

UDC 614.7

DOI: 10.21668/health.risk/2022.1.03.eng

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Research article

## ASSESSMENT OF CARCINOGENIC RISK CAUSED BY ELEVATED 3,4-BENZ(A)PYRENE CONCENTRATION IN SOILS IN AN INDUSTRIAL CITY

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*Apparent destructive changes in soils typically occur in residential areas within urbanized ecological systems formed in large industrial cities. Elevated concentrations of super-toxicants and xenobiotics in soils, polycyclic aromatic hydrocarbons included, create high potential health risks for city population.*

*Our research goal was to assess the carcinogenic risk caused by elevated 3,4-benz(a)pyrene concentrations in soils in Taganrog, an industrial city with its population being about 250 thousand people located in Rostov region. The assessment included individual and population risks. There are two basic reasons which make the research vital. First of all, 3,4-benz(a)pyrene contents in soils in residential areas exceed hygienic standards. Secondly, the incidence of malignant neoplasms has been stably high in the city.*

*We analyzed the results of determining 3,4-benz(a)pyrene concentrations in 384 soil samples taken in 2013–2020 at 19 monitoring posts (located within residential areas close to crossroads with intense traffic, recreational zones, and areas around pre-school children facilities). 3,4-benz(a)pyrene was established to be the priority pollutant in soils in the city with its share contribution to the complex pollution index ( $C_{soil}$ ) being 55.25 %. 3,4-benz(a)pyrene concentrations were higher than maximum permissible ones in 65.63 % of all the examined soil samples; its average and maximum concentrations were equal to 2.298 and 45.525 MPC accordingly. We assessed the individual multi-route carcinogenic risk (CR) caused by elevated 3,4-benz(a)pyrene concentrations in soils. The risk turned out to be high ( $2.4606 \cdot 10^{-3}$ ) and inhalation introduction was established as the priority one (94.84 %).*

**Key words:** social-hygienic monitoring, health risk, chemical soil pollution, 3,4-benz(a)pyrene, malignant neoplasms, carcinogenic risk assessment.

Apparent destructive processes occur in soils in residential areas due to chemical contamination. This situation is typical for most industrial cities. City landscapes are open systems which are closely related to the atmosphere and hydrosphere. High contents of technogenic pollutants, xenobiotics included, in their surface horizons are a highly informative geochemical indicator showing how significantly these landscapes degrade under exposure to anthropogenic technogenic burden. This indicator should be taken into account when examining health disorders among urban population, including studies with their focus on malignant neoplasms. Concentrations of

polycyclic aromatic hydrocarbons (polyarenes or PAHs) are among the most significant indicators included into hygienic and environmental profiles of soils on urbanized territories since many of such chemicals are highly carcinogenic and mutagenic. This creates elevated health risks for population [1–6]. In the Russian Federation 3,4-benz(a)pyrene, a chemical with carcinogenic and mutagenic properties which belongs to the 1<sup>st</sup> hazard category, is often used as the universal indicator and major marker of environmental contamination with PAHs which occurs both due to anthropogenic technogenic sources and natural ones. Organization of additional control over its concentra-

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tion is considered a vital trend in the development of social and hygienic monitoring systems in large cities [3, 7]. A substantial contribution to PAHs emission is made in cities, together with industrial sources and power producing enterprises, by emissions from motor transport due to incomplete fuel combustion in internal-combustion engines and wear and tear of tyres and road surfaces [8–16]. Novocherkasskaya hydro-recirculation power plant is the most powerful energy production in Rostov region and a major source of polycyclic aromatic hydrocarbons (PAHs) emissions into the environment. This is confirmed by results of soils monitoring in a zone influenced by this object since elevated 3,4-benz(a)pyrene levels were detected in roots, stems, and leaves of plants growing there [17]. A substantial contribution to environmental pollution is also made by PAHs which are intensively produced by stove heating, especially when coal is used as a fuel as well as by PAHs coming from various natural sources [18–24]. Soils are a natural depot for polycyclic aromatic hydrocarbons. It is a proven fact that only a part of hazardous chemicals is directly deposited on soil surface from ambient air as well as with precipitations. More than a half of such chemicals primarily accumulate in plants and occur in soils when the vegetation period is over [25, 26]. PAHs absorption from soils through roots of agricultural plants makes for their penetration into food chains with agricultural products and the process has an apparent biomagnification effect. Besides, soils act as the secondary contamination source for ambient air and water by accumulating these pollutants quite actively [27]. Therefore, results produced by monitoring of 3,4-benz(a)pyrene concentrations in urban soils are highly informative integral indicators for creating an objective hygienic and environmental profile of a territory within social and hygienic monitoring activities and environmental analytics [28].

We selected Taganrog as our research object. The city is a large industrial center located in Rostov region; its population is about 250 thousand people. The situation with cancer incidence is stably unfavorable in Taganrog [29]. Thus, average long-term frequency

calculated for all localizations of malignant neoplasms over the last 15 years is equal to 480.11 ‰ which is by 1.25 times higher than the same indicator calculated for the whole Rostov region (373.98 ‰). The city holds the first rank place in the region as per this parameter. Average annual growth in long-term total cancer incidence in Taganrog amounted to +0.28 % in 2006–2020. These stable ascending trends in frequency have been detected for such localizations of malignant neoplasms as oral cavity and pharynx (average annual growth in long-term trend is +1.15 %), rectum (+0.57 %), skin (without melanoma) (+1.89 %), breast (+1.95 %), uterine neck (+3.44 %), prostate (+3.95 %), thyroid gland (+4.42 %), and malignant lymphomas (+1.04 %). The first rank place among oncologic pathologies belongs to malignant neoplasms of skin (without melanoma) with their specific weight being 15.08 %; the second, malignant neoplasms of breast (12.50 %); the third, malignant neoplasms of trachea, bronchi and lung (9.14 %). The next are malignant neoplasms of colon, prostate, and stomach, 6.84, 6.14 and 6.04 % accordingly. Frequency of malignant neoplasms taken in long-term dynamics grew by 2.06 times from 260.35 ‰ in 1986 up to the maximum level registered in 2017 when it was equal to 535.77 ‰ (Figure).

**Our research goal** was to assess an individual and population carcinogenic risk caused by elevated 3,4-benz(a)pyrene concentrations in soils in residential areas in Taganrog, Rostov region, based on the results produced by social and hygienic monitoring activities in 2013–2020.

**Material and methods.** The research was based on the results of examining 384 soil samples which were tested to determine 3,4-benz(a)pyrene contents in them as well as on data taken from the report forms No. 35 “Data on patients with diagnosed malignant neoplasms” over 1985–2015 and No. 7 “Data on malignant neoplasms” over 2016–2020.

Soil samples were taken in 2013–2020 at 19 monitoring points located in residential areas close to crossroads with intense traffic (8 points), areas around municipal preschool

children facilities (8 points), and recreation zones in Taganrog (3 points) including the embankment, a park and a beach. Mass 3,4-benz(a)pyrene concentrations in soils were determined with high performance liquid chromatography (HPLC) using “Stayer” stationary liquid chromatographer with a fluorometer. When assessing a share contribution made by 3,4-benz(a)pyrene into the total soil contamination, we calculated values of separate indicators showing contents of the examined pollutants (in MPC), their concentration coefficients taking into account regional background levels (F), the complex indicator of soil contamination ( $C_{\text{soil}}$ ) and the summated technogenic contamination coefficient ( $Z_c$ ). Results of the studies accomplished by experts from the Academy for Biology and Biotechnology of the D.I. Ivanoskiy’s Southern Federal University in the “Persianovskaya zapovednaya step” State Soil Reserve were taken as regional background levels

of 3,4-benz(a)pyrene contents in common chernozem in Rostov region. Carcinogenic risks were assessed in accordance with the Guide R 2.1.10.1920-04<sup>1</sup>.

We applied our own specialized software “Turbo oncologist”, version 2.01, to create databases and to perform their statistical analysis. The software includes a package which allows implementing algorithms for retrospective epidemiological analysis of frequencies, structure, long-term dynamics, and spatial characteristics based on methods used for partial and complex assessment of an actual (epidemiological) risk. We also used “Turbo Dynamics” software package, version 1.02, (to analyze long-term trends in dynamics, to perform middle-term extrapolation prediction and modeling, non-linear regression and correlation analysis); as well as IBM SPSS Statistics (“Statistical Package for Social Science”) professional statistical software, version 19.0.

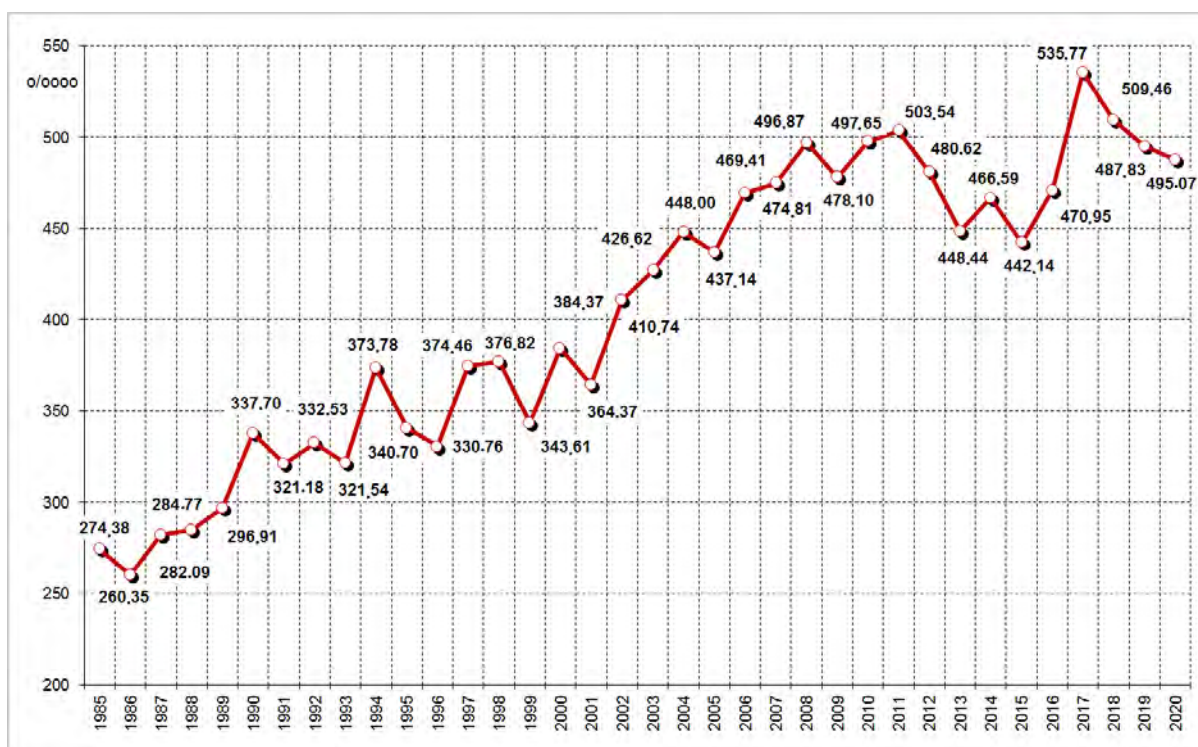


Figure. Total cancer incidence among population in Taganrog, Rostov region, taken in dynamics over 1985–2020

<sup>1</sup> R 2.1.10.1920-04. Rukovodstvo po otsenke riska dlya zdorov'ya naseleniya pri vozdeystvii khimicheskikh veshchestv, zagryaznyayushchikh okruzhayushchuyu sredu [Human Health Risk Assessment from Environmental Chemicals]. Moscow, The Federal State Sanitary and Epidemiological Surveillance Center of the RF Public Healthcare Ministry, 2004, 143 p.

Table 1

Soil contamination with 3,4-benz(a)pyrene in Taganrog, Rostov region, taken in dynamics over 2013–2020

Indicators	Years								2013–2020
	2013	2014	2015	2016	2017	2018	2019	2020	
A share of samples with concentrations higher than MPC, %	72.9	70.8	52.1	62.5	79.2	58.3	66.7	62.5	65.63 ± 4.78
The highest concentration, mg/kg	0.3710	0.1542	0.7610	0.9105	0.1799	0.2170	0.0440	0.1710	0.9105
Contamination indicators (I <sub>max</sub> , MPC)	18.550	7.710	38.050	45.525	8.995	10.850	2.200	8.550	45.525
Average annual concentration (C <sub>ave</sub> , mg/kg)	0.0484	0.0514	0.0471	0.0511	0.0476	0.0510	0.0227	0.0484	0.0460 ± 0.0103
Average contamination indicator (I <sub>ave</sub> , MPC)	2.422	2.570	2.353	2.553	2.382	2.550	1.136	2.422	2.298
C <sub>soil</sub>	4.312	4.590	4.113	4.535	4.248	4.806	3.026	4.312	4.160
A share contribution into C <sub>soil</sub> , %	56.17	56.00	57.20	56.29	56.06	53.05	37.55	56.17	55.25
The summated technogenic contamination coefficient (Z <sub>c</sub> )	6.651	7.120	6.496	7.117	6.890	6.301	6.651	6.651	6.978

**Results.** The results of examinations performed in 2013–2020 indicate that soils in Taganrog are heavily contaminated with 3,4-benz(a)pyrene. Thus, its concentrations were higher than MPC (0.02 mg/kg) in 65.63 ± 4.78 % of 384 examined soil samples and the average actual concentration of the pollutant was 0.0460 ± 0.0103 mg/kg (2.298 MPC). The maximum detected 3,4-benz(a)pyrene concentration was 0.9105 mg/kg (45.525 MPC). We established the priority role played by this pollutant in soil contamination in the city with its share contribution into C<sub>soil</sub> being 55.25 % (Table 1).

The highest contamination with 3,4-benz(a)pyrene was detected in residential areas located close to crossroads with intense traffic (the first group of monitoring points) where a specific share of samples with the pollutant concentrations being higher than MPC amounted to 71.60 ± 5.64 %, over the last eight years and its average content was 0.0475 ± 0.0103 mg/kg (2.376 MPC). The second rank place as per frequency of concentrations exceeding MPC belonged to soils in recreation zones (58.72 ± 9.45 %); however, an average

3,4-benz(a)pyrene content turned out to be even higher there than at monitoring points from the first group (0.0498 ± 0.0241 mg/kg). The Primorskii city park is a recreation zone with the extremely high soil contamination with 3,4-benz(a)pyrene (up to 45.525 MPC); the park is located in a zone which was once influenced by emissions from production workshops of “Taganrogskii harvester plant” PLC and “Kirpichnik” PLC, two large industrial enterprises which are both closed at present. Therefore, soil contamination as per average 3,4-benz(a)pyrene contents can be considered “hazardous” in the first and second monitoring groups according to the assessment criteria applied in evaluation of chemical contamination and stipulated by the Sanitary Rules and Standards SanPiN 2.1.7.1287-03<sup>2</sup>. The third group of monitoring points was located on areas around municipal preschool children facilities. A specific share of soil samples with 3,4-benz(a)pyrene concentration exceeding MPC detected there was 36.00 ± 19.81 %; the average pollutant content was 0.0176 ± 0.0094 mg/kg (0.882 MPC) and its maximum content didn't exceed 3.105 MPC (Table 2).

<sup>2</sup> SanPiN 2.1.7.1287-03. Sanitarno-epidemiologicheskie trebovaniya k kachestvu pochvy: Sanitarno-epidemiologicheskie pravila i normativy [The sanitary-epidemiologic requirements to soil quality: Sanitary-Epidemiological Rules and Standards]. Moscow, The Federal State Sanitary and Epidemiological Surveillance Center of the RF Public Healthcare Ministry, 2004, 16 p.

Table 2

Soil contamination with 3,4-benz(a)pyrene in Taganrog, Rostov region, detected at different monitoring points taken in dynamics over 2013–2020

Indicators	Total at all monitoring points	Including:		
		Residential areas with intense traffic	Recreation zones (embankment, park)	Areas around preschool children facilities
A number of taken soil samples	384	250	109	25
A share of samples with concentrations higher than MPC (%)	65.6 ± 4.8	71.6 ± 5.6	58.7 ± 9.5	36.0 ± 19.8
Average concentration (Cave, mg/kg)	0.0460 ± 0.0103	0.0475 ± 0.0103	0.0498 ± 0.0241	0.0176 ± 0.0094
Contamination indicator as per average concentration (Iave, MPC)	2.298	2.376	2.492	0.882
Lower confidence limit of average concentration ( $p < 0.05$ )	0.0357	0.0372	0.0256	0.0082
Upper confidence limit of average concentration ( $p < 0.05$ )	0.0563	0.0579	0.0740	0.0270
Minimal concentration (Cmin, mg/kg)	0.0026	0.0038	0.0026	0.0040
Contamination indicator as per minimal concentration (Imin, MPC)	0.130	0.190	0.130	0.200
Maximum concentration (Cmax, mg/kg)	0.9105	0.7610	0.9105	0.0621
Contamination indicator as per maximum concentration (Imax, MPC)	45.525	38.050	45.525	3.105

We should note that a significant part of residential areas in Taganrog is private housing with land spots near houses where people grow agricultural products (vegetables, fruits, berries, etc.) and there is a high probability that soils there are heavily contaminated with 3,4-benz(a)pyrene. Given that, it is truly vital to assess carcinogenic risks associated with chemical contamination of soils in the city when social and hygienic monitoring is performed.

We assessed carcinogenic risks associated with elevated 3,4-benz(a)pyrene contents in city soils as per results of laboratory tests performed in 2013–2020. According to recommendation provided in [30–33] a carcinogenic risk was assessed taking into account substantiated maximum exposures calculated on the basis of upper 95 % confidence limits of average 3,4-benz(a)pyrene concentrations.

Individual multi-route carcinogenic risk rates (CR) were calculated within an exposure scenario of a typical residential area as the sums of carcinogenic risk rates caused by exposure to 3,4-benz(a)pyrene contents in soils

at monitoring points taking into account oral (CRo), inhalation (CRi) and dermal (CRd) introduction into the body.

When calculating an individual carcinogenic risk caused by oral 3,4-benz(a)pyrene introduction from soils (CRo), we took a slope factor (SFo) value equal to  $7.3 \text{ (mg/(kg}\cdot\text{day))}^{-1}$ . CRo values calculated for separate calendar years within the examined period varied from  $2.8312 \cdot 10^{-5}$  to  $6.4046 \cdot 10^{-5}$ , and the average long-term CRo rate calculated on the basis of the upper confidence limit ( $p < 0.05$ ) of the average 3,4-benz(a)pyrene concentration turned out to be equal to  $7.2075 \cdot 10^{-5}$  (Table 3).

Therefore, CRo falls within the second range of lifetime individual risk (maximum permissible risk or the upper limit of acceptable risk). This makes the constant control of it essential. The share contribution made by CRo to the structure of the individual multi-route carcinogenic risk (CR) is equal to 2.93 %. The lifetime population carcinogenic risk (an additional to the background absolute number of malignant neoplasms which can occur during an average lifetime of a person equal to 70 years

Table 3

Carcinogenic risk for people living in Taganrog, Rostov region, caused by 3,4-benz(a)pyrene (3,4-BP) concentrations in soils in 2013–2020

Carcinogenic risk rate	Individual multi-route carcinogenic risk	Carcinogenic risk caused by exposure to 3,4-BP in soils (an exposure scenario of a residential area)		
		Oral introduction (CRo)	Inhalation introduction (CRi)	Dermal introduction (CRd)
Risks as per average concentrations in 2013–2020	$2.0203 \cdot 10^{-3}$	$5.9179 \cdot 10^{-5}$	$1.9161 \cdot 10^{-3}$	$4.4976 \cdot 10^{-5}$
Risks as per upper confidence limits of average concentrations in 2013–2020	$2.4606 \cdot 10^{-3}$	$7.2075 \cdot 10^{-5}$	$2.3337 \cdot 10^{-3}$	$5.4777 \cdot 10^{-5}$
Carcinogenic risk structure (%)	100.00	2.93	94.84	2.23

Table 4

Population carcinogenic risk taken as per the upper confidence limits of average concentrations ( $p < 0.05$ ) over 2013–2020

Indicator	Total risk PCR	Oral exposure (PCRo)	Inhalation exposure (PCRi)	Dermal exposure (PCRd)
Expected number of malignant neoplasms during the whole lifetime (70 years) calculated per 252,309 people (average city population)	621	18	589	14
Annual population carcinogenic risk per 100 thousand people	3.515	0.103	3.334	0.078
Contribution made by 3,4-BP in soils to cancer incidence (%)	0.729	0.021	0.692	0.016

calculated as per the average population number in Taganrog which is 252,309 people) is equal to 18 cases under oral introduction of 3,4-benz(a)pyrene from soils (PCRo). Accordingly, the annual population carcinogenic risk is equal to 0.103 ‰ per 100 thousand people and the share contribution made by 3,4-benz(a)pyrene in soils to the overall cancer incidence under oral introduction equals to 0.021 % (Table 4).

Since there are no data on gastro-intestinal absorptivity (GIABS) for 3,4-benz(a)pyrene, its value was taken as equal to 1.0 which means that 100 % of the pollutant quantity introduced into the body is absorbed [30, 31]. Therefore, when calculating the individual carcinogenic risk under dermal introduction, we took the slope factor value (SFd) as equal to the slope factor value under oral exposure (SFo), that is,  $7.3 \text{ (mg/(kg}\cdot\text{day))}^{-1}$ . Rates of the annual individual carcinogenic risk under dermal exposure (CRd) over the last eight years varied from  $2.1517 \cdot 10^{-5}$  to  $4.8675 \cdot 10^{-5}$

and its average long-term rate was  $5.4777 \cdot 10^{-5}$  (the second range of the individual lifetime risk). The specific share of CRd in the individual multi-route carcinogenic risk (CR) was equal to 2.23 %. The lifetime population carcinogenic risk (PCRd) for population in Taganrog equals to 14 cases over 70 years; the annual population carcinogenic risk per 100 thousand people is 0.078 ‰; the share contribution made by 3,4-benz(a)pyrene in soils to the overall cancer incidence is 0.016 % under dermal introduction.

When assessing the individual carcinogenic risk under inhalation exposure to 3,4-benz(a)pyrene in soils (CRi), we took a slope factor (SF<sub>i</sub>) equal to  $3.9 \text{ (mg/(kg}\cdot\text{day))}^{-1}$ . When calculating its vaporization factor (VF), we took a value equal to  $0.034 \text{ Pa}\cdot\text{m}^3/\text{mol}$  as the value of the Henry's law constant. We established that the annual CR<sub>i</sub> rates varied from  $9.1672 \cdot 10^{-4}$  to  $2.0737 \cdot 10^{-3}$ ; CR rates, from  $9.6655 \cdot 10^{-4}$  to  $2.1865 \cdot 10^{-3}$ . Their average long-term values were  $2.3337 \cdot 10^{-3}$  and

$2.4606 \cdot 10^{-3}$  accordingly thus falling within the fourth risk range (more than  $1.0 \cdot 10^{-3}$ ). This means there is *De manifestis Risk*, which is unacceptable, both for population in general or specific occupational groups. Therefore, inhalation introduction of 3,4-benz(a)pyrene from soils is the priority one with the share contribution made by CRi to the individual multi-route carcinogenic risk (CR) being equal to 94.84 %. The lifetime population carcinogenic risk for population in Taganrog under inhalation introduction of 3,4-benz(a)pyrene from soils (PCRi) is equal to 589 cases over 70 years; the lifetime multi-route population carcinogenic risk (PCR), 621 cases; the annual population carcinogenic risks per 100 thousand people are equal to 3.334 ‰ and 3.515 ‰ accordingly with the share contribution made by 3,4-benz(a)pyrene in soils to the overall cancer incidence being equal to 0.692 % and 0.729 % accordingly (Table 4).

Therefore, we corroborated that soils in Taganrog were heavily contaminated with 3,4-benz(a)pyrene and the pollutant created high carcinogenic risks for the city population. This chemical has several peculiarities: it tends to accumulate in soils and to persist there preserving its biological activity for a long period of time; it is able to create secondary contami-

nation of ambient air and to penetrate into food chains through roots of agricultural plants; its carcinogenic and mutagenic effects on the human body are well-proven. Given that, it is advisable to include dynamic monitoring of 3,4-benz(a)pyrene concentrations into the list of mandatory soil examinations within social and hygienic monitoring activities. There are some promising trends in developing information and analytical support for hygienic assessment of chemical contamination in soils within the social and hygienic monitoring system. They include integration of profile databases into regional geoinformation systems (GIS) and new software enabling assessment of risks related to developing carcinogenic effects caused by chemical contamination in soils in Rostov region. There are plans to perform sampling studies on 3,4-benz(a)pyrene contents in fruits and vegetables grown on private land spots on the territory of Taganrog provided that such studies are included into the program of social and hygienic monitoring in the region.

**Funding.** The research was not granted any financial support.

**Conflict of interests.** The authors declare there is no any conflict of interests.

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*Deryabkina L.A., Marchenko B.I., Tarasenko K.S. Assessment of carcinogenic risk caused by elevated 3,4-benz(a)pyrene concentration in soils in an industrial city. Health Risk Analysis, 2022, no. 1, pp. 27–35. DOI: 10.21668/health.risk/2022.1.03.eng*

Received: 14.12.2021

Approved: 21.02.2022

Accepted for publication: 21.03.2022