Dietary intake of arsenic in the Slovak Republic

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Summary

The aim of this study was to evaluate arsenic contents in food commodities and its intake via the dietary pathway in the Slovak Republic. The estimated daily doses of arsenic ingested via food and beverages were based on arsenic findings in analysed commodities and the respective data on consumption, and were calculated for an average inhabitant and for children of various age groups. For the survey, analytical findings in the total of 68 513 samples of foods and beverages, collected at agricultural holdings, food industry enterprises, the retail network and households were utilized. Two types of consumption models were used: a model based on Household Budget Survey (HBS) and models based on the rational diet. The estimated daily doses of arsenic for an average inhabitant related to body weight (bw) during the evaluated period (1994–2005) ranged between 0.25 μg.kg^{-1} bw and 0.60 μg.kg^{-1} bw (HBS-based model, mean findings). As regards children, the most burdened were pre-school children (ages 4–6 years), with daily doses ranging from 0.57 μg.kg^{-1} bw to 1.16 μg.kg^{-1} bw. According to data from 2005, the major contribution to dietary intake of arsenic was from fish and fish products. Also, these commodities had the highest findings of arsenic.

Keywords

arsenic; dietary intake; risk assessment; food; HBS model; rational diet model

Arsenic occurs in the general environment both released from its natural sources or as a result of human activity [1, 2]. In the Slovak Republic, the main sources of arsenic emissions primarily include the energy sector (power station Nováky SE in the town of Zemianske Kostoľany) and metallurgy (e.g., factory Kovohuty in the town of Krompachy). The coal-burning power station Nováky in central Slovakia annually emitted up to 200 tones of arsenic between 1953 and 1989. Since then, pollution-control measures have reduced arsenic emissions to less than 2 t a year [3].

Arsenic occurs in both inorganic and organic forms, with inorganic forms being more toxic [4, 5]. In bodies of living organisms, inorganic arsenic gets bound with carbon and hydrogen to its organic forms [1]. Bioaccumulation of organic arsenic compounds, after their biogenesis, occurs mainly in aquatic organisms, and its bioconcentration in freshwater invertebrates and fish is lower than in marine organisms [6]. Limited data indicate that less toxic, organic forms of arsenic generally prevail in food commodities, and approximately only 25% of the total arsenic in food is inorganic [6-9]. However, this depends highly on the type of food,

e.g. various cereals have higher levels of inorganic arsenic, while in fish and shellfish, the portion of inorganic arsenic is low (< 1%) [6].

Non-occupational human exposure to arsenic is primarily through the dietary pathway, namely ingestion. Daily intake of arsenic from food and beverages is generally between 20 and 300 μg [6]. Also, inhalation of contaminated air and inhalation of cigarette smoke represent a significant source of exposure. Pulmonary exposure may be up to 10 μg per day for smokers and about 1 μg per day for non-smokers, but the pulmonary exposure to arsenic may be even higher in polluted areas [1, 6, 10].

In the human body, arsenic is detoxified in liver and is eliminated mainly by urine within several days [11-14]. Most mammals are able to methylate inorganic arsenic to dimethylarsenic acid (DMA). However, this compound has organ-specific toxicity and represents a promoter of tumours in a variety of organs, as confirmed by animal studies [10, 11]. Also, inorganic arsenic has been classified by IARC (The International Agency for Research on Cancer) and US EPA (United States Environmental Protection Agency) among the group of

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substances with a carcinogenic potential for humans [15, 16]. Arsenic compounds may also cause mutagenic and teratogenic effects [4]. Chronic toxicity due to arsenic may lead to skin lesions, nerve damage, skin cancer and diseases of the blood vessels. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) has evaluated arsenic and established a provisional tolerable weekly intake (PTWI) of 15 μg.kg^{-1} bw (bw – body weight) to inorganic arsenic [17]. However, most surveys including this one have measured total arsenic, since it is difficult to distinguish analytically between the different forms of arsenic in food. Since the exposure limit value for total arsenic has not been stated, the risk of total arsenic intake cannot be accurately evaluated and, in accordance with the PTWI value for more toxic inorganic form, it would be overestimated. For this reason, our survey is focused only on the assessment of total arsenic contents in analysed food commodities as well as on evaluation of daily intake of total arsenic via dietary pathway within 1994–2005. This approach was already applied in studies of the dietary exposure to various contaminants (e.g. lead, cadmium, mercury, polychlorinated biphenyls and nitrates) which were accomplished in the Slovak Republic [18-23] and makes possible a more complete view of the exposure of population to food contaminants.

MATERIALS AND METHODS

For the evaluation of arsenic contents in food commodities and assessment of daily intake of arsenic via dietary pathway, we utilized the extensive Database on Occurrence of Contaminants in Food (Centre for Evaluation of Contaminant Occurrence, Bratislava, Slovakia) as well as professional experience acquired from a variety of national projects (Partial Informational System on Contaminants; Partial Monitoring System "Food and Feed Contamination"; projects of the National Programme on Health Support). Arsenic was chosen as one of many parameters monitored, due to the results of long-term toxicological studies, references on health risk, international recommendations and course of the food surveillance system in the Slovak Republic. Also, the list of priority contaminants in foods set up by GEMS FOOD WHO was used [24].

During the period of 1994–2005, a total of 68 513 samples of basic food raw materials, foods and beverages were analysed for arsenic. The samples were collected in the territory of the Slovak Republic at agricultural holdings, food industry enterprises, the retail network as well as households. The following institutions participated in the sampling and analyses: State Veterinary and Food Administration of the Slovak Republic (Regional Veterinary and Food Administrations, State Veterinary and Food Institutes); Food Research Institute, Bratislava; Dairy Research Institute, Žilina; Administrations of Public Health; and Slovak Medical University, Bratislava. Since 1994, laboratories of the agriculture sector have been involved in the national system of AQA (Analytical Quality Assurance) for food, and since 2000, they have regularly taken part in the international competency tests focusing exclusively on food analyses, in the FAPAS (Food Analyses Performance Assessment Scheme) programmes, and in the GEMS/Food (Global Environmental Monitoring System).

The paper encloses results of assessment of arsenic contents in selected food commodities in the Slovak Republic as well as annual time series of estimated daily doses of arsenic ingested via dietary pathway (μg per day). For the assessment of exposure doses of arsenic, data on chemical residues in each analysed food type were combined with data on the consumption of respective food commodities to produce an estimate of total daily contaminant intake via the dietary pathway. The exposure doses of dietary arsenic were calculated for an average consumer in the Slovak Republic as well as for selected age categories of children (preschool children, younger school-age children and senior school-age children). The values of daily doses of arsenic were also expressed as related to body weight of the consumer (μg.kg^{-1} bw). A value of 70 kg was used as the body weight of an average consumer, and for children of various age groups, average body weights of 18 kg, 33.5 kg and 47 kg were used. Also, a part of the paper was focused on the contribution of selected commodity groups to total daily intake of arsenic from food and beverages.

Commodities analysed in the survey were divided into 93 groups (Tab. 1).

For data on arsenic contents, the analytical data from the food inspection system (State Veterinary and Food Administration of the Slovak Republic) as well as from the monitoring programme were utilized. Mean values of all arsenic findings obtained during each year of the observation period were used for the assessment. There was a considerable amount of samples in which arsenic was not detected. For such cases, a value of 50% of the limit of quantification was used.

For the consumption data, the following models of consumption were used (Tab. 1):

Tab. 1. Dietary consumption models used for arsenic intake assessment
(amount of consumed food commodities is expressed in g per person per day).

Commodity	HBS–based model (average consumer)	Consumed food commodities [g per person per day]			
		Models based on rational diet			
		Adult consumer	Children (aged 4–6)	Children (aged 7–11)	Children (aged 12–15)
Beef and veal	6.05	42.61	20.45	24.86	28.11
Pork	27.98	16.08	27.15	30.70	35.03
Mutton	0.04	–	–	–	–
Other meat	0.67	–	–	–	–
Offal and bones	1.00	–	–	–	–
Chicken	24.50	22.02	12.95	14.53	16.54
Hen meat	1.59	–	1.55	1.64	1.63
Goose meat	0.34	–	–	–	–
Duck meat	1.03	–	–	–	–
Turkey meat	1.90	–	–	–	–
Poultry offal	3.05	2.57	–	–	–
Poultry products	3.02	–	–	–	–
Canned meat	4.13	–	4.29	6.93	8.50
Meat preserves	0.22	–	11.34	11.95	13.30
Canned poultry	0.23	–	–	–	–
Durable sausages	4.71	–	–	–	–
Soft sausages	11.07	9.80	0.61	1.08	1.39
Small meat products	13.13	–	7.14	8.57	11.43
Boiled meat products	4.48	–	–	–	–
Special meat products	0.98	–	–	–	–
Smoked meat	3.48	7.46	1.07	2.14	2.86
Other meat products	2.05	–	–	–	–
Marine fish	3.07	23.55	–	–	–
Freshwater fish	0.67	–	–	–	–
Smoked fish products	0.26	–	–	–	–
Marinated fish products	1.08	–	–	–	–
Canned fish products	3.36	–	0.17	0.26	0.35
Special fish products	0.97	14.15	–	–	–
Milk	184.60	221.69	81.29	86.42	90.30
Hard cheese	4.66	13.43	5.58	7.08	8.19
Soft cheese	0.21	–	3.57	7.14	7.14
Mould cheese	1.40	–	1.13	1.66	2.12
Processed cheese	4.66	5.43	–	–	–
Other cheese	1.36	–	–	1.31	1.58
Fermented milk products	24.79	50.00	51.56	55.18	60.89
Ice creams	6.17	–	–	–	–
Cottage cheese	6.17	31.70	5.43	7.02	8.69
Cream	6.67	1.27	1.87	2.27	2.90
Canned milk products	1.37	–	–	–	–
Savouryed milk products	0.73	17.14	0.07	0.11	0.11
Powdered milk products	0.44	–	–	–	–
Baby foods	0.62	–	–	–	–
Eggs	19.78	44.25	6.22	7.33	9.93
Egg products	1.40	–	–	–	–
Edible vegetable oils	22.36	18.74	7.82	10.85	12.76
Margarines	13.37	1.43	0.62	0.83	0.93
Butter	6.44	23.70	12.18	14.92	17.32
Lard	1.78	1.49	3.40	4.73	5.72
Bread	122.80	107.14	84.15	145.71	212.24
Rolls	33.15	45.00	21.56	22.46	22.65
Fine pastries	8.41	21.57	37.85	66.56	74.02
Vegetable products	4.43	43.14	9.32	12.63	15.33

Tab. 1. *continued*

Commodity	Consumed food commodities [g per person per day]				
	HBS–based model (average consumer)	Models based on rational diet			
		Adult consumer	Children (aged 4–6)	Children (aged 7–11)	Children (aged 12–15)
Cabbage	14.35	15.12	9.37	10.51	12.41
Pulses	2.36	2.86	12.21	14.96	16.94
Fruiting vegetables	35.84	131.85	25.89	35.69	40.25
Leafy vegetables	1.98	5.55	0.33	0.36	0.36
Root vegetables	12.08	50.39	30.44	35.17	37.82
Potatoes	72.10	211.59	48.13	70.76	92.50
Mushrooms	0.52	–	–	–	–
Legumes	0.21	–	–	–	–
Fruit products	2.48	154.60	15.58	20.52	20.95
Grapes	2.66	47.31	21.43	21.43	21.43
Pomaceous fruits	39.55	43.02	33.53	33.84	33.84
Stone fruits	9.95	–	12.22	13.67	14.39
Citrus fruits	16.84	3.97	9.69	9.81	9.81
Bananas	10.74	32.23	17.14	21.43	21.43
Tropical fruits	1.87	–	–	–	–
Berries	5.91	–	–	–	–
Nuts	1.81	–	0.27	0.34	0.34
Sugar	39.51	31.43	8.68	11.60	12.22
Chocolates	7.12	–	3.57	7.14	7.14
Candies	6.80	11.43	–	–	–
Cocoa powder	1.64	6.19	1.10	1.20	1.98
Waffles	5.42	–	16.79	22.14	22.14
Cereal products	74.27	–	56.37	78.85	91.37
Condiments	0.76	1.57	0.07	0.13	0.15
Mustard and other ingredients	3.91	0.25	0.62	0.68	1.15
Delicatessen	2.31	–	–	–	–
Jams and marmalades	4.75	–	1.23	1.71	2.07
Preservatives	5.79	2.18	0.44	0.66	0.81
Salt	4.94	8.05	3.15	3.94	4.80
Other foodstuffs	4.80	3.25	0.72	0.87	1.01
Coffee and coffee substitutes	4.30	0.77	0.19	0.25	0.25
Tea	0.80	5.34	0.19	0.24	0.24
Soft drinks	238.49	121.43	10.71	10.71	10.71
Beer	47.07	–	–	–	–
Spirits	7.48	–	–	–	–
Wine	16.80	14.29	–	–	–
Syrups	12.44	6.88	2.84	3.58	3.65
Prepared dumplings	3.32	–	–	–	–
Powdered soup	1.86	–	–	–	–
Prepared meals	1.15	–	–	–	–
Dish portion	0.36	–	–	–	–
Total amount	1330.10	1696.91	763.20	989.04	1154.11

Consumption model based on household budget survey

Data on consumption of an average inhabitant of the Slovak Republic are based on household budget survey (HBS) data from the Statistical Office of the Slovak Republic [25]. HBS assumes per-capita food consumption, based on expenditure and income of the selected households. The selected households were household budget correspondents for two consequent months. They provided information about their expenditure and income in gross values (the first month) and detailed divisions, data of household structure and housing conditions (the second month). Natural incomes, income in kind as well as expenditures for food commodities, non-alcoholic and alcoholic

beverages in restaurants, cafés, bars, confectioneries and canteens have also been included for the assumption of consumed commodities. Within the model, the values of the consumed commodities were expressed in net values, i.e. in terms of edible share of the particular commodity. In general, however, models are only an approximation of the reality which often brings about a certain level of inaccuracy within the assumption. The weakness of this model is the fact that there is no information on the proportion of the food actually consumed by the individual members of the household and on the other part of the purchased food, which was wasted.

**Consumption model
based on rational diet for adult inhabitant**

The model was derived from rational nutrition menu, based on recommended dietary allowances (RDA) for average adult inhabitant of the Slovak Republic through diet modelling. For modelling, data of the Slovak Food Composition Database (VÚP Food Research Institute, Bratislava, Slovakia) as well as the nutritional software Alimenta 4.2 (VÚP Food Research Institute, Bratislava, Slovakia) were utilized.

**Consumption model
based on rational diet for children**

As a separate objective, also dietary intake of arsenic was calculated for children of various ages. As there are no statistical data on the consumption of various age categories available, these values could only be estimated based on modelled consumption for children, referring to recom-

mended dietary allowances (RDA) and serving sizes for 3 age categories of children: pre-school children (aged 4–6), younger school-age children (aged 7–11) and senior school age children (aged 12–15).

RESULTS AND DISCUSSION

Evaluation of arsenic contents in food commodities

Estimated data of arsenic contents (mean annual values of all the findings measured during the period of 1994–2005) in chosen food commodities are shown in Tab. 2 (fish and fish products), Tab. 3 (meat), Tab. 4 (meat products), Tab. 5 (poultry and poultry products), Tab. 6 (vegetables and vegetable products), Tab. 7 (dairy products) and Tab. 8 (bakery products).

Fish and fish products

Reported data on arsenic contents in food commodities indicate that the most contaminated commodities are fish and fish products, with the mean value of all findings in analysed samples from this category during the period of 1994–2005 reaching 277 μg.kg^{-1}, while the mean annual values of arsenic findings in particular groups of fish and fish products (Tab. 2) ranged between 10 μg.kg^{-1} and 1 222 μg.kg^{-1}. The values of arsenic contents reached the highest levels in case of commodities like marine fish, with the mean value of 383 μg.kg^{-1} for all findings in the given period. On the other hand, the lowest amount of arsenic was found in freshwater fish (59 μg.kg^{-1}). The share

Tab. 2. Arsenic findings (mean values) in fish and fish products.

	Arsenic findings (mean values) [μg.kg^{-1}]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Marine fish	95	80	77	113	78	92	484	995	1222	941	800	645
Freshwater fish	113	39	23	28	37	19	38	95	302	71	76	87
Smoked fish products	219	162	98	155	164	29	470	258	759	521	10	10
Marinated fish products	63	71	219	41	46	148	228	125	340	166	166	26
Canned fish products	147	186	138	94	106	70	212	652	849	541	609	1070
Special fish products	75	140	167	125	146	140	707	310	307	656	50	35

Tab. 3. Arsenic findings (mean values) in meat.

	Arsenic findings (mean values) [μg.kg^{-1}]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Beef and veal	19	20	16	14	10	11	15	13	16	11	12	8
Pork	16	18	12	14	9	9	13	11	14	10	13	8
Mutton	21	7	15	6	10	9	20	5	9	8	5	6
Other meat	26	36	19	24	17	9	20	16	15	11	15	8

Tab. 4. Arsenic findings (mean values) in meat products.

	Arsenic findings (mean values) [μg.kg^{-1}]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Offal and bones	44	33	22	30	17	13	24	18	26	14	22	9
Canned meat	43	33	33	25	13	15	18	18	22	24	15	62
Meat preserves	15	18	26	20	17	15	27	16	27	30	13	13
Durable sausages	21	20	29	25	16	16	36	24	30	23	16	16
Soft sausages	17	16	23	21	15	19	32	18	23	24	37	37
Small meat products	18	20	12	12	20	22	33	21	12	18	12	23
Boiled meat products	20	9	33	24	7	21	31	18	26	19	23	23
Special meat products	21	57	23	29	18	17	34	36	28	20	20	20
Smoked meat	17	17	22	30	16	14	32	22	17	18	12	10
Other meat products	48	30	3	15	26	39	39	20	20	12	12	12

Tab. 5. Arsenic findings (mean values) in poultry and poultry products.

	Arsenic findings (mean values) [μg.kg^{-1}]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Chicken	18	21	12	14	11	11	19	15	17	17	12	10
Hen meat	9	13	17	23	17	14	10	10	17	9	9	11
Goose meat	14	18	19	7	7	12	12	10	10	33	20	20
Duck meat	15	12	15	6	9	12	18	6	13	19	20	20
Turkey meat	15	22	9	9	5	10	14	14	26	15	18	12
Poultry offal	80	104	169	43	83	94	19	30	27	22	17	10
Poultry products	18	23	20	18	10	20	33	26	14	17	17	17

Tab. 6. Arsenic findings (mean values) in vegetables and vegetable products.

	Arsenic findings (mean values) [μg.kg^{-1}]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Vegetable products	47	22	54	20	27	10	78	189	78	33	43	43
Cabbage	30	7	14	8	8	8	8	25	15	9	15	5
Pulses	42	21	79	35	57	54	53	80	40	16	45	57
Fruiting vegetables	11	15	10	9	8	7	10	24	24	9	10	5
Leafy vegetables	10	4	10	10	27	14	14	14	1	1	1	1
Root vegetables	21	16	14	11	16	14	25	44	44	26	15	6
Potatoes	38	16	14	6	6	8	12	41	21	14	15	6
Mushrooms	107	107	107	107	107	107	107	100	118	90	80	6
Legumes	10	16	30	14	46	59	59	20	20	20	20	22

Tab. 7. Arsenic findings (mean values) in dairy products.

	Arsenic findings (mean values) [μg.kg^{-1}]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Fermented milk products	17	14	7	15	5	7	11	10	18	10	9	9
Ice creams	9	12	19	20	9	14	18	17	18	18	5	5
Cottage cheese	24	10	6	12	6	13	21	17	26	17	18	6
Cream	27	21	15	15	2	5	18	16	26	13	7	9
Canned milk products	13	37	11	27	5	20	36	26	28	14	30	30
Savouryed milk products	14	42	20	12	9	7	16	15	23	17	19	19
Powdered milk products	35	2	16	16	14	29	54	24	25	32	26	25
Baby foods	25	2	10	8	7	7	18	23	29	14	9	9
Hard cheese	23	44	30	26	18	13	28	16	37	9	11	11
Soft cheese	32	12	14	14	10	15	10	13	13	9	5	5
Mould cheese	6	28	43	43	24	16	19	11	14	7	5	5
Processed cheese	19	23	29	18	15	21	45	16	61	35	10	11
Other cheese	36	41	242	54	24	6	12	24	5	5	13	5

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Tab. 8. Arsenic findings (mean values) in bread and bakery products.

	Arsenic findings (mean values) [μg.kg^{-1}]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Bread	28	13	11	12	22	7	25	32	30	30	30	30
Rolls	26	9	12	30	23	5	21	40	40	40	40	40
Fine pastries	23	160	160	28	4	22	12	10	10	7	7	25

of values of arsenic contents (mean annual values of all the findings) in marine fish obtained in this survey, in comparison to the current limit in the Slovak Republic (Decree of the Ministry of Agriculture of the Slovak Republic and the Ministry of Health of the Slovak Republic No. 07174/2005-SL) for arsenic contents in marine fish (5000 μg.kg^{-1}), ranged between 1.5% and 24.4% [26].

Meat and meat products

Findings of arsenic in this category of commodities did not reach high values (Tab. 3 and Tab. 4). The mean annual values of arsenic contents measured in various meats ranged from 5 μg.kg^{-1} to 36 μg.kg^{-1}, which correspond to shares of 5% to 36% of the current limit value stated for meat and meat products (100 μg.kg^{-1}) [26], and from 7 μg.kg^{-1} to 57 μg.kg^{-1} in various meat products, which correspond to shares of 7% to 57% of the limit value.

Other food commodities

Mean annual values of arsenic contents in samples from the category of poultry and poultry products (Tab. 5) ranged between 5 μg.kg^{-1} and 169 μg.kg^{-1}, with the mean value of all the findings from this category for the period 1994–2005 reaching 20 μg.kg^{-1}. In vegetables and vegetable products (Tab. 6), these values ranged between 1 μg.kg^{-1} and 189 μg.kg^{-1}, with the mean value of all the findings from this category for the given period reaching 22 μg.kg^{-1}, and with mushrooms being the commodity with the highest contents (mean value of 84 μg.kg^{-1}). In the category of dairy products (Tab. 7), these values ranged between 5 μg.kg^{-1} and 242 μg.kg^{-1}. A higher level of arsenic was found in various cheeses. As regards bread and bakery products (Tab. 8), contamination with arsenic was not at a high level. The mean annual values of arsenic contents in this group of products ranged between 5 μg.kg^{-1} and 160 μg.kg^{-1}.

In general, besides commodities like fish and fish products, higher values of arsenic were found in delicatessen (where the mean annual values of arsenic ranged between 15 μg.kg^{-1} and 484 μg.kg^{-1}, with the mean of all findings as high as 225 μg.kg^{-1}), in tea (where the mean annual

values of arsenic ranged between 14 μg.kg^{-1} and 4625 μg.kg^{-1}, with the mean of all findings as high as 153 μg.kg^{-1}), in cocoa powder (where the mean annual values of arsenic ranged between 10 μg.kg^{-1} and 281 μg.kg^{-1}, with the mean of all findings as high as 123 μg.kg^{-1}), and salt (where these values ranged between 1 μg.kg^{-1} and 861 μg.kg^{-1}, with the mean of all findings as high as 172 μg.kg^{-1}). However, the exposure doses of arsenic ingested via food and beverages are influenced by the consumption of the particular commodity, and therefore higher contents found in a particular commodity do not mean also a high exposure dose of the contaminant. For example, in case of salt, the intake of arsenic was less significant because of the less extensive consumption. According to results from 2005 (Fig. 1), exposure doses of arsenic suggested that the sources accounting for the major portion of arsenic intake calculated for HBS-based model of consumption, included fish and fish products (25.3%) followed by bread and bakery products which contributed by 23.4% to the overall daily intake of 0.32 μg.kg^{-1} bw, even though the respective mean value of arsenic in bread and bakery products was not high (25 μg.kg^{-1}).

According to data acquired from the assessment of daily doses from the model based on rational diet, intake from fish and fish products, besides the higher arsenic contents in these commodities, was also magnified by their higher modelled consumption. Arsenic intake from vegetables and vegetable products, in particular potatoes, was also higher due to the same reason. Based on the rational diet model of consumption, fish and fish products contributed most (43.9%) to the overall daily intake of 0.51 μg.kg^{-1} bw. Bread and bakery products contributed with 15.5% to the daily intake of arsenic and vegetables and vegetable products with 12.4%. Although arsenic contents as well as its intake from commodities such as fish and fish products were at a high level, a major portion of arsenic in such commodities is in its less toxic, organic form.

Dietary exposure to arsenic in the Slovak Republic

The ranges of daily doses of arsenic, calculated from mean annual findings for the period of obser-
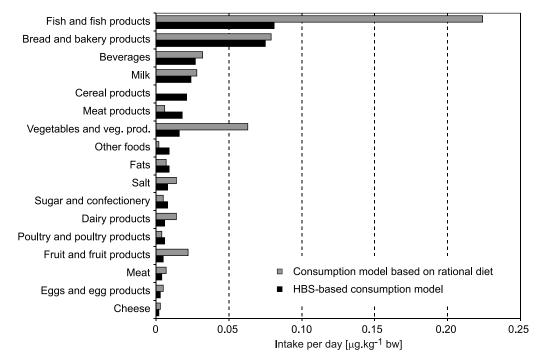
Fig. 1. Arsenic intake from individual commodities, calculated for consumption model based on HBS data and consumption model based on rational diet for an average inhabitant of the Slovak Republic in 2005.

vation (1994–2005), using models of consumption for the average consumer in the Slovak Republic as well as for selected age groups of children, are presented in Tab. 9.

Trends of estimated daily doses of arsenic for HBS-based consumption model

Daily doses of arsenic calculated from HBS-based consumption data and mean annual findings suggested a decreasing tendency of the exposure doses during the first period of observation (1994–1998), ranging between 17.49 μg and 41.66 μg (corresponding to 0.25 μg.kg^{-1} and

0.60 μg.kg^{-1} bw); the maximum value calculated in the first year of the observation period at the same time represented the highest value for the entire period of observation (Tab. 9, Fig. 2). Increasing tendencies of the daily arsenic intake values were recorded from 1999 until 2001, reaching levels as high as 38.63 μg, with a subsequent reduction of the daily intake values after 2002, down to 22.41 μg in 2005.

Comparison shows that the daily arsenic doses based on the value of the 95th percentile, which could be used to estimate the highest prospective intake, reached markedly higher

Tab. 9. Daily doses [μg] of arsenic from food and beverages within 1994-2005 for an average inhabitant of the Slovak Republic (mean findings; consumption model based on HBS data and consumption model based on rational diet) as well as for the selected age groups of children (consumption model based on rational diet for 3 age categories of children; mean findings).

	Daily doses of arsenic from food and beverages [μg]											
	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Average inhabitant (HBS - based MODEL)	41.66	27.71	21.28	20.74	17.49	18.11	30.74	38.63	35.88	28.18	24.58	22.41
Average inhabitant (Rational diet model)	81.1	35.6	29.58	26.76	22.06	23.15	56.42	76.04	75.37	59.17	43.66	35.5
Children in age of 4-6	20.80	17.71	16.57	12.06	10.31	11.36	17.13	20.52	20.69	13.81	13.37	11.77
Children in age of 7 -11	27.56	25.60	24.60	16.05	13.92	15.06	22.46	27.33	26.58	18.39	18.19	16.39
Children in age of 12-15	32.82	29.74	28.49	18.82	16.74	17.37	26.73	32.76	31.26	22.14	22.07	19.94

values, ranging between 48.25 μg and 126.93 μg (1.44–3.59 μg.kg^{-1} bw). The highest values were obtained in 1994, the lowest ones in 2005. The value of 95th percentile, however, describes an extreme situation and it is not likely in reality in all the food commodities within the diet.

Trends of estimated daily doses of arsenic for model based on rational diet

Estimated daily doses of arsenic based on data of the rational diet model are higher than those estimated for HBS-based consumption model when considering the mean findings (Tab. 9, Fig. 3). Taking mean annual findings as the basis, the estimated daily doses of arsenic ranged between 22.06 μg (0.32 μg.kg^{-1} bw) in 1998 and 81.10 μg (1.16 μg.kg^{-1} bw) in 1994. Similarly as in the case of HBS-based consumption model, an increase of arsenic doses

was noticed in 2000 to 2002, with a subsequent decreasing tendency. Markedly higher values were obtained when using the 95th percentile as the basis, when during the period of observation, the estimated values of daily arsenic intake ranged between 64.50 μg in 1999 and 213.89 μg in 2001.

The higher values of arsenic doses calculated for model based on rational diet were generally influenced by the higher modelled consumption. In particular, this was evident in respect to commodities such as sea fish, vegetable products, potatoes, fish specialties, and root vegetables (Table 1).

Daily doses of arsenic calculated for selected age groups of children

The values of daily arsenic doses (μg) in respect of children of various age groups calculated for the period of observation are presented in Tab. 9. The
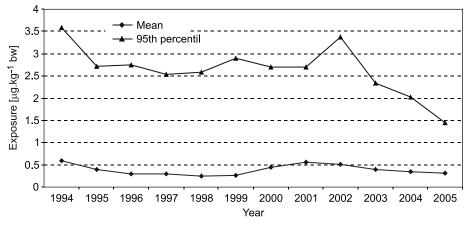

Fig. 2. Time trends of daily exposure doses [μg.kg^{-1} bw] of arsenic from food and beverages within 1994–2005 (average inhabitant of the Slovak Republic; mean findings; 95th percentile; consumption model based on HBS data).
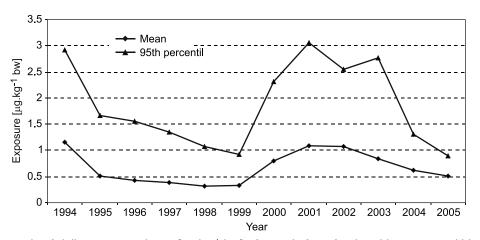

Fig. 3. Time trends of daily exposure doses [μg.kg^{-1} bw] of arsenic from food and beverages within 1994–2005 (average inhabitant of the Slovak Republic; mean findings; 95th percentile; consumption model based on rational diet).
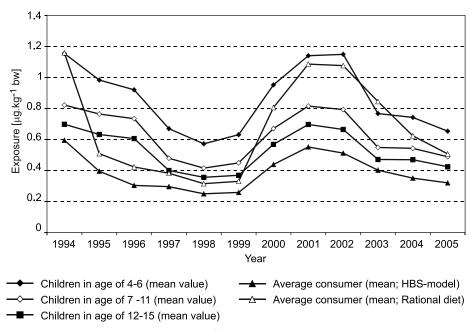
Fig. 4. Time trends of daily exposure doses [μg.kg^{-1} bw] of arsenic from food and beverages within 1994–2005 (age groups of children and average inhabitant of the Slovak Republic; mean findings).

obtained results indicate that the most burdened were pre-school children (ages 4–6 years) for whom the daily doses of arsenic (based on mean values) during the period of observation ranged from 10.31 μg to 20.80 μg, which corresponds to values from 0.57 μg.kg^{-1} bw to 1.16 μg.kg^{-1} bw (Fig. 4). The younger school-age group (ages 7–11) was less burdened, with the daily doses of arsenic ranging from 13.92 μg to 27.56 μg (0.42 μg.kg^{-1} bw to 0.82 μg.kg^{-1} bw). The least burdened group was that of the senior school-age children (ages 12–15), for whom the daily doses of arsenic during the period of observation ranged from 16.74 μg to 32.82 μg (0.36 μg.kg^{-1}bw to 0.70 μg.kg^{-1} bw).

As compared with arsenic doses for the average inhabitant, those calculated for children were higher (Tab. 9). However, it should be underlined that the consumption model based on rational diet, set up for the adult population as well as for the particular groups of children, assigns the highest consumption of food related to body weight (in terms of g.kg^{-1} bw) to the younger demographic groups. Due to this, the intake of arsenic was also higher for those groups. However, there was a lack of consumption data for considerable amount of food commodities in the models of consumption used for the intake assessment, in particular, for fish and fish products, which are commodities with highest arsenic contents. This drawback was evident in case of models used for assessed age categories of children, and therefore, the exposure

doses of ingested arsenic for children might be even higher than those assessed.

Regarding the arsenic contents in food commodities, higher values were found in rice and vegetables, where inorganic arsenic made up 87% of total arsenic in the former and 96% in the latter [27]. Some wines may also contain higher amount of arsenic. Arsenic present in wines probably originates mostly from insecticides used to protect grapes [2]. Higher contents of arsenic were also found in oat and cocoa beans. Mushrooms and poultry meat are also considerable sources of exposure. In some areas, drinking-water can be a significant source of exposure to inorganic arsenic. In general, arsenic contents in food, except for marine animals, is generally below 1000 μg.kg^{-1} and sea fish contain less than 5000 μg.kg^{-1} [11, 28, 29].

Dietary exposure to total arsenic has been estimated also in other countries; however, comparison of data between countries is complicated. One of the reasons is connected with the fact that various countries have rather complex and very different systems and methodologies for the evaluation of the dietary intake of food contaminants. Some of the observations were based on the results of total diet studies, which were based on findings in prepared foods (washed, peeled or cooked) rather than on findings in raw commodities of the market basket survey. Also, in some of the countries, the evaluation was done separately for male and female consumers with the respective data for

their average body weight. In addition, each country has its specific eating habits and therefore, it is quite impossible to compare results of the observation between continental countries and coastal states, where the consumption of fish and other sea food is higher. According to available data, Slovak Republic can be classified among countries with the lowest arsenic exposure values (17.49 μg to 41.66 μg per day). The highest exposure doses were reported for Spain, Japan and New Zealand. Exposure of male adults in New Zealand was estimated to be 150 μg per day [30], a survey carried out in the Basque Country (Spain) in 1990 and 1991 found a mean total arsenic exposure of 286 μg per day [31] and a duplicate diet study of Japanese adults reported an exposure of 182 μg per day [32]. However, in these countries, the high exposure doses originates from high consumption of fish. The portion of fish consumption in the national diet in the Slovak Republic is only 0.9%.

The risk of arsenic intake cannot be accurately evaluated since the exposure limit value for total arsenic has not been stated. In accordance with the PTWI value for inorganic arsenic (15 μg.kg^{-1} bw), the share of the values obtained in our survey would range from 11.7% to 27.8% PTWI (mean values; adult inhabitant; HBS-based model of consumption). However, the PTWI value, mentioned above has been stated for more toxic inorganic form, and seeing that only up to 25% of the total arsenic in food is presented in this form, the risk evaluated in accordance with this value would be overestimated.

CONCLUSION

The exposure of the Slovak Republic population to arsenic within the period of observation did not reach values which might be associated with an increased probability of health risk. Although the trend of exposure to arsenic in the Slovak Republic has been showing a slightly decreasing tendency in recent years, arsenic will remain a focus of the organizations of food surveillance as the results obtained from localities monitored during the years of the observation period represent a basis for the calculations of exposure dose trends as well as for the identification of significant sources of dietary exposure.

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