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ВЛИЯНИЕ БЕНЗ(А)ПИРЕНА НА КАЧЕСТВО ОКРУЖАЮЩЕЙ СРЕДЫ И ЗДОРОВЬЕ НАСЕЛЕНИЯ (НА ПРИМЕРЕ Г. ИВАНОВО)

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В работе приводятся результаты измерений содержания бенз(а)пирена (БП) в снежном покрове на территории г. Иваново. Выявлено, что концентрация БП в контролируемых точках в среднем в 2,7 раза превышает фоновый уровень. Степень загрязнения снежного покрова в г. Иваново значительно меньше, чем в г. Москва (уровень превышения варьируется в диапазоне 2,3 - 89 раз). Оценены уровни поступления (от 0,02 мкг/м^2 до 3,76 мкг/м^2) и плотности выпадения (0,02-1,88 нг/м^2·сут+) БП, которые соизмеримы с величинами, характерными для таких районов, как городские территории Германии и Канады. Интерполяция значений концентраций БП в снеге позволила выявить на территории города аномалии с уровнем содержания бенз(а)пирена 35 нг/л, что в 7 раз выше среднего значения в целом по городу. Полученные данные позволили оценить концентрацию БП в атмосферном воздухе, которая в среднем равна 0,7·10^{-6} мг/м^3, что составляет 0,7 долей ПДКсс. Рассчитаны величины индивидуального канцерогенного риска для здоровья населения, которые составили 0,6·10^{-6} и 1,6·10^{-6}, что соответствует пренебрежимо малому индивидуальному риску (для взрослого населения) или соответствует предельно допустимому уровню (для детей) соответственно. Полученные результаты позволили оценить экологический риск от загрязнения БП снежного покрова, уровень которого соответствует умеренному, однако свидетельствует о потенциальной опасности для здоровья населения, связанной с способностью БП накапливаться в объектах окружающей среды. Установлено, что наиболее вероятным источником поступления бенз(а)пирена на территории города является автомобильный транспорт, вклад топливно-энергетического комплекса должен быть существенно меньше.

Ключевые слова: бенз(а)пирен, ПАУ, снежный покров, канцероген, экологический риск, мониторинг

INFLUENCE OF BENZO(A)PYRENE ON ENVIRONMENTAL QUALITY AND POPULATION HEALTH (BY EXAMPLE OF IVANOVO)

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The paper presents the results of measurements of the content of benzo(a)pyrene (BaP) in the snow cover in the territory of the Ivanovo city. It was revealed that the concentration of BaP at the controlled points is 2.7 times higher than the background level on average. The degree of pollution of the snow cover in Ivanovo is much less than in Moscow (the level of excess varies in the range 2.3 - 89 times). The intake levels (from 0.02 μg/m^2 to 3.76 μg/m^2) and deposition density

(0.02 - 1.88 ng/m^2 · day^{-1}) of BaP are estimated that are commensurate with the values typical for such areas, as urban areas of Germany and Canada. Interpolation the BaP concentrations in snow is allowed to reveal anomalies in the city with BaP content of 35 ng/l, which is 7 times higher than the average value for the whole city. The obtained data made it possible to estimate the BaP concentration in atmospheric air, which on the average is 0.7· 10^{-6} mg/m^3, which is 0.7 fraction of the maximum permissible concentration. The values of individual carcinogenic risk for public health were calculated, which amounted to 0.6·10^{-6} and 1.6·10^{-6}, which corresponds to negligible individual risk (for the adult population) or corresponds to the maximum allowable level (for children), respectively. The obtained results allowed to evaluate the environmental risk from pollution of the snow cover with BaP, the level of which corresponds to a moderate one, however, indicates a potential public health hazard related to the ability of the BaP to accumulate in environmental objects. It is established that the most likely source of BaP is automobile transport. The contribution of the fuel and energy complex should be much less.

Key words: benzo(a)pyrene, polycyclic aromatic hydrocarbons, snow cover, carcinogen, environmental risk, monitoring

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INTRODUCTION

Polycyclic aromatic hydrocarbons are among the priority pollutants of the environment [1]. Many polycyclic aromatic hydrocarbons (PAHs), having a high activity in biological objects, contribute to the occurrence of carcinogenic, teratogenic or mutagenic changes [2].

In Russia, the content of carcinogenic PAHs in environmental objects is normalized only by one, the most dangerous compound – BaP. Hygienic standards of BaP content in environmental objects are given in the literature [3-6].

The priority of BaP is due not only to high toxicity, but also to the scale of environmental pollution. The results of monitoring the level of atmospheric air pollution in the territory of the Russian Federation show that its average concentration in Russia is 1.4 times higher than 1 MAC (maximum allowable concentration), and the maximum – in 5.6 times [7].

BaP transformation products (peroxides, quinones, sulfur and azo derivatives) are also toxic to living organisms [8, 9]. BaP concentration in the atmosphere can serve as an indicator of anthropogenic activity, as well as an indirect factor of human influence on the climate [8-10]. The main sources of BaP supply to the environment are the fuel and energy complex and motor transport. Heavy industry, uncontrolled fires and incineration of waste also make a significant contribution to the overall pollution of the BaP [10]. Most of the BaP is located in the lower layers of the atmosphere in the form of aerosol particles that can penetrate into the respiratory system. Being chemically relatively stable, BaP can be transported over long distances and migrate for a long time in the environment. Thus, monitoring the BaP content in environmental objects, as well as identifying the main channels for its entry and withdrawal, is an urgent task. It should be noted that, in recent years, more and more research has been devoted to studying the processes of migration and transformation of PAHs in the snow cover [11-13].

Snow cover, being a relatively stable mechanical system, is not simultaneously active either chemically or biologically, such as soil. Therefore, snow can be considered an indicator of the previous pollution of the atmosphere [14]. Obviously, the main channel of BaP entering the environment is the combustion processes, followed by air pollution, as a result of which the BaP propagates over long distances from the sources of intake, and the main channel of removal is the processes of dry and wet deposition. Consequently, in the cold period, especially in urban areas, it is solid atmospheric precipitation that is the main process of removing BaP from the atmosphere. Being an effective reservoir for the accumulation of various chemical compounds, including highly toxic organic compounds, such as BaP, in the spring, when all accumulated pollutants are washed out in a short period of thawing. Characteristic for PAH transformation processes in aqueous systems, such as photolysis, bioaccumulation and biodegradation, do not occur in the snow cover, as a result of which, snow causes intense stress on the hydrosphere and the lithosphere [15].

The sampling of snow is fairly simple and does not require complicated equipment as compared to air sampling. Stratified sampling of the snow cover allows to obtain the dynamics of pollutants arrival during the winter season, and one sample over the entire height of the snow cover gives an idea of the contamination for the entire period from the establishment of the shelter to the moment of sampling. It allows to solve the problem of determining the total parameters of contamination (dry and wet deposition). Snow naturally provides concentration of impurities in comparison with the air environment, which simplifies the subsequent task of analyzing pollutants. Also, the study of snow will make it possible to develop models of BaP deposition to identify the most polluted areas and to develop measures aimed at minimizing the impact of highly toxic compounds present in urban thawed effluents.

EXPERIMENTAL

The object of the study was the city of Ivanovo. Climatic conditions in the cold season: the maximum temperature is -0.4 °C; the minimum is -11.7 °C; the average is -5.8 °C. The main type of precipitation from the cold period is snow, and the average amount of precipitation per season is 228 mm. The average depth of the snow cover in the study area in the end of March varied in the range of 0.4-0.6 m. The BaP emissions in the study area are mainly characterized by anthropogenic sources associated with the combustion of fuel at the heat power station and the traffic of road transport, and an additional entry channel may be a transboundary transfer from adjacent territories.

According to recommendations [16], samples of snow were collected in March 2017 (that is at the time when a stable snow cover was established and heavy snowfalls were observed). For snow samples sampling, a standard snow-homer was used, through which individual cores were cut at the points of the snow-covered route cut to the full depth of the snow cover without capturing soil particles. At least 3 snow samples were collected and analyzed at each point in the specified period. Samples of the snow cover were taken from all over the city of Ivanovo, for which the whole area of the city was divided into 12 squares, in each of which 3 to 5 samples were taken. During the entire observation period, 51 samples of snow were analyzed.

The level of background contamination with the BaP was estimated by sampling snow outside the local anthropogenic impact area, at a sufficient distance from the settlements (on the windward side), at least 500 m from the roads.

Analysis of snow samples was carried out according to [17]. The snow samples were melted, the melt water was filtered and extracted in an ultrasonic bath at room temperature with n-hexane from the dried filter, the extract was evaporated and the dry residue was dissolved in acetonitrile. The resulting solutions were analyzed by high performance liquid chromatography (HPLC) using a FLUORAT-02M liquid analyzer as a fluorimetric detector with a column and a pre-column filled with a reversed-phase sorbent "Kromasil C18". Detection of the optical signal was carried out in the excitation wavelength range of 270-360 nm and the registration of 390-450 nm, providing maximum sensitivity and selectivity. The relative error is ± 10% with a confidence level of 0.95 over the entire range of measured concentrations. The random error component does not exceed ± 6.0% (RMS ≤ 3.0%) [18].

The concentration of BaP in the volume of the analyzed snow sample was estimated by the formula:

$$X = \frac{C_c \cdot V_к \cdot Q \cdot 10^3}{V_{pr}}, \text{ ng/l},$$

where: X – concentration of BaP in the sample, mg/dm^3; C_c – concentration of BaP in the sample concentrate, mg/dm^3; V_k is the volume of the sample concentrate, cm^3; V_{pr} – volume of water sample, cm^3; Q is the dilution factor of the sample concentrate.

Conversion of bulk concentration of BaP into bulk (ng/g) was carried out based on snow density equal to 0.25 g/cm^3 [16].

To estimate the level of snow contamination with BaP, the experimental values of the content in the samples were converted into μg/l. Table 1 shows the results of studies at all points of sampling. The results obtained allowed us to estimate the level of BaP supply to the snow cover [15]:

$$m = \frac{C_c \cdot V}{S},$$

where m is the BaP input level, μg/m^2; C_c – concentration of BaP in snow, μg/l; V – volume of melt water, l; S – snow sampling area, m^2.

The calculated levels of BaP receipt are given in Table 2. The results obtained represent the rate of accumulation of BaP in the snow over a time interval of 5 months, which varies from 0.02 μg/m^2 to 3.76 μg/m^2. The obtained values are in good agreement with the studies of other authors [15, 19], the results of which are given in Table 3.

Table 1

The content of BaP in snow samples, ng/l
***Таблица 1*. Содержание БП в пробах снега, нг/л**

Sampling point																
1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**
13.5	3.8	18.2	-	-	10.5	2.5	2.6	-	13.8	1,0	-	8.2	17.3	3.8	-	-
18	**19**	**20**	**21**	**22**	**23**	**24**	**25**	**26**	**27**	**28**	**29**	**30**	**31**	**32**	**33**	**34**
11,4	-	-	-	41.1	-	5.5	-	-	0.1	0.1	-	0.1	-	14.0	0.2	14.6
35	**36**	**37**	**38**	**39**	**40**	**41**	**42**	**43**	**44**	**45**	**46**	**47**	**48**	**49**	**50**	**51**
-	15.6	29.5	0.2	-	-	20.9	-	-	-	-	0.8	-	0.7	-	-	-

Table 2

Incoming levels of BaP, μg/m² (for 5 months)
***Таблица 2*. Уровни поступления БП, мкг/м² (за 5 месяцев)**

Sampling point																
1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**
1.72	0.49	2.31	-	-	1.33	0.32	0.33	-	1.76	0.11	-	1.04	2.20	0.49	-	-
18	**19**	**20**	**21**	**22**	**23**	**24**	**25**	**26**	**27**	**28**	**29**	**30**	**31**	**32**	**33**	**34**
1.45	-	-	-	5.24	-	0.69	-	-	0.01	0.01	-	0.02	-	1.78	0.02	1.86
35	**36**	**37**	**38**	**39**	**40**	**41**	**42**	**43**	**44**	**45**	**46**	**47**	**48**	**49**	**50**	**51**
-	1.99	3.76	0.03	-	-	2.66	-	-	-	-	0.10	-	0.09	-	-	-

Table 3

Comparison of the levels of BaP incoming in the snow cover
***Таблица 3*. Сравнение уровней поступления БП в снежный покров**

Ivanovo city, Russia	Su-Sainte-Marie city, Canada [15]	Berlin, Frankfurt, Dusseldorf, Germany [19]	England (motorway M6) [19]
0.02-3.76	0.26-2.73	0.5-2.0	4.9

The BaP is a compound that can accumulate in the snow cover during the entire observation period, because is characterized by a high partition coefficient in the octanol-water system (low solubility in water) and is present in the atmosphere in the form of an aerosol, especially in urban environments characterized by a high content of suspended particles [15]. That is the main way of migration of BaP in environment is the physical and mechanical one [20]. This fact agrees well with the experimental data obtained in the analysis of suspended substances from snow cover and snow filtrate.

According to research [15] it is precisely the heavy PAHs such as benzo(k)fluoranthene, benzo(a)pyrene, benzo(g,h,i)pyrene, indeno(1,2,3-cd)pyrene, completely are bound by solid particles present in the snow, and the light (phenanthrene, fluoranthene, pyrene, benzo(b)fluoranthene) are present in both the solid and liquid phases. Therefore, the intensity of atmospheric deposition of BaP can be estimated from the dust load [21]:

$$D = P_n \cdot C,$$

where D is the intensity of atmospheric precipitation of BaP with snow, ng/m²·day; P_n – daily dust load, g/m²; C is the concentration of BaP in the solid fraction of snow, ng/g.

Another complex index of snow cover contamination, characterizing the arrival of solid precipitation (dust) on the snow cover for a certain period of time, can be dust deposition density (ρ_d), kg/km²·day^{-1} [22], which is determined by the formula:

$$\rho_d = \frac{p}{S \cdot t},$$

where p is the mass of the dust precipitated by snow or the dust mass in the sample of the solid snow sediment (mg; kg); S – the area of the pit (m²; km²); t – time interval in the days between the moment of sampling and the date of establishment of a stable snow cover (day).

When assessing the level of dust precipitation (water-insoluble forms) in the snow cover, the concentration of suspended solids, received during the cold season, was taken into account. The duration of snow cover was 5 months. The results of the calculations are given in Table 4. It should be noted that the level of pollution of the snow cover in the territory of Ivanovo is much lower than in Moscow [23], where the level of pollution varies from 154 to 5732 ng/g.

Interpolation of BaP concentration values in the snow cover revealed an anomaly in the center of the city, with a BaP content of 35 ng/l at the center to 5 ng/l at the periphery. Also, a minor anomaly is observed in the south-west of the city. It should be noted that it is in these areas that the main facilities of the fuel and energy complex of the city (CHP-2 and

CHP-3) are located, as well as intensive traffic of vehicles, and, probably, they make the main contribution to the pollution of the snow cover. It is possible that local sources are responsible for forming the quality of the environment, which is confirmed by the results of earlier studies on the assessment of soil contamination level in Ivanovo [24]. This assumption can be confirmed in the analysis of seasonal fluctuations in the level of BaP in the components of the environment.

An analysis of the spatial distribution of BaP in the area under study (Fig.) made it possible to estimate the level of excess of atmospheric deposition of BaP (Kd = C_v/C_{av}) relative to the city average of 4.1 ng/l (Table 4), which varies in range 0-2.8 times.

Table 4

The level of contamination of the snow cover in Ivanovo
***Таблица 4.* Уровень загрязненности снежного покрова г. Иваново**

Index	Squares											
	1	2	3	4	5	6	7	8	9	10	11	12
Average concentration of suspended solids in snow, g/l	0.024	0.06	0.05	0,01	0,03	0,04	0,04	0,02	0,03	0,02	0,03	0,02
P_n, kg/км^2·day^{-1}	20.8	51.5	38.3	10.3	27.2	36.2	31.7	17.3	29.0	14.1	21.7	14.7
The average mass concentration of BaP in the snow cover, ng/g (C_m)	41	24	11	0	69	37	2	14	58	66	1	0
Intensity of BaP deposition, ng/m^2·day^{-1}	0.853	1.43	0.42	0	1.88	1.34	0.04	0.24	1.68	0.93	0.02	0
The average volumetric concentration of BaP in the snow cover, ng/l (C_v)	10.1	7.2	3.0	0	1.17	9.32	0.02	3.5	11.3	6	7.5	0.3
The maximum concentration of BaP, ng/l	18.2	13.5	8.5	0	11.2	39.9	0.05	13.2	28.5	19.1	16.2	0.7
The level of excess of atmospheric deposition, K_d	2.5	1.8	0.7	0	0.3	2.3	0.0	0.9	2.8	1.5	1.8	0.1

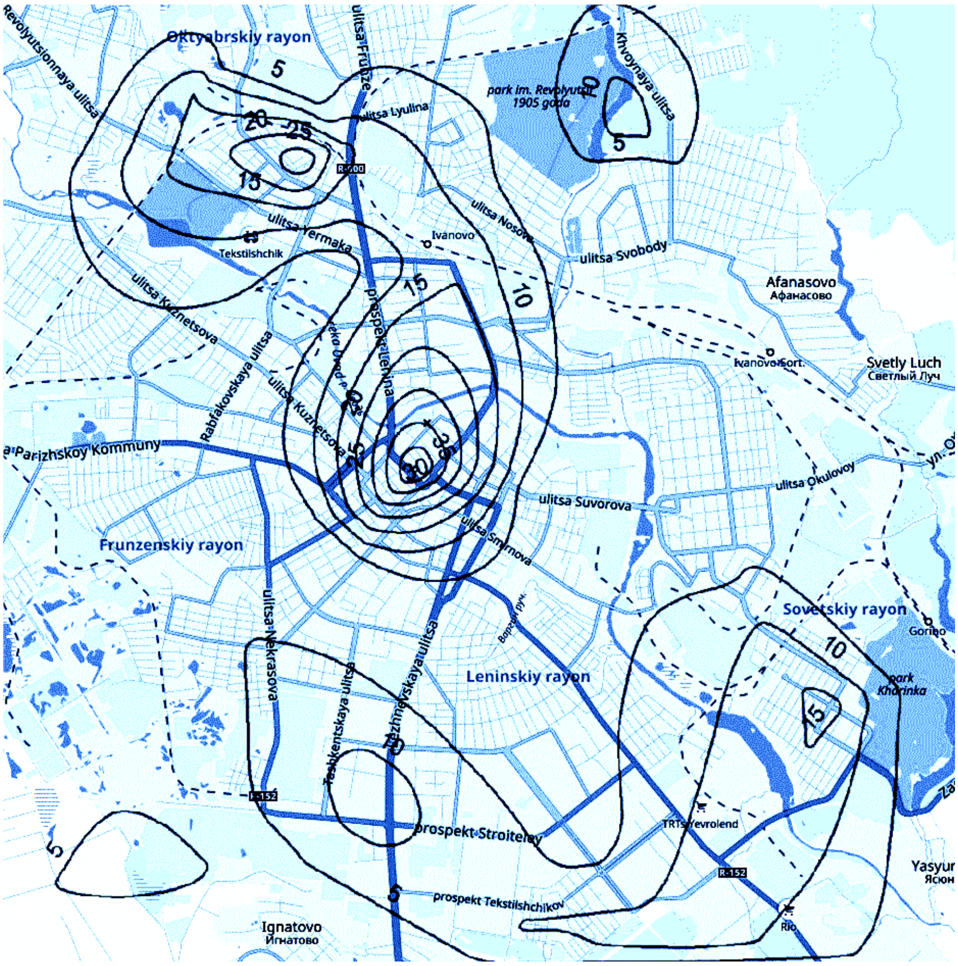

Fig. Spatial distribution of BaP in the city
Рис. Пространственное распространение БП на территории города

Table 5

Assessment of the magnitude of the health risks of different population groups

Таблица 5. Оценка величины риска для здоровья различных групп населения

Population category	SF	CR, m^3/day	EF, days/year	ED, years	BW, kg	AT, years	C_s, mg/m^3	C, mg/m^3	R
men	3.9	15.2	350	70	70	70	0.0044/ 0.0409*	$0.7\cdot10^{-6}$/ $1.3\cdot10^{-6}$	$0.6\cdot10^{-6}$ / $1.1\cdot10^{-6}$
women		11.3		70	70	70			$0.4\cdot10^{-6}$ / $0.8\cdot10^{-6}$
children		8.7		6	14	6			$1.6\cdot10^{-6}$ / $3.0\cdot10^{-6}$

* - the average and maximum concentration of BAP in the city, respectively

* - средняя и максимальная концентрация БП по городу соответственно

Since the snow cover is an indicator of air pollution, the results allow to estimate the magnitude of the risk of adverse effects on public health from the inhalation effect of BaP, which is a substance with carcinogenic properties. This approach allows us to identify the finely-tuned action of the pollutant on the human body and determine the probability of unfavorable consequences for human health [25]. The assessment of the magnitude of individual carcinogenic risk in the territory of the Russian Federation is carried out in accordance with [26] according to the formula:

$$R = LADD\cdot SF,$$

where LADD is the average daily dose of a toxicant that enters the body during life (mg/(kg·day)); SF is the carcinogenic potential factor (mg/(kg·day)$^{-1}$).

$$LADD = \frac{C\cdot CR\cdot ED\cdot EF}{BW\cdot AT\cdot 365},$$

where C is the concentration of the substance in atmospheric air (mg/m^3); CR – rate of the incoming medium (m^3/day); EF – frequency of exposure, days/year; ED – duration of exposure, years; BW – body weight, kg; AT – the period of exposure averaging for carcinogens, years.

The B AP concentration in atmospheric air was determined using the correlation equation obtained by the authors of the article [27]:

$$C_s = 0,83\cdot e^{\left(C/0,00027\right)},$$

where C is the BAP concentration in atmospheric air, μg/m^3; C_s is the concentration of BAP in the snow cover, μg/m^3.

The initial data and the results of calculating the risk values are given in Table 5.

According to [26], risk tolerance criteria are used to characterize the public health risk caused by exposure to environmental pollutants an individual lifelong risk equal to or less than $1\cdot10^{-6}$, characterizes such levels of risk that are perceived by all people as negligible, not differing from ordinary, everyday risks. Individual risk throughout life is more than $1\cdot10^{-6}$, but less than $1\cdot10^{-4}$, corresponds to the maximum allowable risk, i.e. the upper limit of acceptable risk. Thus, the results of the calculation show that the values of the individual risks are either negligible (for the adult population) or correspond to the maximum allowable level (for children).

The authors of the work [28] used the risk calculation methodology (RQ) to assess environmental risk:

$$RQ = \frac{C_{BaP}}{C_{QV}},\quad RQ_{NCs} = \frac{C_{BaP(NCs)}}{C_{QV(NCs)}},\quad RQ_{MPCs} = \frac{C_{Bap(MPCs)}}{C_{QV(MPCs)}},$$

where C_{BaP} is the average concentration of BaP in the investigated sites, ng/l; $C_{BaP(NCs)}$, $C_{BaP(MPCs)}$ – the minimum and maximum concentrations of BaP in the investigated objects, respectively, ng/l; $C_{QV(NCs)}$, $C_{QV(MPCs)}$ – minimum and maximum values of BaP concentrations, leading to environmental risk, ng/l.

Since the values of standard risk values are not established for the snow cover, values for water objects are used for the calculation. The results of the calculation are given in Table 6.

The obtained results show that the level of pollution of the snow cover with BaP corresponds to low environmental risk (Table 7) - the magnitude of environmental risks are at the same level as the risks typical for the Fields Peninsula (Arctic). However, there is probably a potential danger both for public health and for anthropogenically-modified ecosystems in the city of Ivanovo, because of the possibility of the BaP to accumulate in environmental objects.

Table 6

Assessment of environmental risk by the method of risk factors

Таблица 6. **Оценка экологического риска по методике факторов риска**

$C_{QV(NCs)}$, ng/l	$C_{QV(MPCs)}$, ng/l	$C_{BaP(NCs)}$ ng/l	$C_{BaP(MPCs)}$ ng/l	RQ_{NCs}	RQ_{MPCs}	RQ_{NCs}	RQ_{MPCs}
				Ivanovo		Peninsula Fields (Arctic) [28]	
0.5	50	4.4	40.9	8.8	0.8	7.9	0.79

Table 7

Classification of environmental risk [28]

***Таблица 7.* Классификация экологического риска [28]**

Level of environmental risk	RQ_{NCs}	RQ_{MPCs}
Absence of risk	0	
Moderate risk	≥ 1	< 1
High risk		≥ 1

CONCLUSION

The content of benzo(a)pyrene in the snow cover on the territory of the Ivanovo city and its intake levels from the atmosphere are determined. It was found that the BaP concentration exceeds the background level at almost all sampling points on average. The obtained data made it possible to estimate the concentration of BAP in the atmospheric air, which on the average is $0.7 \cdot 10^{-6}$ mg/m^3, which is 0.7% of the maximum permissible concentration (MPC_{AS}).

The individual carcinogenic risk to public health was estimated to be $0.6 \cdot 10^{-6}$ and $1.6 \cdot 10^{-6}$, which corresponds to negligible individual risk (for the adult population) or corresponds to the maximum allowable level (for children), respectively. The calculated values of ecological risk correspond to the moderate level of risk typical for territories with low anthropogenic impact.

It is established that the most likely source of benzo(a)pyrene is automobile transport. The contribution of the fuel and energy complex should be much less.

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Л И Т Е Р А Т У Р А

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