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ОЦЕНКА УРОВНЯ ЗАГРЯЗНЕНИЯ ПОЧВ Г. ИВАНОВО ПОЛИЦИКЛИЧЕСКИМИ АРОМАТИЧЕСКИМИ УГЛЕВОДОРОДАМИ

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В работе приводятся результаты исследования уровня загрязнения почвенного покрова г. Иванова полициклическими ароматическими углеводородами (ПАУ). В пробах почв было идентифицировано 13 из 16 ПАУ, рекомендованных к контролю Агентством по охране окружающей среды США. Из приоритетного перечня в почвенном покрове города отсутствовали ацетонафтилен, ацетонафтен и индено(1,2,3-cd)пирен. Суммарная концентрация ПАУ в почвенном покрове изменяется в диапазоне от 869 до 23343 мкг/кг, при медианном значении 6805 мкг/кг. Анализ распределения концентраций ПАУ относительно их молекулярной массы показал, что основной вклад в суммарное содержание вносят ПАУ с низкой молекулярной массой, вклад которых составляет ~ 80 %. Установлено, что источниками поступления флуорена, хризена, бенз(a)антрацена, бенз(a)пирена и дибенз(a,h)антрацен, могут быть как локальные, так и трансграничные. Показано, что среднее суммарное содержания ПАУ в почвах города в 50 раз выше фонового уровня, поэтому основным каналом их поступления являются все же локальные источники выброса. Выявлено, что вклад канцерогенных ПАУ в их суммарное содержание не превышает 12 %, причем ~ 85 % приходится на бенз(b)флуорантен, а доля бенз(a)пирена не превышает 12,5 %. Проведена оценка величин индивидуального канцерогенного и неканцерогенного рисков для населения города, которая показала, что найденные величины соответствуют пренебрежимо малому индивидуальному риску. Уровень рисков канцерогенных эффектов у населения, проявляющийся при загрязнении почвенного покрова ПАУ, соизмерим с величиной рисков здоровью от загрязнения почвы тяжелыми металлами, а для неканцерогенных эффектов – на порядок ниже.

Ключевые слова: ПАУ, бенз(а)пирен, городские почвы, канцероген, экологический риск, мониторинг

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ESTIMATION OF LEVEL OF SOIL POLLUTION IN IVANOVO CITY BY POLYCYCLIC AROMATIC HYDROCARBONS

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The paper presents the results of a study of the level of soil pollution in the city of Ivanovo with polycyclic aromatic hydrocarbons (PAHs). In soil samples, 13 of the 16 PAHs recommended for control by the US EPA were identified. Acetonaphthylene, acetonaphthene and indeno(1,2,3-cd)pyrene were absent from the priority list in the soil cover of the city. The total concentration of PAHs in the soil cover varies from 869 to 23343 μg/kg, with a median value of 6805 μg/kg. Analysis of the distribution of PAH concentrations relative to their molecular weight showed that the main contribution to the total content is made by PAHs with a low molecular weight, the contribution of which is about 80%. It was found that the sources of fluorene, chrysene, benzo(a)anthracene, benzo(a)pyrene, and dibenz(a,h)anthracene can be both local and transboundary. It is shown that the average total content of PAHs in the soils of the city is 50 times higher than the background level. Therefore, the main channel of their input is still local emission sources. It was revealed that the contribution of carcinogenic PAHs to their total content does not exceed 12%, with ~ 85% accounted for by benzo(b)fluoranthene, and the proportion of benzo(a)pyrene does not exceed 12.5%. An assessment of the values of individual carcinogenic and non-carcinogenic risks for the city population was carried out, which showed that the found values correspond to a negligible individual risk. The level of risks of carcinogenic effects in the population, which manifests itself when the soil cover is contaminated with PAHs, is commensurate with the magnitude of health risks from soil contamination with heavy metals, and for non-carcinogenic effects it is an order of magnitude lower.

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

INTRODUCTION

To identify the level of the load of economic activity on the environment and to develop measures to reduce it, it is necessary to organize and conduct environmental monitoring. One of the aspects of ecological assessment of the state of the environment is the study of the chemical composition of soils and analysis of its changes under the influence of anthropogenic load. Priority environmental pollutants include polycyclic aromatic hydrocarbons (PAHs), which are a large group of organic compounds containing in its structure there are two or more benzene rings [1]. Most PAHs have carcinogenic and mutagenic properties and can concentrate when moving along trophic chains [2]. Due to the resistance of PAHs to degradation in natural environments, as well as their ability to bioaccumulate and high toxicity, they are classified as persistent organic pollutants [1]. Thus, the US Environmental Protection Agency (US EPA) has isolated 16 PAHs, which are the most common in the environment and are highly toxic, including benzo[a]anthracene, benzo[a]pyrene, benzo[k]fluoranthene, benzo(b)fluoranthene, chrysene, dibenzo(a)anthracene, and indo(1,2,3-c,d)pyrene have been classified as carcinogens [3]. PAHs are formed as a result of natural (forest fires, volcanic activity) and anthropogenic (fuel combustion, asphalt production, vehicle exhaust gases) processes [1, 4] and enter the atmosphere, from where, due to the processes of “dry” and “wet” washout, they enter the into the soil, where they are firmly bound by the humic substance [5, 6]. Therefore, for PAHs, soils are the main depositing medium, absorbing up to 90% of the total PAH content in environmental objects [7]. The soil cover is a link between the biotic and abiotic components of terrestrial ecosystems [8], which makes biological processes the main channel for the removal of PAHs from soils.

The soil, especially in the urban environment, is heterogeneous and can be characterized by a significant difference in the concentration of pollutants even when sampling in small areas, which leads to difficulties in assessing the actual level of pollution [9]. To solve this problem, it is necessary to increase the number of sampling points, or to use geostatistical analysis [10]. Geostatistical analysis makes it possible to obtain maps of spatial distribution based on a scattered set of concentrations of pollutants and to estimate the general level of pollution of territories [9-11].

Earlier, in [12, 13], it was shown that the level of atmospheric pollution with PAHs in a number of districts of Ivanovo poses a potential hazard to public health. In this study, the level of PAH contamination of the soil cover in Ivanovo was assessed and the main channels for the intake of these compounds were identified. Also, the assessment of the magnitude of the risk to the health of the population, which manifests itself in soil contamination with PAHs [2].

EXPERIMENTAL PART

To assess the level of PAH content in the soil cover, the territory of Ivanovo was divided into 10 squares with different anthropogenic load, in which sampling was carried out. The level of background pollution was assessed by sampling in a place not subject to local anthropogenic impact (at a sufficient distance from industrial enterprises, as well as at a distance of at least 500 m from highways).

Sampling was carried out in accordance with the requirements of [14] from the surface layer of the soil by the "envelope" method to a depth of 0.30 m. The determination of the PAH content in the samples was carried out by high performance liquid chromatography (HPLC) using a FLUORAT-02 liquid analyzer as a detector [15]. The identification and concentration of monitored compounds in extracts was carried out by comparing them with a known amount of PAHs prepared in an aqueous solution from a standard sample containing 16 PAHs included in the EPA list. The relative error was ± 20% with a confidence level of 0.95 in the entire range of measured concentrations [15].

RESULTS AND DISCUSSION

In soil samples from the city territory, out of 16 priority PAHs, 13 compounds were identified, including 6 carcinogenic (Table 1). It should be noted that not all priority PAHs were found in the analyzed samples, and their concentrations have a wide scatter of values depending on the sampling sites (Table 1).

Table 1

Concentration of PAHs in the soil of the city of Ivanovo (μg/kg)

***Таблица 1.* Концентрация ПАУ в почве города Иваново (мкг/кг)**

PAH	Mean value	Median value	Range of values	Background level	The multiplicity of exceeding the background	
					relative to the mean	relative to the median value
Naphthalene	218.16	140.7	16.6-768.6	20.1	10.9	7.0
Fluoren	37.6	18.3	4.9-176	<6	6.3	3.1
Phenanthrene	3019.19	1993	465.5-12660	< 6	503.2	332.2
Anthracene	3798.85	3622	416.6-11900	132.4	28.7	27.4
Fluorantin	106.33	49.5	3.9-384	< 3	35.4	16.5
Pyrene	407.03	359.15	81.4-877.6	< 20	20.4	18.0
*Chrysen**	3.17	1.1	0.03-37.4	1.6	2.0	0.7
Benz(a)anthracene	6.71	6.9	0.8-17	< 1	6.7	6.9
Benz(k)fluoranthene	7.85	2.4	0.3-82	< 1	7.9	2.4
Benz(b)fluoranthene	847.13	615.1	83.1-2787	< 6	141.2	102.5
Benz(a)pyrene	124.09	64	6.5-831.7	18.4	6.7	3.5
Dibenz(a, h) anthracene	7.05	4.1	1.1-30.2	2.1	3.4	2.0
Benz(ghi)perylene	15.61	12.8	0.6-67.05	< 0.6	26.0	21.3
ΣPAH	8598.77	6805,15	869.1-23342.5	174.6	49.2	38.9

Note: * PAHs with carcinogenic properties are italicized

Примечание: * курсивом выделены ПАУ, обладающие канцерогенными свойствами

According to the research results, the minimum median values of concentrations in soil samples are typical for chrysene (1.1 μg/kg), benzo(k)fluoranthene (2.4 μg/kg) and dibenz(a,h)anthracene (4.1 μg/kg), and the maximum – for anthracene (3622 μg/kg), phenanthrene (1993 μg/kg). It should be noted that the main contribution to soil pollution in the territory of Ivanovo is made by PAHs with a low molecular weight (containing less than 4 aromatic rings in the structure [16, 17]), the content of which is ~ 80% of the total PAH content. From the data of Table 1 it follows that the sources of the following compounds: fluorene, chrysene, benzo(a)anthracene, benzo(a)pyrene, dibenz(a,h)anthracene, can be both local and transboundary. This conclusion is based on the multiplicity of the excess of the average concentrations of these compounds in the soil with the background level. Since the average level of the total content of PAHs in the soil is 50 times higher than the background level (Table 1), the main channel for their entry is local emission sources.

In Russia, only benzo(a)pyrene (BP) is subject to mandatory monitoring of PAHs in soil, the value of the maximum permissible concentration (MPC) in soil for which is taken at the level of 20 μg/kg [18]. The median value of BP concentration in soil in the city is 64 μg/kg, which is 3 times higher than the MPC value (excess of MPC is observed in 75% of soil samples).

The share of carcinogenic PAHs in the soils of the city is about 12% of their total content. Benzo(b)fluoranthene accounts for the maximum proportion of carcinogens and is ~ 85%. BP's share does not exceed 12.5%. The rest of PAHs account for 2.5%.

The results of monitoring the content of PAHs in the soil of Ivanovo (Table 1) make it possible to assess the risk of adverse effects on human health. When the soil is contaminated with chemical compounds, the impact on the human body can occur by inhalation, oral administration, and cutaneous [19]. Since the PAH group includes compounds with both general toxic and carcinogenic properties, it is possible to assess the level of individual lifelong carcinogenic risk and the level of risk from non-carcinogenic effects in chronic exposure. A more detailed procedure for assessing the risk of adverse effects on human health from soil contamination is given in [8]. The results of assessing the risk levels of adverse effects in the population from chemical soil contamination are given in Table 2.

According to [19], the risk acceptance criteria are used to characterize the risk to public health caused by exposure to chemicals that pollute the environment. If the individual risk throughout life is equal to or less than $1\cdot10^{-6}$, then these levels of risk are negligible (no different from the usual, everyday risks). If the individual risk lies in the range of $1\cdot10^{-4}$ - $1\cdot10^{-6}$, then this corresponds to the maximum permissible risk (the upper limit of the acceptable risk). It should be noted that the magnitudes of the risks of carcinogenic effects in the population, manifested when the soil cover is contaminated with PAHs, are at the same level as the magnitudes of health risks from soil contamination with heavy metals, and for non-carcinogenic effects they are an order of magnitude lower.

Because the value of the level of individual risk of both carcinogenic and non-carcinogenic effects in the population, manifested when the soil cover of Ivanovo is contaminated with PAH, does not exceed the maximum permissible values. Consequently, there is no need to develop environmental protection measures aimed at reducing their content in the soil cover. However, the cumulative assessment of the level of the ecological state of the soil cover shows that it is necessary to control the content of both PAHs and heavy metals in specific territories of the Russian Federation, because the level of their content varies widely [20, 21].

Table 2

The results of assessing the risk of adverse health effects in Ivanovo in case of soil contamination

Таблица 2. **Результаты оценки риска неблагоприятных последствий для здоровья населения г. Иваново при загрязнении почвенного покрова**

Population group	Carcinogenic risk level (CR)	Risk level of non-carcinogenic effects under chronic exposure (HQ)	Level of acceptable risk (R) [19]
Polycyclic aromatic hydrocarbons			
Children	$1.55\cdot10^{-7}$	$7.13\cdot10^{-7}$	less than $1\cdot10^{-6}$
Adults	$6.62\cdot10^{-8}$	$3.01\cdot10^{-7}$	
Heavy metals [8]			
Children	$1.23\cdot10^{-7}$	$8.36\cdot10^{-6}$	less than $1\cdot10^{-6}$
Adults	$2.60\cdot10^{-7}$	$5.01\cdot10^{-5}$	

CONCLUSIONS

In the study of the soil cover in Ivanovo, 13 out of 16 PAHs recommended for control were identified. It was revealed that the total content of PAHs exceeds the background level by 50 times, which indicates local sources of their input. It was found that anthracene (3622 μg/kg) and phenanthrene (1993 μg/kg) are the priority compounds that make the main contribution to soil pollution. The values of individual carcinogenic and non-carcinogenic risks to the health of the population were estimated, which amounted to $1.55\cdot10^{-7}$ and $7.13\cdot10^{-7}$, which does not exceed the maximum permissible levels.

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The authors declare the absence a conflict of interest warranting disclosure in this article.

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