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Research article

INTEGRAL ASSESSMENT OF FOOD PRODUCTS CONTAMINATION WITH PRIORITY POLYAROMATIC HYDROCARBONS

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Polyaromatic hydrocarbons (PAH) occur in the environment as complex mixtures and each congener has different carcinogenic and mutagenic activity.

Our research goal was to accomplish an integral assessment of food products contamination with priority PAH basing on their determination with high precision procedures.

We validated a procedure for determining the said substances and hygienically assessed contamination of certain food products with benzpyrene, as well as with different carcinogenic and mutagenic PAH equivalents taking into account samples with low contamination. Quantitative determination limit for benz(a)anthracene and benzpyrene was fixed at 0.01 µg/kg; benz(b)fluoranthene and chrysene, 0.1 µg/kg. Detection limit for benz(a)anthracene and benzpyrene amounted to $0.003 \ \mu g/kg$ in our research; for benz(b)fluoranthene and chrysene, $0.03 \ \mu g/kg$. A procedure for integral assessment of contamination with the examined compounds allowed us to calculate benz(a)anthracene, benzpyrene, benz(b)fluoranthene, and chrysene contents in certain food products taking into account mixture of the examined substances, their individual contributions into aggregated contamination, and their different toxic and mutagenic activity. Median food products contamination with benzpyrene amounted to 0.0065–0.42 µg/kg; PAH taking into account carcinogenic equivalents, 0.03–0.55 µg/kg; PAH based on mutagenic equivalents, 0.04–0.81 µg/kg. Maximum concentrations of benzpyrene and PAH based on carcinogenic and mutagenic equivalents are due to a combination of subsequent technological processes that make for occurrence of the examined substances and also due to physical and chemical properties of the examined food products.

Key words: risk assessment, integral assessment, polycyclic aromatic hydrocarbons, contamination, food products, congeners, toxic equivalent, mutagenic equivalent.

providing sanitary-epidemiologic welfare in Belarus. Food products may get contaminated with polyaromatic hydrocarbons (PAHs), substances with carcinogenic and mutagenic properties, due to surface contamination as well as due to these substances occurring when food products are being manufactured [1, 2].

PAHs are toxic organic compounds with two or more condensed aromatic rings. Experts established relations between exposure

Food product safety is a key element in to PAHs mixtures and unfavorable outcomes at birth, neurologic and behavioral effects, and poorer fertility [3, 4]. Experiments performed on animals allowed revealing that certain PAHs were carcinogenic and made for occurrence of some oncologic diseases including breast cancer, lung cancer, and malignant neoplasms in distal intestines. More than 100 PAHs congeners have been examined so far and 16 out of them are determined as priority contaminants by the US Environmental Protection Agency due to their toxic



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properties; 7 PAHs are considered potentially carcinogenic for people [3]. According to a classification by the International Agency for Research on Cancer, benz(a)pyrene belongs to Group 1 carcinogens (is carcinogenic for humans); benz(a)anthracene, chrysene, and benz(b)fluoranthene belong to group 2B carcinogens (are possibly carcinogenic for humans) [4–8, 10].

There are multiple natural and anthropogenic PAHs sources in the environment. These substances occur when organic compounds are being burnt or during technological processes applied to produce food products [10–13]. PAHs contents in food products depend on procedures applied to treat and manufacture specific food products (smoking, grilling, application of smoking flavoring agents, frying, etc.) and on quantitative and qualitative characteristics of a technological process as well. Food products that are primary components in any ration (milk products and bread) get contaminated due to PAHs migration along food chains and surface contamination of grain cultures [1, 3, 6, 9–11].

When identifying hazards and describing risks related to PAHs alimentary introduction, it is necessary to take into account carcinogenic and mutagenic equivalence factors that are used to describe overall toxicity and mutagenicity of the chemicals being considered in this work [3, 4]. All the above mentioned indicates it is quite relevant to perform integral assessment of food products contamination with PAHs.

Our research goal was to integrally assess food products contamination with PAHs basing on their determination with high precision techniques.

Data and methods. PAHs were determined according to the State Standard GOST $31745-2012^1$. The said methodical guide contains the following parameters: quantitative

determination limit amounts to 2.0 µg/kg, and detection limit varies from 0.1 to 5.0 μ g/kg for specific PAHs, for example, 1.0 µg/kg for benz(a)anthracene; 1.0 µg/kg for chrysene; $\mu g/kg$ for benz(b)fluoranthene; 0.25 and 0.5 µg/kg for benz(a)pyrene. Similar requirements are fixed to these PAHs contents in the European Union. However, according to the European Union legislation², detection limit for all the above mentioned PAHs should be ≤ 0.30 µg/kg and quantitative determination limit should be $\leq 0.90 \ \mu g/kg \ [3, 14, 15]$. Given that, it seemed necessary to validate the procedure in order to achieve better sensitivity, as well as precision and accuracy of measurements aimed at determining benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene contents. We calculated linearity, repeatability, intermediate precision, accuracy (extraction), uncertainty, detection limit and quantitative determination limit of our procedure and compared them to the above mentioned EU Commission Regulation.

Contaminants were quantitatively determined via absolute calibration. All the obtained data were processed with Agilent Open LAB CDS software package. To build up a calibration curve, we established dependence between peak square and a corresponding concentration of benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene in calibration solutions. Contents of each substance in PAHs mixture amounted to 4 μ g/cm³. Calibration solutions of concentrations equal to 0.0004; 0.0008; 0.0040; 0.0100 and 0.0200 μ g/cm³ were prepared via diluting.

To calculate calibration curves, we applied the least-square procedure. Correlation coefficient R^2 was a linearity criterion.

Repeatability and intermediate precision as parameters showing actual precision of a procedure were determined in accordance with State Standard ISO 5725-2-2002, item 7, and

¹ GOST 31745-2012. Food products. Determining polycyclic aromatic hydrocarbons with high performance liquid chromatography. Minsk, Gosstandart Publ., 2014, 8 p. (in Russian).

² Commission Regulation (EU) No. 836/2011 of August 19 2011, that fixes procedures for sampling and analyzing aimed at official control over contents of lead, cadmium, mercury, non-organic tin, 3-MCPD and benz(a)pyrene in food products. *EuroLex*. Available at: https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32011R0836&qid=1574851930841&from=EN (27.08.2019).

State Standard ISO 5725-3-2002, item $8.2^{3,4}$. Shifting was assessed as per State Standard ISO 5725-4-2002, item 5⁵. To assess precision, we obtained statistic data via analyzing working samples of canned fish (sprats), vegetable oils, spread, and mayonnaise.

Accuracy of results obtained with the procedure was studied with validation examinations under repeatability conditions via analyzing samples containing benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene in a quantity equal to 0.01 mg/kg.

Extraction degree was calculated as a ratio of measured benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene contents in a sample with addition and calculated benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene contents in a sample with addition according to experimental data.

Contamination level is a variable used to assess exposure. And here PAHs can occur in food products in quantities lower than detection limit or quantitative determination limit. In such cases substitute values are required. Modeling is applied when a share of product samples with low contamination is higher than 60 %. Otherwise, non-significant contamination levels are considered to be equal to zero. We applied models that involved assessing upper and lower limits as well as an average level. Values for lower limit were equated with detection limit; for upper limit, with quantitative determination limit; and as for an average level, we took simple mean of the above mentioned parameters for it 6 [4, 10].

We examined benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene contents in 278 food product samples. PAHs contents in coffee and tea as drinks were calculated taking into account a percent of them that was transferred from coffee beans and tea leaves into liquid drinks [16, 17] (Table 1). A % of PAHs that is transferred from initial coffee beans and tea leaves into liquid drinks

Food product	% of transferred PAH
Dark roasted ground coffee	7.0
Light roasted ground coffee	9.0
Instant coffee	0
Black tea	0.86
Green tea	5.9
Tea drink	1.0

All the obtained data were statistically processed with STATISTICA 12.0 software program. We applied Shapiro-Wilkes test and Kolmogorov-Smirnov test with Lilliefors correction to assess normal distribution. When significance was p < 0.05, data distribution was considered to be non-parametric. Hygienic assessment of examined products contamination with PAHs was performed using median (Me), interquartile range (25 %÷75 %), and 95 % percentile (95P). Validity of discrepancies between upper and lower limits compared to an average food products contamination as per median was determined at p < 0.05 as per Mann-Whitney U-test.

Basing on application instruction No. 004-1618 and according to the research works [18, 19], we performed an integral assessment of food products contamination with PAHs mixture⁶.

Results and discussion. Performed validation tests allowed assessing uncertainty in measured mass concentration of benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene that included the following (Table 2):

- a) repeatability factor;
- b) building up and using calibration curve;
- c) sample preparation for analysis.

³ GOST R ISO 5725-2-2002. Accuracy (correctness and precision) of measurement procedures and results. Part 2. The basic procedure for determining repeatability and reproducibility of a standard measurement technique. Moscow, Standartinform Publ., 2009, 42 p. (in Russian).

⁴ GOST R ISO 5725-3-2002. Accuracy (correctness and precision) of measurement procedures and results. Part 3. Intermediate precision parameters of a standard measurement technique. Moscow, GOSSTANDART Rossii Publ., 2002, 29 p. (in Russian).

⁵ GOST R ISO 5725-4-2002. Accuracy (correctness and precision) of measurement procedures and results. Part 4. Basic procedures for determining correctness of a standard measurement technique. Moscow, Standartinform Publ., 2009, 24 p. (in Russian).

⁶The procedure for hygienic assessment of polyaromatic hydrocarbons contents in food products: application guide No. 004-1618. Approved by the Deputy Minister, Chief Sanitary Inspector of Belarus on June 22, 2018. Minsk, 2018, 14 p. (in Russian).

Source	Relative standard, %					
Source	Benz(a)anthracene	Benz(b)fluoranthene	Chrysene	Benz(a)pyrene		
Repeatability of measurement results in a sample	5.28	4.20	2.16	6.29		
Sample treatment	3.53	3.53	3.53	3.53		
Extraction	2.35	1.72	1.11	2.31		
Building up and using a calibration curve	10.3	6.2	8.6	10.8		
Total standard uncertainty	21.46	15.65	15.4	22.93		
Maximum extended measurement uncertainty $(k = 2)$	42.92	31.30	30.8	45.86		

Uncertainty budget in PAH measurement

Table 3

Metrologic properties of the procedure for PAHs determination

Metrologic property	Benz(a)anthracene	Benz(b)fluoranthene	Chrysene	Benz(a)pyrene
QDL, µg/kg	0.01	0.10	0.10	0.01
Repeatability, %	5.69	5.94	3.05	8.90
Intermediate precision, %	6.92	7.11	4.57	9.18
Repeatability limit, %	20.89	16.63	8.54	24.92
Intermediate precision limit, %	26.71	19.91	12.80	25.70
Determination accuracy, %	86.38	90.24	94.24	89.17
Shifting, %	2.35	1.72	1.11	2.31
Extended uncertainty for a range of measurements, %	42.92	31.30	30.80	45.86

The performed research allowed establishing metrologic properties of the procedure for PAHs determinaiton; they are given in Table 3.

Therefore, performed validation of the procedure allowed establishing quantitative determination limit; it amounted to 0.01 µg/kg for benz(a)anthracene and benz(a)pyrene and to 0.01 µg/kg for benz(b)fluoranthene and chrysene. The European Food Safety Authority (EFSA) recommends the ratio of detection limit to quantitative determination limit to be not less than 3.3; or detection limit should be equal to 1/10 of a standard deviation from a background signal. Consequently, detection limit for benz(a)anthracene and benz(a)pyrene amounts to 0.003 μ g/kg in our research; and it amounted to 0.03 μ g/kg for benz(b)fluoranthene and chrysene. It corresponds to requirements fixed in the EU Commission Regulation.

Having applied the conventional procedure taking into account its validation, we quantitatively determined benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene in certain food products. Number of samples that contained these substances in quantities lower than quantitative determination limit varied from 0 to 81.4 % for oils and vegetable fats, processed cacao products, smoked fish and meat products and cheese [20]; from 0 to 90 % for bread and bakery; from 36.7 to 93.3 % for milk products; from 0 to 43.3 % for tea; from 50 to 100 % for coffee.

We modeled samples with low contamination via applying substitute values instead of contamination levels lower than quantitative determination limit and we also applied toxic and mutagenic equivalent factors to characterize benz(a)anthracene, benz(b)fluoranthene, and chrysene contents [20]. It allowed us to determine range of contamination levels for the examined food products with both benz(a)pyrene itself and in values equivalent for the contaminant. Benz(a)pyrene contents in various food products are given in Table 4.

Contamination with benz(a)pyrene varied within 0.003-0.01 µg/kg range for processed cocoa products, milk products, and coffee (hereinafter meant as a ready drink) and within $0.009-0.0013 \mu g/kg$ range for smoked cheese. Discrepancies between lower and upper limits against the average level of contamination with benz(a)pyrene were statistically significant for processed cocoa products (U = 584, Z = -2.94, p < 0.05), milk products (U = 58, Z = -5.79, p < 0.05), and coffee (U = 29.5, Z = -6.21, p < 0.05). We didn't reveal any discrepancies between the examined level in other food products and it may be due to small size of examined samplings and a great number of values being higher than the quantitative determination limit chosen for the procedure applied in this work.

Average contamination with benz(a)pyrene taken as per median amounted to 0.42 μ g/kg for tea (hereinafter meant as a ready drink); 0.20 μ g/kg for vegetable oils and fats; 0.05 μ g/kg for smoked fish products and bread and bakery; 0.02 μ g/kg for smoked meat products; 0.01 μ g/kg for smoked cheese; 0.0065 μ g/kg for milk products, coffee, and processed cocoa products.

The highest (95P) benz(a)pyrene quantity reached 4.84 μ g/kg in tea and 1.29 μ g/kg in vegetable oils and fats.

Table 5 contains data on PAH mixture levels in various food products taking into account their carcinogenic equivalents.

Contamination with PAH mixture determined with using carcinogenic equivalents amounted to 0.03–0.04 µg/kg, 0.07–0.08 µg/kg, 0.05–0.06 µg/kg, and 0.01–0.03 µg/kg for processed cocoa products and milk products, smoked meat products, smoked cheese, and coffee accordingly. Discrepancies between upper and lower limits against average level of contamination with PAH mixture taking into account its carcinogenic equivalents were detected for processed cocoa products (U=636, Z=-2.49, p<0.05), milk products (U=311, Z=-2.05, p<0.05) and coffee (U=270, Z=-2.65, p<0.05).

Average levels of contamination with PAH mixture basing on its carcinogenic equivalents as per median amounted to 0.55 μ g/kg for tea; 0.36 μ g/kg for vegetable oils and fats; 0.14 μ g/kg for bread and bakery; 0.10 μ g/kg for smoked fish products; 0.08 μ g/kg for smoked meat products; 0.06 μ g/kg for smoked cheese; 0.04 μ g/kg for milk products; 0.03 μ g/kg for processed co-coa products; 0.02 μ g/kg for coffee.

Table 4

		Me (25 % ÷75 %)				95P			
Food product	Ν	Lower limit	Average level	Upper limit	Lower limit	Average level	Upper limit		
Vegetable oils and fats	45		0.20 (0.12 ÷0.60)				1.29		
Processed cacao prod- ucts	43	0.003* (0.003 ÷ 0.03)	$\begin{array}{c} 0.0065 \\ (0.0065 \div 0.03) \end{array}$	0.01* (0.01 ÷ 0.03)		0.28			
Fish products (smoked)	30		0.05 (0.02÷0.21)			0.58			
Meat products (smoked)	30	0.02 (0.003 ÷ 0.06)	$0.02 \\ (0.0065 \div 0.06)$	0.02 (0.01 ÷ 0.06)	0.99				
Cheese (smoked)	10	$\begin{array}{c} 0.009 \\ (0.003 \div 0.05) \end{array}$	$\begin{array}{c} 0.011 \\ (0.0065 \div 0.05) \end{array}$	$0.013 \\ (0.01 \div 0.05)$	0.61				
Bread and bakery	30	0.05 (0.003 ÷ 0.20)	$0.05 \\ (0.0065 \div 0.20)$	0.05 (0.01 ÷ 0.20)	0.35				
Milk products	30	0.003* (0.003 ÷ 0.03)	$0.0065 \\ (0.0065 \div 0.0065)$	0.01* (0.01 ÷ 0.01)	0.04				
Tea (ready drink)	30	0.42 (0.08 ÷ 1.64)				4.84			
Coffee (ready drink)	30	0.003* (0.003 ÷ 0.003)	$\begin{array}{c} 0.0065 \\ (0.0065 \div 0.0065) \end{array}$	0.01* (0.01 ÷ 0.01)	0.003	0.0065	0.01		

Benz(a)pyrene contents in certain food products (µg/kg)

N o t e : * means validity of discrepancy between lower limit, average level, and upper limit (p < 0.05).

	N	95P						
Food products	Lower limit	A viana da lavial	Upper limit	Lower	Average	Upper		
	Lower mint	Average level	Opper minit	limit	level	limit		
Vegetable oils and fats	0	.36 (0.22 ÷0.73)			1.57			
Dupped apped whether	0.03*	0.03	0.04*	0.36	0.3	0		
Processed cocoa products	$(0.02 \div 0.05)$	$(0.02 \div 0.06)$	$(0.03 \div 0.06)$	0.50	0.38			
Fish maduate (smalled)	0.10	0.10		0.97	0.00			
Fish products (smoked)	(0.04÷0.31)	$(0.05 \div$	0.31)	0.87	0.88			
Maat was bests (see also d)	0.07	0.0	1.68					
Meat products (smoked)	$(0.04 \div 0.15)$	$(0.05 \div$						
Chasse (amaliad)	0.05	0.06		0.87	0.88			
Cheese (smoked)	$(0.03 \div 0.07)$	(0.03 ÷	0.87					
Bread and bakery	0	.14 (0.03 ÷ 0.28)		0.64				
Milk products	0.03*	0.04	0.04*	0.09		0.10		
	$(0.01 \div 0.06)$	$(0.01 \div 0.07)$	$(0.02 \div 0.07)$					
Tea (ready drink)	0	.55 (0.11 ÷ 2.14)	6.06					
Coffee (ready drink)	0.01*	0.02	0.03*	0.48		0.49		
	$(0.01 \div 0.03)$	(0.01 ÷ 0.04)	$(0.02 \div 0.04)$					

PAH mixture levels in certain food products basing on their carcinogenic equivalents ($\mu g/kg$)

N o t e : * means validity of discrepancy between lower limit, average level, and upper limit (p < 0.05).

Highest (95P) contamination levels reached 6.06 μ g/kg in tea and 1.57 μ g/kg in vegetable oils and fats.

Table 6 contains data on PAHs mixture contents in different food products using mutagenic equivalents.

Contamination with PAHs mixture taken as per its mutagenic equivalents varied within 0.03–0.05 µg/kg for processed cocoa products and milk products; $0.11-0.12 \mu g/kg$ for smoked fish products; 0.05-0.07 µg/kg for smoked cheese; 0.02–0.04 µg/kg for coffee. Discrepancies between upper and lower limits against average level of contamination with PAHs mixture taking into account its mutagenic equivalents were detected for processed cocoa products (U = 418, Z = -4.37, p < 0.05), milk products (U = 278, Z = -2.54, p < 0.05), and coffee (U = 135, Z = -4.65, p < 0.05). Average levels of contamination with PAH mixture basing on its mutagenic equivalents as per median amounted to 0.81 μ g/kg for tea; 0.44 μ g/kg for vegetable oils and fats; 0.14 µg/kg for bread and bakery; 0.12 µg/kg for smoked fish products; 0.11 µg/kg for smoked meat products; 0.07 µg/kg for smoked cheese; 0.03 µg/kg for smoked coffee; 0.04 µg/kg for milk products and processed cocoa products.

Maximum (95P) PAHs mixture contents recalculated as per its mutagenic equivalent reached 6.63 µg/kg in tea and 1.85 µg/kg in smoked meat products.

We didn't detect violated maximum permissible concentrations of benz(a)pyrene and mixture of benz(a)anthracene, benz(b)fluoranthene, chrysene, and benz(a)pyrene fixed in hygienic standards existing in Belarus, the Eurasian Economic Union, and in the European Union in all the examined food products samples^{7, 8, 9}.

⁷Commission Regulation (EU) No 835/2011 of 19 August 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in food stuffs. EuroLex. Available at: http://eur-lex.europa.eu/legalcontent/EN/TXT/PDF/?uri=CELEX:32002R0178&gid= %201429 076106145 (29.03.2019).

⁸ The hygienic standard «Parameters of food products and food raw materials safety for human». Approved by the Order of the Belarus Public Healthcare Ministry on June 21, 2013 No. 52. Minsk, The Republican Center for Hygiene, Epidemiology, and Public health Publ., 2013. Available at: http://minzdrav.gov.by/ru/dlya-spetsialistov/normativno-pravovaya-baza/tekhnicheskie-normativnyepravovye-akty/teksty-tekhnicheskikh-normativnykh-aktov/pishchevye-produkty-i-pishchevye-dobavki.php (29.03.2019). ⁹ TR CU 021/2011. On food safety from 15.12.2011. Minsk, BelGISS Publ., 2015, 160 p. (in Russian).

]	Me (25 % ÷75 %	95P				
Food products	Lower limit	Average level	Upper limit	Lower limit	Average level	Upper limit	
Vegetable oils and fats		0.44 (0.27 ÷0.78))	1.62			
Processed cocoa	0.03*	0.04	0.05*		0.45		
products	$(0.02 \div 0.05)$	$(0.03 \div 0.06)$	$(0.04 \div 0.07)$				
Fish products	0.11	0.	0.80 0.00				
(smoked)	(0.05÷0.34)	(0.06-	÷0.34)	0.89 0.90			
Meat products	0.	11	0.11	1.95			
(smoked)	(0.07 -	÷ 0.17)	$(0.08 \div 0.17)$	1.85			
Cheese (smoked)	0.05	0.07	0.07	0.86		0,87	
Cheese (shloked)	$(0.03 \div 0.08)$	$(0.04 \div 0.08)$	$(0.05 \div 0.09)$				
Bread and bakery	0.12	0.14	0.15	0.89			
	$(0.03 \div 0.28)$	$(0.05 \div 0.29)$	$(0.06 \div 0.30)$			_	
Milk products	0.03*	0.04	0.05*	0.21		0,22	
	$(0.01 \div 0.09)$	$(0.02 \div 0.10)$	$(0.04 \div 0.11)$				
Tea (ready drink)	(6.63				
Coffee (ready drink)	0.02*	0.03	0.04*	1.13			
	$(0.01 \div 0.03)$	$(0.02 \div 0.04)$	$(0.04 \div 0.06)$				

PAHs mixture levels in certain food products basing on their mutagenic equivalents (µg/kg)

N o t e : * means validity of discrepancy between lower limit, average level, and upper limit (p < 0.05).

Our results revealed that the highest contamination with benz(a)pyrene and PAHs mixture taking into account carcinogenic and mutagenic equivalents occurred in vegetable oils and fats and tea. It can be due to a sequence of technological processes that make for occurrence of the examined substances and to physical and chemical properties of the said food products. Greater contamination with the examined substances occurring in vegetable oils and fats can be due to high temperatures being applied when oil culture seeds are being dried and then roasted [1-3, 8]. Ether oils that are contained in tea can act as co-solvents for certain lipophilic compounds, PAHs included, and it results in greater contents of the examined substances in tea drinks [17, 18].

Lower benz(a)pyrene and PAH contents recalculated as per its carcinogenic and mutagenic equivalents in smoked meat, fish, and cheese are due to technological peculiarities typical for their production, namely, smoked flavoring agents or up-to-date equipment applied in production processes allowing to control conditions for smoked products manufacturing. Bread and bakery, processed cocoa goods, and coffee get contaminated due to such technological operations as raw materials drying and roasting. These processes usually involve high temperatures for roasting [1-2, 8, 16]. Besides, cocoa and coffee beans get contaminated with PAH during their storage and transportation in jute or sisal bags treated with textile oil [1-2, 8, 16].

Milk products get contaminated due to PAH migration along food chains and surface contamination of grain cultures.

We didn't detect the examined substances in instant coffee; it is due to PAHs being hydrophobic and peculiarities of applied technological processes as ground coffee beans are treated with hot water under 15 atm (steam treatment), and then extracted soluble substances are dried with hot air. Therefore, the examined compounds that occur in ground coffee do not penetrate a ready product during extraction.

Conclusion. Validation of the procedure for quantitative PAH determination in food products allowed establishing lower detection limit for benz(a)anthracene and benz(a)pyrene, namely, down to 0.003 μ g/kg; and down to $0.03 \ \mu g/kg$ for benz(b)fluoranthene and chrysene. Average contamination with benz(a)pyrene as per median varied from 0.0065 µg/kg in processed cocoa products and milk products to 0.42 μ g/kg in tea; from 0.03 μ g/kg to 0.55 μ g/kg basing on carcinogenic equivalents; from $0.04 \,\mu g/kg$ to $0.81 \,\mu g/kg$ basing on mutagenic equivalents. Contamination with benz(a)pyrene that was close to its maximum levels (95P) taking into account carcinogenic and mutagenic equivalents amounted to 4.84 μ g/kg, 6.06 μ g/kg, and 6.63 μ g/kg in tea. We didn't detect maximum permissible concentrations of benz(a)pyrene and a mixture of benz(a)anthracene, benz(b)fluoranthene, and chrysene in any examined food product samples. Our research results revealed that the highest contamination with the examined substances was

typical for food products manufactured via smoking, roasting, and drying as well as for products that contain fats and ether oils. Integral assessment of food products contamination with PAHs allowed determining contents of the examined substances as a mixture taking into account their individual contributions into overall contamination and degree of their carcinogenic and mutagenic activity.

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