} Bq/kg dry weight; of ^{137}Cs - 1.5×10^{11} Bq/kg dry weight; of α-emitting radionuclides (mainly isotopes of Pu, Am, Cm) – 1.7×10^6 Bq/kg dry weight [8]. Until 1951, reservoir R-9 was a natural lake-swamp Karachay with time-dependent feeding patterns. The water body was rather small, very shallow (the depth <2 m) in its previous natural state and shallow (the depth was 2–5 m) in the period of its usage. The work on eliminating the open water area of Lake Karachay began in 1968. They were finished on 26 November 2015.

Samples in reservoir R-9 were taken in summer 2015. The works on filling the banks to eliminate open water area were conducted on an ongoing basis. Therefore, the reservoir at the time of the study was a narrow, elongated aquatic object 5–10 m in width with a surface area of about 0.5 hectares. Sampling was performed from banks.

Reservoir R-4 (former Metlynsky pond) is a part of the Techa Cascade of Reservoirs. It is a storage reservoir of low-level liquid radioactive waste and is being used in a continuous flow mode. The mean depth of the water body is 1.8 m (0.4–3.2 m), surface area 1.86 km^2, volume 3.3 $\times 10^6$ m^3.

Sampling from reservoir R-4 was performed in summer 2016 at 4 sampling stations (Fig. 1). Sampling station R-4/1 is located in the upper reaches of the reservoir where the hydrologic regime is similar to that of a river. Sampling station R-4/2 is located close to the middle reaches of the reservoir. Sampling station R-4/3 is located in the riverside area. Sampling station R-4/4 is located in the deepest part of the reservoir with the lowest flow.

2.1.2. Sampling and radionuclide content measurement

In reservoir R-4, water to measure the radionuclide content was sampled from the epipedon (1.5 l), bottom sediments were sampled with a grab sampler. Bottom sediments from reservoir R-9 were not studied.

Content of ^{137}Cs, ^{60}Co, ^{241}Am was measured with gamma spectrometry using a spectrometer with semiconductor detector USK Gamma-plus with highly pure germanium (HPGe). The measurement error ranged from 3% to 20%, depending on the radioactivity of the sample and the measurement time.

In low active samples ^{137}Cs was determined using the radiochemical antimony iodide method.

^{90}Sr concentration in samples was measured by radiochemical isolation of ^{90}Y with subsequent measurement of its activity using low-background β-metric unit of UMF-2000 type and flame-photometric control of the emission of strontium carrier. The error of ^{90}Sr measurement was 10%.

2.1.3. Control of airborne contamination

The control of airborne contamination with radionuclides in the Mayak PA area was arranged in 1952. Over the years, the monitoring system was developed and improved on an ongoing basis. Nowadays, monitoring is performed with the use of tablets (sedimentation method) and air filter units (AFU) (aspiration method) [9,10]. All the main studies on investigating the source of aqueous aerosol formation were conducted using the results of the monitoring at the control site "Novogorny" as it is located just 6 km away from the reservoir R-9. Moreover, we had the most complete (tablets and AFU), reliable (only certified methods and measurement tools were used) set of monitoring
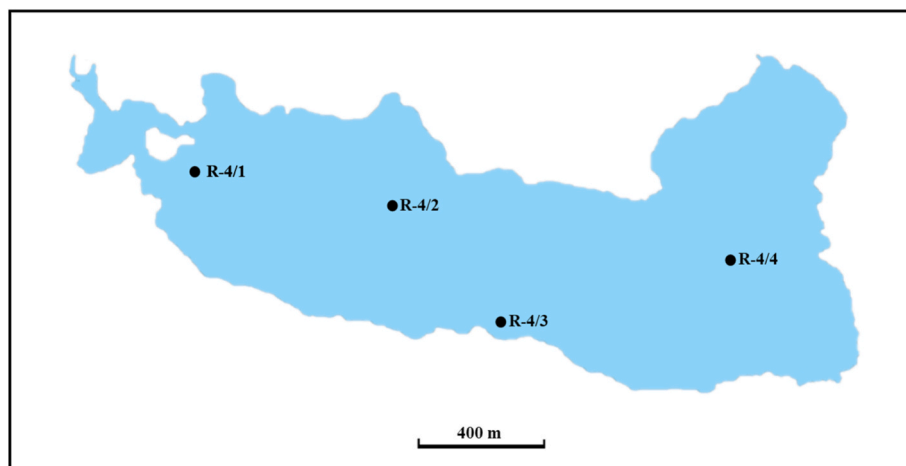
Fig. 1. Location of the sampling station in reservoir R-4.

data covering a long time for this control site (starting from 1968).

2.1.4. Measurement of gas volume and composition

The volume of the gas formed under 1 m^2 of water surface and rising to the surface in bubbles (the amount of dissolved gases was not taken into account) in reservoir R-9 was measured at one sampling station. For this purpose, a unit to collect gas bubbles (gas trap) was used (Fig. 2). The surface of the unit was 188.7 cm^2. Two experiments to collect gases were performed. In the first case, the unit was placed into water of the reservoir and left there for 42 days (in June–July 2015). In the second case, it was left in water for 13 days (on August 2015). At the end of the exposure, the accumulated gas was collected. Its volume was measured using a syringe. The gas volume was calculated per 1 m^2 of the water surface.

In reservoir R-4, the gas volume was measured using the above-described method at three sampling sites (R-4/1, R-4/2, and R-4/4). The gas collecting unit was kept in the water of the reservoir for 11 days on August 2016. At the end of exposure the accumulated gas was collected, its volume was measured using the syringe. The gas volume was calculated per 1 m^2 of the water surface.

Quantitative analysis of gas samples was conducted using the method of gas chromatography with the hardware and software system for medical examinations based on the chromatographer "Chromatech-crystall 5000.2". Ar, O$_2$, N$_2$ were determined with a thermal conductivity detector (TCD), CH$_4$ and CO$_2$ - with a flame ionization detector (FID), H$_2$ - with a catalytic-combustion detector (CCD), and N$_2$O – with an electron-capture detector. Analyzed gases were injected with 25 ml syringes for gas chromatography of SGE - Analytical Science series/type. For each sampling site the measurements were done in triplicate.

Mean amount of each of the formed gases detected in a sample in terms of mmol/m^2 per day was calculated based on the data on the percentage of various gases in samples and sample volume (volume of the gases collected with the collecting unit at each of the stations).

2.1.5. Phytoplankton

Samples of phytoplankton in reservoir R-9 were taken five times over the period from May to September 2015. Sampling was done at the bank. Water samples (0.5 l) were taken from the water surface. After that, they were concentrated to 3 ml by filtration through a membrane filter (with a pore diameter of 0.45 μm), and Lugol's solution fixative was added.

Samples of phytoplankton in reservoir R-4 were taken three times at four sampling sites over the period from June to August 2016. Ruttner's bathometer was used for sampling. Water samples were taken from the surface and further down, every 0.5 m, to the bottom. Samples from different layers were mixed. A 0.5 l sample was taken, concentrated to 3 ml by filtration through the membrane filter (with a pore diameter of

0.45 μm). After that, Lugol's solution fixative was added.

Species identification of phytoplankton and calculation of its abundance were performed according to the standard hydrobiological methods using the respective keys [43,44]. Phytoplankton biomass was determined using the volume-weight method based on the assessment of the abundance of each species and the volume of cells in the specimens of a given species, assuming the density of algae equal to 1 g/ml. The results were used to calculate the mean biomass value of the phytoplankton belonging to various taxonomic groups.

2.2. Results

2.2.1. Assessment of radioactive contamination level in reservoirs

The radionuclide content in the water and bottom sediments of the studied reservoirs is given in Tables 1 and 2. The ^{90}Sr activity concentration in the water of reservoir R-4 was 5.0–6.1 kBq/l, and the ^{137}Cs activity concentration was 0.5–0.7 kBq/l. The activity concentration of bottom sediments was higher: the content of ^{90}Sr in bottom sediments of reservoir R-4 was on average 2.8 MBq/kg dry weight, and ^{137}Cs was 9.7 MBq/kg dry weight.

The radionuclide content in the water of reservoir R-9 was four orders of magnitude higher than that in reservoir R-4.

2.2.2. Assessment of airborne contamination

Some parameters of the source of formation and transfer of the aqueous aerosol from the water area of the reservoir were determined in the study [3] with the use of standard Gaussian model of pollutant ventilation in the atmosphere, experimental data on the density of radioactive fallout of ^{137}Cs in the area of Mayak PA location, and data on the changes in the surface area of the water area and activity concentration of the water of reservoir R-9:

1. Assuming equal ^{137}Cs activity concentration in the water of the reservoir and aqueous aerosol, we evaluated the effective mean annual rate of aqueous aerosol formation, which made up ~20 ml per 1 m^2 of the surface area of reservoir R-9 aquatorium per year. If the surface area of the aquatorium is 0.30 km^2, then the annual efflux of water from the reservoir with aqueous aerosol is expected to be ~6 m^3 of water.

2. It was demonstrated that the rate of the source of aqueous aerosol formation above the aquatorium of the reservoir could vary significantly within a calendar year. Table 3 provides results of the calculation of the annual change (on a monthly basis) of the rate of the source of water efflux from Lake Karachay over the course of 1970, 1974, and 1980 [3].
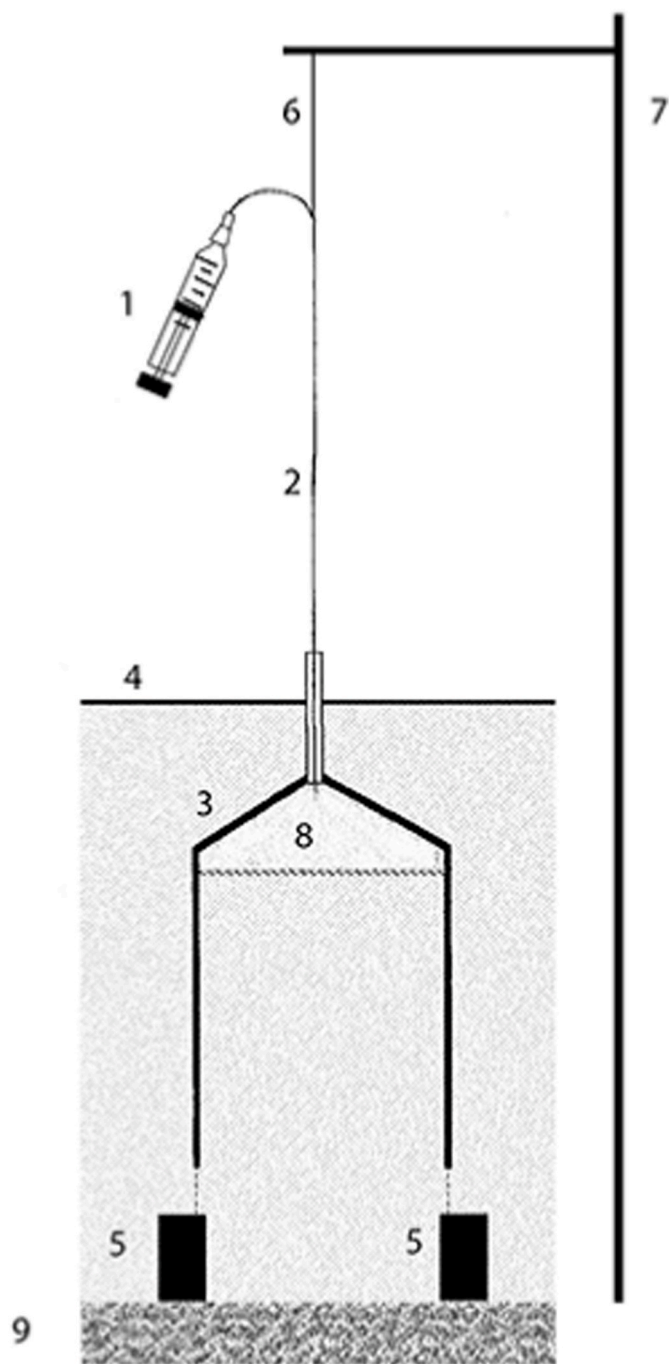
Fig. 2. Gas bubbles collecting unit (gas trap)
1 – syringe to collect gas, 2 – tube, 3- collecting cone, 4 – water surface, 5- weights, 6 – rope, 7 – stand rod, 8 – collected gases, 9 – bottom of the reservoir.

Table 1
Radionuclide content in water of the studied reservoirs (R-4 – June 2016, R-9 – June 2015 г.).

Sampling station	Water, Bq/l	
	^{90}Sr	^{137}Cs
R-4/1	$(5.8 \pm 0.8) \cdot 10^3$	$(6.9 \pm 1.0) \cdot 10^2$
R-4/2	$(5.0 \pm 0.7) \cdot 10^3$	$(4.6 \pm 0.7) \cdot 10^2$
R-4/3	$(6.1 \pm 0.8) \cdot 10^3$	$(5.1 \pm 0.8) \cdot 10^2$
R-4/4	$(5.3 \pm 0.7) \cdot 10^3$	$(5.3 \pm 0.8) \cdot 10^2$
R-9	$(9.9 \pm 1.5) \cdot 10^7$	$(1.14 \pm 0.20) \cdot 10^7$

Table 2
Radionuclide content in bottom sediments of the reservoir R-4, June 2016, Bq/ kg dry weight.

Sampling station	Bottom sediments, Bq/kg dry weight			
	^{90}Sr	^{137}Cs	^{60}Co	^{241}Am
R-4/1	$(8.4 \pm 1.2) \cdot 10^5$	$(1.49 \pm 0.13) \cdot 10^7$	-	$(3.5 \pm 0.5) \cdot 10^5$
R-4/2	$(2.2 \pm 0.3) \cdot 10^6$	$(6.8 \pm 0.6) \cdot 10^6$	$(7.1 \pm 1.0) \cdot 10^3$	$(7.1 \pm 1.0) \cdot 10^5$
R-4/3	$(4.8 \pm 0.7) \cdot 10^6$	$(7.2 \pm 0.6) \cdot 10^6$	$(1.02 \pm 0.10) \cdot 10^5$	$(6.3 \pm 0.9) \cdot 10^5$
R-4/4	$(3.2 \pm 0.5) \cdot 10^6$	$(9.8 \pm 0.9) \cdot 10^6$	$(1.01 \pm 0.13) \cdot 10^5$	$(7.6 \pm 1.1) \cdot 10^5$

Table 3
Expected annual change of absolute ($m^3 \cdot month^{-1}$) and relative rate of the source of water carry-over from the water surface of the reservoir over the course of 1970, 1974, and 1980.

Month	1970		1974		1980	
	m^3/month	%	m^3/month	%	m^3/month	%
January	0.0	0.0	0.005	0.09	0.014	0.16
February	0.004	0.037	0.012	0.22	0.13	1.5
March	0.020	0.19	0.082	1.5	0.074	0.86
April	0.262	2.43	0.315	5.9	0.260	3.02
May	2.53	23.4	1.92	35.8	0.574	6.67
June	4.46	41.30	1.00	18.6	1.07	12.4
July	1.84	17.0	0.548	10.2	1.83	21.3
August	1.08	10.0	0.751	14.0	4.10	47.7
September	0.407	3.8	0.528	9.8	0.333	3.87
October	0.14	1.3	0.092	1.7	0.16	1.9
November	0.017	0.16	0.076	1.4	0.027	0.31
December	0.022	0.20	0.047	0.9	0.029	0.34
In a year	10.8	100.0	5.37	100.0	8.60	100.0

Based on the analysis of the data presented in table 3, it should be noted that [3]:

- in the winter period (from November to March), when the reservoir is covered with ice, there is no activity carry-over from the reservoir water area, and the "apparent carry-over" of activity (less than 1% per month) is due to other sources;
- maximum aerosol efflux from the reservoir water area occurs in summer (June–August) at the maximum water temperature and can reach 50% per month of the annual value of carry-over (up to 10 ml/ m^2/month).

3. The notion of the fractionation factor (accumulation) of radionuclide X in the water aerosol of reservoir R-9 in relation to the reference radionuclide (^{137}Cs) is introduced: F(X/Cs). The range of possible values of the fractionation factor was estimated for the following radionuclides: ^{90}Sr and ^{134}Cs ($\sim$ 1.0); ^{103,106}Ru (0.5–3); ^{95}Zr+^{95}Nb (2–8); ^{125}Sb (1–4); ^{144}Ce (4.5–33) Plutonium fractionation factor in aqueous aerosol relative to ^{137}Cs was estimated. The mean value was 27.4. The variation range was 12.5–50.3.

4. A model of the source power of atmospheric contamination by aqueous aerosol formed above the reservoir surface is proposed. Fig. 3 shows a comparison of calculated and experimental data on ^{137}Cs fallout density at the "Navogorny" control site for the period 1970–2010.

2.2.3. Assessment of the ebullition level

The release of gas bubbles from the bottom of the reservoir was clearly visible in the water of reservoir R-9. The following results were obtained with the gas trap when measuring the amount of gases produced: the gas bubble emission on the surface of reservoir R-9 was 88.3 ml/m^2 day at exposure for 42 days in June–July 2015 and 285 ml/m^2 day at exposure for 13 days in August 2015; the amount of dissolved
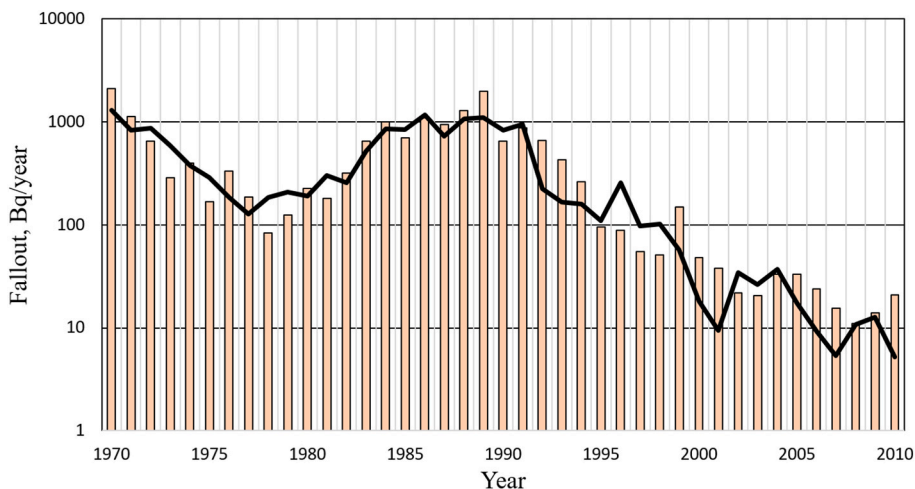
Fig. 3. Comparison of experimental data on ^{137}Cs activity fallout in "Navogorny" control site (bars) with the corresponding calculated fallout as part of the aqueous aerosol (curve) for 1970–2010 [10].

gases was not considered.

In reservoir R-4, the amount of gases rising from the bottom sediment surface and in the water column was on average 370 ± 60 ml per day from 1 m^2 in August 2016.

The composition of gases collected at reservoir R-4 is presented in Table 4. According to the data obtained, we can see that the composition of gases rising from the surface of bottom sediments differs from that of the atmospheric pool. The gases rising from the bottom sediments consist mainly of methane and nitrogen, followed by oxygen and carbon dioxide in much smaller amounts.

In reservoir R-4, methane emission with bubbles from bottom sediments averaged 10 ± 3 mmol/m^2 day in 2016, nitrogen 5.1 ± 1.4 mmol/m^2 day, oxygen 0.37 ± 0.06 mmol/m^2 day, carbon dioxide 0.28 ± 0.21 mmol/m^2 day, and hydrogen 0.00015 ± 0.00003 mmol/m^2 day (Table 5). No nitrogen oxide was detected in the gas samples. The presence of methane, nitrogen, and carbon dioxide in the composition of gases indicates their biological origin [27,32,46–48].

2.2.4. Phytoplankton

Despite a high level of radioactive contamination, phytoplankton develops in the studied reservoirs. In the summer period, abundant microalgae blooms were registered in water bodies.

The phytoplankton biomass in the water of reservoir R-9 is presented in Table 6. The phytoplankton biomass in the water of reservoir R-4 is

Table 4
Composition of gases rising from the surface of bottom sediments and in the water column in reservoir R-4, 2016, % of volume.

Gas	Atmosphere gas composition (published data [45])	Atmosphere gas composition (measurement results)	Sampling station R-4/1	Sampling station R-4/2	Sampling station R-4/4
N$_2$	78.084	79.29	11.6 ± 1	50.15 ± 0.26	42 ± 1.2
O$_2$	20.9476	21.53	2.28 ± 0.2	3.19 ± 0.22	1.58 ± 0.16
Ar	0.934	0.93	0.183 ± 0.015	0.640 ± 0.025	0.590 ± 0.025
CO$_2$	0.038	0.04	3.4 ± 0.5	0.39 ± 0.04	0.66 ± 0.026
CH$_4$	0.00017	0	80.9 ± 0.7	44.86 ± 0.17	53.5 ± 1.2
H$_2$	0.000055	0	0.001 ± 0	0.001 ± 0	0.0007 ± 0.0003
N$_2$O	0	0	0	0	0

Table 5
Composition of gases rising from the surface of bottom sediments and in the water column in reservoir R-4, 2016, mmol/m^2 day.

Gas	Sampling station R-4/1	Sampling station R-4/2	Sampling station R-4/4
N$_2$	2.39 ± 0,20	6.04 ± 0.03	6.87 ± 0.20
O$_2$	0.47 ± 0,04	0.38 ± 0.03	0.26 ± 0.03
Ar	0.038 ± 0.003	0.077 ± 0.003	0.096 ± 0.004
CO$_2$	0.70 ± 0.11	0.047 ± 0.005	0.108 ± 0.004
CH$_4$	16.7 ± 0.14	5.4 ± 0.02	8.75 ± 0.20
H$_2$	0.0002 ± 0.0002	0.0001 ± 0.0001	0.00011 ± 0.00005
N$_2$O	0	0	0
Gas volume, ml/ m^2 × day	462	270	366

Table 6
Phytoplankton biomass in reservoir R-9 in 2015, g/m^3.

Taxon	June	August	September
Total	6.0 ± 0.4	14.3 ± 1.1	5.8 ± 0.3
Cyanobacteria	3.6 ± 0.4	14.3 ± 1.0	5.6 ± 0.3
Diatomeae	0.03 ± 0.03	0.01 ± 0.01	0.02 ± 0.02
Green algae	< 0.01	< 0.01	< 0.01
Cryptophyte	2.33 ± 0.05	0.08 ± 0.03	0.18 ± 0.01

given in Table 7.

The maximum amount of phytoplankton in reservoir R-9 in 2015 was observed in August (biomass reached 14 g/m^3), and the minimum amount of phytoplankton biomass was registered in June (biomass amounted to 6 g/m^3). Thus, algal biomass varied significantly during the summer. It is worth noting that cyanobacteria dominated the composition of phytoplankton; they accounted for 60–99% of the total biomass.

A total of 13 species of microalgae were identified in reservoir R-9 (this is a very insignificant number of species for natural water bodies);

Table 7
Phytoplankton biomass in reservoir R-4 in 2016, g/m^3.

Taxon	June	July	August
Total	19.1 ± 1.1	42 ± 16	17 ± 4
Cyanobacteria	1.66 ± 0.7	16 ± 6	11.1 ± 2.8
Diatomeae	3.8 ± 1.3	20 ± 10	1.1 ± 0.6
Green algae	5.9 ± 1.7	2.08 ± 0.23	2.7 ± 0.9
Other	8 ± 4	3.18 ± 0.29	2.1 ± 0.5

the cyanobacterial species *Microcystis aeruginosa* dominated. As for the environmental characteristics, all microalgae registered in reservoir R-9 are widespread, resistant to increased water salinity, and inhabit reservoirs of any size and hydrological regime [49]. The predominance of cyanobacteria in the composition of phytoplankton is generally characteristic of man-made areas and extreme habitats; cyanobacteria are resistant to a wide range of adverse factors, including ionizing radiation [50].

The phytoplankton community of reservoir R-4 was characterized by higher species diversity, with a total of 86 species identified in phytoplankton samples. As in reservoir R-9, cyanobacteria dominated (40–65% of phytoplankton biomass in July and August), although green and cryptophytic algae were observed here in significant numbers in early summer. The maximum value of algal biomass in reservoir R-4 was higher, than that in reservoir R-9 and amounted to 42 ± 16 g/m^3 in July. The species composition of phytoplankton, algae biomass, and complexes of dominant phytoplankton groups are typical of standing water bodies with a pronounced eutrophication process and high biological productivity against the background of water body saturation with biogenic elements.

The correlation analysis revealed the presence of a positive, statistically significant relationship between the volume of ebullition gas and phytoplankton biomass (Spearman's $\rho = 0.82$; $p = 0.046$).

3. Discussion

Some aquatic ecosystems were radioactively contaminated due to nuclear weapon testing [51], contamination of the environment in the vicinity of the nuclear weapon complex facilities in the USA [52] and the USSR [6–8], radionuclide inflow into aquatic ecosystems at the time of radiation accidents at nuclear power plants, as it has happened at the Chernobyl Nuclear Power Plant and Fukushima-1 Nuclear Power Plant [53], or when natural water bodies are used as coolers at the nuclear power plants [54].

Previous studies have found that in the area where the Russian nuclear facility, Mayak PA (Ozersk, Chelyabinsk Region, Russia), is located, radioactive contamination of the atmosphere is largely determined by the radionuclide-containing aqueous aerosol wind-blown from radioactively contaminated water bodies [10,3,14].

For example, in the studies [9], it was shown that during the entire period of observations of atmospheric pollution by ^{90}Sr and ^{137}Cs at the Novogorny control site (1968–2015) near Mayak PA, the density of radioactive fallout of these nuclides was almost completely determined by carry-over of aqueous aerosol from the water area of reservoir R-9 (Fig. 3).

There are different mechanisms for aqueous aerosol formation. Some of them are related to physical factors, such as wind and rain. Aerosol is also formed when gas bubbles rising to the water surface from bottom sediments collapse [11,18,24,25].

When studying the wind efflux of radionuclides with aqueous aerosol from reservoir R-9 (Lake Karachay), it was noted that atmospheric precipitation had no significant effect on the rate of generation and wind transport of aqueous aerosol [13]. The maximum radioactive contamination of the atmosphere near Lake Karachay was observed in the summer months (May–August) [3,11,15]. It decreased when the water temperature dropped in the fall, despite the presence of other factors of aerosol formation (rain, wind, etc.). It has been suggested that the main factor influencing the formation of aqueous aerosol above the radioactively contaminated water bodies at Mayak PA is the collapse of gas bubbles formed in the water body as a result of biological processes.

In the bottom sediments of water bodies, organic matter is transformed into inorganic substances, including gases, as a result of the activity of decomposers. These gases accumulate in the bottom sediments and then rise in the form of bubbles to the water surface [27]. If the net gas production is higher than its diffusive transfer from bottom sediments to water, then gas bubbles consisting mainly of methane and

nitrogen are formed and released from silt [32,55]. In freshwater sediments, microorganisms produce significant amounts of methane, nitrogen, and carbon dioxide during anaerobic decomposition of organic matter [27,32,46–48,56,57].

In our study, gas bubbles collected in reservoir R-4 averaged 60 ± 11 % methane, 35 ± 12 % nitrogen, 2.4 ± 0.5 % oxygen, 1.5 ± 1.0 % carbon dioxide, 0.47 ± 0.14 % argon, and 0.00089 ± 0.00011 % hydrogen. A very similar composition of gases was observed in natural water bodies in Siberia. For example, samples of gas bubbles were taken using gas traps in two lakes in Siberia (the technique was similar to the one used in our study). The gas composition was: methane 64 ± 16 % and 55 ± 7 %; nitrogen 30 ± 15 % and 40 ± 8 %; oxygen 6 ± 2 % and 7 ± 2 %; carbon dioxide 0.3 ± 0.3 % and 0.2 ± 0.1 % [46]. In an artificially aerated shallow eutrophic freshwater lake in Southern California, methane produced as a result of microbiological processes in bottom sediments made up 80–90%, while carbon dioxide accounted for 2–3% of bubble-forming gas [33]. For a small hypertrophic freshwater lake in Great Britain, the methane content in bubbles collected using a gas trap varied from 44 to 88% [32]. The composition of gases identified in the bubbles rising to the water surface of reservoir R-4 coincides with the composition of gases in the bubbles collected in "non-radioactively contaminated" natural eutrophic water bodies. This confirms the hypothesis of a biogenic origin for bubbles related to the decomposition of detritus in the bottom sediments of the water body.

The formation of gas bubbles in the bottom sediments of a water body depends on such factors as depth, silt temperature, air temperature, atmospheric pressure, composition of organic matter, and the amount of oxygen [33,58–62], as well as on the local features of the water body bottom, which vary greatly in time and space [63].

In addition to the biologically mediated formation of gas bubbles, it is possible to assume the influence of water radiolysis on bubble formation. Under the action of ionizing radiation, water molecules decompose, forming a number of free radicals, the main end products of such processes are hydrogen and hydrogen peroxide. It can be assumed that hydrogen formed as a result of water radiolysis could form bubbles. It should be noted that in our study, hydrogen was detected in the bubbles. Its proportion in the gases collected using gas traps was 0.0009%. Calculations based on the content of radionuclides in bottom sediments (Table 2), mean energy of emitted gammas, electrons, and alpha particles (https://oecd-nea. org/jcms/pl_39910/janis), assuming that all radionuclide decay events in bottom sediments will not be absorbed by the solid substrate of the bottom sediments or dispersed but will result in water radiolysis, give a hydrogen production estimate of 0.000028 mmol/m^2 day, whereas in our study hydrogen production was 0.0001 mmol/m$^2 \times$ day. That is, the detected hydrogen is contained in gas bubbles in a negligible quantity. Most likely, this hydrogen is formed as a result of other processes than water radiolysis.

According to our estimates, the emission of free, undissolved gas that is capable of forming bubbles in July–August was 88–290 ml/m^2 day in reservoir R-9 in 2015 and 270–460 ml/m^2 day in reservoir R-4 in 2016. The obtained results do not contradict the estimates of the volume of gases emitted by bubbles for different natural water bodies. For example, in a small hypertrophic freshwater lake in Great Britain, the bubble methane emission, excluding the contribution of other gases, was 12 mmol/m^2 day (269 ml/m^2 day) [32], in a eutrophic lake in Finland, methane bubble emission was 36–46 mg/m^2 day (50–64 ml/m^2 day) [64], in a shallow eutrophic lake in the hot climate of Southern California, methane bubble emission reached 96 mmol/m$^2 \times$ day (2150 ml/m^2 day) [33].

It should be noted that in order to obtain an accurate estimate of gas emissions from the entire body of water, a greater number of measurement points using gas traps is required since there is a considerable heterogeneity in bubble gas emissions depending on the bottom topography, depth, and other conditions [62,63,65–67].

Findings from many years of research indicate the importance of assessing the trophic status of a water body to predict methane and

nitrogen emissions in the form of bubbles [38,68–70]. Eutrophic shallow lakes have a disproportional share in methane emissions [71,72] because the concentration of nutrients and the number of primary producers – phytoplankton, and higher aquatic plants – increases in the reservoir [73,74]. The levels of methane emissions correlate with the productivity of water reservoirs throughout the world [34]; the rate of methanogenesis in freshwater sediments increases with the biomass of phytoplankton as a whole [38,41] and cyanobacteria within phytoplankton in particular [37]. Bartosiewicz et al. [36,37] established a causal relationship between methane emissions and cyanobacteria biomass and calculated a quantitative dependence between these parameters. The same authors have shown that cyanobacteria blooms lead to methane emissions several times higher than the baseline levels for a water body. This is associated with both an increase in autochthonous carbon delivered to the bottom after algal cell death [39,40,42] and with the depletion of dissolved oxygen reserves in a water body. This promotes the process of anaerobic methanogenesis [37]. Our investigation also revealed a positive, statistically significant correlation between phytoplankton biomass and the volume of ebullition gas. The increase in the number of microalgae cells itself may also be associated with an increase in the number of water droplets formed above the surface of the aquatic environment and forming an aerosol [16].

Water blooming, i.e., mass development of phytoplankton, is observed in the storage reservoirs of liquid radioactive waste in Mayak PA in the summer period [75–80]. According to our data, in reservoir R-9 in 2015, algae biomass reached high values, up to 14 g/m^3, and in reservoir R-4, more than 40 g/m^3. Reservoirs R-9 and R-4 are shallow and well warmed in the summer period. The phytoplankton composition of these bodies was dominated by cyanobacteria. In terms of phytoplankton biomass and the complex of dominant algal species, these water bodies are characterized as eutrophic. All these features are factors contributing to the active process of methanogenesis. In May–September when the water in water bodies warms up, mass development of phytoplankton occurs there. It is one of the main reasons for methane, nitrogen, and carbon dioxide formation in bottom sediments, which can rise to the surface in the form of bubbles and collapse with the formation of water aerosol.

In general, the ratio of ions in the water aerosol corresponds to the ratio of ions in the bulk water, however, some organic substances and inorganic ions are characterized by significant enrichment in the aerosol compared to the bulk water. The bubbles form an aerosol from the water surface film. It is important to note that the concentration of organic and inorganic matter in the surface film differs from that in bulk water [81,82]. It has been widely recognized that both soluble and insoluble components can be substantially enriched in jet drops at a much higher concentration compared to that in the bulk water, up to a thousand-fold [24]. Arising bubbles collect particles from the contaminated water by interception. Small particles with negligible inertia follow the streamlines, with only those close to the bubble surface being intercepted and carried by the bubble. During bubble bursting, bubble cavity collapse excites capillary waves propagating along the bubble cavity surface, which sweep the particles towards the bubble bottom pole. A jet then forms and eventually pinches off, ejecting a top jet drop with an enriched particle concentration [24].

Radionuclides exhibit biochemical and environmental behaviors similar to those of stable analogues. For example, strontium-90 is a calcium analogue, and cesium-137 is a potassium analogue. So, the basis for the mechanism of radionuclide capture in aqueous aerosol has to be very close to the capture of analogue stable ions in the waterbody. The enrichment of Ca^{2+} in sea spray aerosol, relative to seawater, has been identified in a number of ambient marine and nascent sea spray aerosol studies. Cation enrichment has also been observed in the form of Mg^{2+} and K^+ [83]. Ca^{2+} has been observed to be enriched by up to 500 %, relative to seawater. Observations of organic carbon particles that contain inorganic cations (such as Ca^{2+}, Mg^{2+}, and K^+) but no Cl^- have also been reported, suggesting that in some cases these species complex

with organic material [83]. Several authors have referred to a significant organic matter content in the submicrometer fraction of remote marine aerosol. The volume of organic matter in sea salt aerosol has been found to be between 20 and 50% of the volume of sea salt. This level of enrichment corresponds to an enrichment factor for organic material in total marine aerosol (i.e., cumulative across all sizes) of between 7000 and 17000 times that found in bulk seawater [84]. Thus, one of the possible mechanisms of enrichment of an aqueous aerosol with some radionuclides may be the formation of compounds of radionuclides and organic substances in the water of radioactively contaminated reservoirs and the enrichment of water aerosols with the compounds of organic matter with radionuclides.

As a result, the radionuclide composition in the aerosol may differ from the one in the water body over which it is formed [11,85]. In addition, rising to the water surface, gas bubbles are capable of adsorbing and transporting solid particles from bottom sediments in industrial, freshwater, and marine water bodies. Delwiche et al. [23] showed that the transport of particles with methane bubbles from bottom sediments was positively correlated with the volume of gas bubbles. The concentrations of heavy metals and cyanobacteria cells in the particles transported by bubbles were similar to those in bottom sediments. In radioactively contaminated water bodies, the concentration of radionuclides in the bottom sediments is orders of magnitude higher than that in the water bulk. Thus, the transfer of radionuclides with gas bubbles into the aqueous aerosol can be significant.

Our observations allow us to assume that eutrophication and rapid development of phytoplankton in radioactively contaminated reservoirs are associated with bubble emission of gases from bottom sediments and, therefore, with radioactive contamination of the atmosphere around radioactively contaminated water bodies due to the formation of aqueous aerosol as a result of bubble collapse on the water surface. Thus, to reduce the aerosol efflux of radionuclides from radioactively contaminated reservoirs, it is necessary to control the development of phytoplankton and the inflow of biogenic elements that cause eutrophication of such water bodies.

4. Summary

The conducted studies show that radioactive pollution of the atmosphere near radioactively contaminated water bodies occurs due to the formation of aerosols as a result of the collapse of gas bubbles rising from bottom sediments. Gas ebullition in bottom sediments results from the activity mainly of methane-forming and nitrate-reducing bacteria in bottom sediments and depends on the biomass of the substrate, which is a nutrient solution for such bacteria. The main source of substrate for bottom bacteria is phytoplankton (microalgae). Its mass development (blooming) is characteristic of eutrophic (with a high level of organic pollution) water bodies. Thus, the findings of the research indicate the need for a detailed study of the relationship between eutrophication of radioactively contaminated water bodies, the development of phytoplankton in them, the gas ebullition rate as a result of the bacteria activity in bottom sediments, the behavior pattern of gas bubbles, their collapse on the surface of water bodies, and aqueous aerosol formation, as well as modeling of these processes depending on climate and geographical features and conditions to predict and manage the processes of radioactive contamination of the air near radioactively contaminated reservoirs.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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