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PESTICIDES RESIDUE DETERMINATION IN VEGETABLES AND FRUITS COMMONLY USED IN REPUBLIC OF MOLDOVA AND ESTIMATION OF HUMAN INTAKE

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Abstract. The purpose of this research was to assess the residual concentrations of pesticides in fruits and vegetables and to estimate the health risk associated with the consumption of pesticide-contaminated vegetables and fruits. A total number of 5206 samples from twenty one different vegetables and fruits were collected during 2009-2017. Pesticides in concentrations exceeding the maximum residue levels were found in 1194 analysed samples (22.9% of total). Thirteen insecticides, four fungicides, two acaricides and one herbicide were detected in the analysed samples. The estimated daily intake (EDI) has been established between 0.000001 and 0.0002 mg/kg of body weight/day. Calculated values of EDIs are lower than the levels of acceptable daily intake. The calculated hazard indices ranged from 0.000004 up to 0.15 for the analysed pesticides and the highest value of hazard index was calculated for diazinon, being of 0.15. It might be concluded that the long-term consumption of vegetables and fruits could pose a health risk for the population of the Republic of Moldova, since the minimum norms used for risk estimation do not reflect real food consumption pattern in the Republic of Moldova.

Keywords: pesticide residue, fruit, vegetable, estimated daily intake, risk assessment.

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Introduction

Pesticides are chemicals substances that are mainly used worldwide in agriculture or in public health sector in order to protect plants from diseases, pests, or weed and people from vector-borne diseases, such as contacting dengue, malaria, Chagas disease, leishmaniasis and schistosomiasis. The agricultural sector of the Republic of Moldova uses 838 plant protection products based on 211 active substances of different chemical groups, including: compounds of copper and sulphur, carbamates and thiocarbamates, organophosphorus and synthetic pyrethroids, chlorophenoxy compounds, neonicotinoid derivatives, sulphonylureas, strobilurins, and others. The total amount of pesticides used in 2017, in Republic of Moldova, was estimated to 2064 tonnes. Pesticides together with fertilizers have contributed to substantial increases in agricultural crop yields production, causing though many health risks. The numerous adverse health effects in humans that have been associated with chemical pesticides include dermatological, gastrointestinal, respiratory, neurological, carcinogenic, reproductive and endocrine problems [1-13]. Some pesticides that are used in agriculture are classified as carcinogenic, mutagenic or causing hormonal system disorders. The severity of the health problems depends on the quantity and the route in which a person is exposed to potentially toxic pesticides. An increasing number of cases of lethal pesticide poisoning for both adults and children was registered over past years in the Republic of Moldova [14]. On a global level, there are three million cases of acute and severe pesticide poisonings with about 200,000 deaths [15-20]. Many studies were focused on determining indirect exposure of pesticides, including exposure on consumers [21-25]. As pesticides are toxic and deliberately spread in the environment, the production, distribution and use of pesticides requires strict regulation and survey. In this context, regular monitoring of pesticide residues in food is required.

Vegetables and fruits are important components of human diet since they provide essential nutrients that are required for most of the biochemical reactions occurring in the human body. Like other crops, fruits and vegetables are attacked by pests and diseases during production and storage, leading to damages that reduce the quality and the harvest. In order to reduce the loss and maintain the quality of fruits and vegetables harvest, pesticides are used. However, the use of pesticides during production often leads to the presence of pesticide residues after harvest [15]. The presence of pesticide residue in vegetables and fruits has always been a matter of serious concern for the public health of Republic of Moldova. Even in residual quantities pesticides can lead to serious chronic health hazards, especially, when food commodities are consumed fresh [26]. Therefore, the monitoring of pesticides levels can be regarded as a contemporary public health problem that could help guarantee food quality and evaluate the health risks. Pesticide residues in food are usually monitored with reference to *maximum residue levels* (MRLs) and *acceptable daily intakes* (ADIs). The MRL is an index that represents the highest concentration of pesticide residue that is legally accepted in a food commodity after the use of pesticides. National MRLs were established in Sanitary Regulation [27]. The MRLs are always set far below levels considered to be safe for humans. Safety limits are assessed in comparison with ADI. ADI is an estimated amount of a chemical in food that can be ingested daily over a life time without appreciable health risk to the consumer [26].

The purpose of the present study was to assess the residual concentrations of pesticides in fruits and vegetables commonly consumed in Republic of Moldova using gas chromatography/mass spectrometry analysis. Additionally, the human health risks associated with daily intake from agricultural products were evaluated.

Experimental

Sample collection

A total number of 5206 samples from twenty-one different vegetables and fruits (of which 4365 were apple samples, 125 tomato samples, sixty three cucumber samples, twenty two eggplant samples, sixty eight head cabbage samples, twenty seven plum samples, 278 samples of table grapes, *etc*.) from the harvests of 2009–2017 were collected from different agricultural fields of the Republic of Moldova.

Pesticide residues analysis

The pesticide residues analysis was carried out according to Moldovan standard SM EN 15662:2015 [28]. Determination of pesticide residues using gas and liquid chromatography-mass spectrometry analysis was performed following the acetonitrile extraction/partitioning and clean-up by dispersive SPE-QuEChERS-method (quick, easy, cheap, effective, rugged and safe). This method involves liquid-liquid partitioning using acetonitrile and purifying the extract using dispersive solid phase extraction.

The general procedure consists in using an amount of 10 g of homogeneous sample in frozen conditions to for the extraction by acetonitrile. After addition of magnesium sulphate, sodium chloride and citrate buffer (pH 5.0 to 5.5) the mixture is shaken and centrifuged for phase separation. An aliquot of the organic phase (6 mL) is cleaned-up by DSPE using magnesium sulphate (900 mg). The extracts are purified with amino-sorbents (150 mg PSA) afterwards 500 µL of extracted sample with 25 µL of formic acid is subjected to the gas and liquid chromatography-mass spectrometry analysis.

Equipment

The gas and liquid chromatography-mass spectrometry analyses were performed using the Agilent GC/MS 7890/7000 Triple Quadrupole system and Agilent LC/MS 7890 Triple Quadrupole system.

Hazard identification and characterization

The final results of pesticide residues in selected samples were compared with MRLs. The MRLs for all crops and all pesticides can be found in the European Commission MRL online database [29].

The estimated daily intake (EDI) was calculated according to Eq.(1):

$$EDI = C \times F/W \quad (1)$$

where, C is the concentration of pesticide residues in each commodity (mg/kg);
F is the mean daily intake of food per person (kg/day);
W is the mean body weight (in this study, W was considered equal to 60 kg) [23].

Dietary intake of fruits and vegetables per person was estimated considering the minimum norms of the food included in a living-wage food basket (Table 1) [30].

Risk estimation

The calculated EDI was compared with ADI. Hazard indices were calculated by dividing EDI value by ADI value. Toxicological information (ADI) is available at EU pesticide database [29].

Results and discussion

In the Republic of Moldova, the consumers basic diet structure includes about thirty-forty plant based food commodities. The selection of apples, potatoes, tomatoes, cucumbers, eggplants, head cabbage, garlic, onion, plums, table grapes and other listed hereinafter crops was based on their popularity and high consumption rates by Republic of Moldova population. The assessment of pesticide monitoring conducted between 2009-2017 shows that the majority of the pesticides residues were in concentrations below the detection level. In 1194 analysed samples (22.9%) pesticides' residues were found in concentrations exceeding the MRLs. Information regarding contamination with pesticide residues of different vegetables and fruits, number of samples analysed from each commodity, number of samples above MRL, the minimum, the maximum and mean values of pesticides in commodities is presented in Table 2.

Table 1

Estimated food consumption rate per capita according to the food minimum norms included in a living-wage food basket, kg/day [30].

Vegetables	*Consumption*	*Fruits*	*Consumption*
Potato	0.320	Melon	0.017
Onion	0.041	Apple	0.095
Cucumber	0.022	Table grapes	0.028
Tomato	0.050	Pear	0.034
Sweet pepper	0.016	Plum	0.034
Cabbage	0.064	Peach	0.034
Pumpkin	0.015	Apricot	0.034
Eggplant	0.007		
Garlic	0.004		

Table 2

Concentration of pesticide residues detected in vegetables and fruits samples, mg/kg.

Food commodities	*Total no. of samples*	*Detected pesticide*	*No. of samples exceeding MRL*	*Mean*	*Maximum*	*Minimum*	*St. dev.**	*MRL*
Potato	48	Acetamiprid	1	0.0250	0.025	0.025	n/a**	0.01
		Cymoxanil	1	0.0200	0.020	0.020	n/a	0.01
		Dimethoate	7	0.0129	0.020	0.010	0.005	0.01
Onion	26	Dimethoate	11	0.0104	0.020	0.004	0.004	0.01
Cucumber	63	Dimethoate	9	0.0138	0.020	0.010	0.005	0.01
		Phosalone	4	0.0305	0.080	0.002	0.034	0.01
		Propargite	5	0.0820	0.100	0.010	0.040	0.01
Tomato	125	Dimethoate	21	0.0201	0.100	0.001	0.021	0.01
		Phosalone	11	0.0149	0.080	0.002	0.023	0.01
		Methamidophos	20	0.0121	0.025	0.001	0.007	0.01
		Methomyl	1	0.0500	0.050	0.050	n/a	0.01
		Propargite	12	0.0370	0.100	0.004	0.039	0.01
		Pirimiphos-methyl	2	0.0600	0.100	0.020	0.057	0.01
Sweet pepper	75	Dimethoate	19	0.0146	0.020	0.008	0.006	0.01
		Phosalone	6	0.0150	0.080	0.002	0.032	0.01
		Nicosulphuron	1	0.0300	0.030	0.030	n/a	0.01
		Propargite	14	0.0257	0.100	0.010	0.029	0.01
		Procymidone	1	0.0200	0.020	0.020	n/a	0.01
Pepper	1	Dimethoate	1	0.0200	0.020	0.020	n/a	0.01
Cabbage	68	Dimethoate	19	0.0172	0.030	0.010	0.008	0.01
		Malathion	1	0.0400	0.040	0.040	n/a	0.02
		Propargite	2	0.0175	0.025	0.010	0.011	0.01
		Thiamethoxam	2	0.0750	0.100	0.050	0.035	0.02
Cauliflower	10	Dimethoate	2	0.0250	0.040	0.010	0.021	0.02
		Methamidophos	3	0.0107	0.020	0.004	0.008	0.01
		Propargite	2	0.0550	0.100	0.010	0.064	0.01
Pumpkin	16	Phosalone	3	0.0307	0.080	0.002	0.043	0.01
Eggplant	22	Phosalone	4	0.0170	0.060	0.002	0.029	0.01
		Propargite	4	0.0363	0.100	0.010	0.043	0.01
Garlic	17	Dimethoate	7	0.0171	0.020	0.010	0.005	0.01

Continuation of Table 2

Food commodities	*Total no. of samples*	*Detected pesticide*	*No. of samples exceeding MRL*	*Mean*	*Maximum*	*Minimum*	*St. dev.* *	*MRL*
Lettuce leaves	10	Clofentezine	1	0.0400	0.040	0.040	n/a	0.02
		Dimethoate	3	0.0150	0.020	0.010	0.007	0.01
		Propargite	1	0.0200	0.020	0.020	n/a	0.01
Asparagus	5	Propargite	1	0.0250	0.025	0.025	n/a	0.01
Melon	8	Dimethoate	2	0.0200	0.020	0.020	0.000	0.01
		Propargite	1	0.0250	0.025	0.025	n/a	0.01
Watermelon	16	Dimethoate	4	0.0167	0.020	0.010	0.006	0.01
		Phosalone	4	0.0155	0.020	0.002	0.009	0.01
		Methamidophos	3	0.0200	0.020	0.020	n/a	0.01
Apple	4365	Diazinon	60	0.0167	0.050	0.002	0.018	0.01
		Dimethoate	678	0.0207	1.100	0.001	0.074	0.01
		Dimethomorph	1	0.0200	0.020	0.020	n/a	0.01
		Fenitrothion	60	0.0145	0.080	0.002	0.024	0.01
		Malathion	7	0.1186	0.640	0.004	0.231	0.02
		Nicosulphuron	15	0.0583	0.670	0.004	0.170	0.01
		Propargite	47	0.0548	0.180	0.004	0.037	0.01
		Pyridaben	27	0.0781	0.150	0.020	0.040	0.05
Pear	24	Dimethoate	7	0.0126	0.020	0.008	0.005	0.01
		Phosalone	4	0.0215	0.080	0.002	0.039	0.01
		Propargite	2	0.0250	0.025	0.025	0.000	0.01
Table grapes	278	Clofentezine	2	0.2000	0.200	0.200	0.000	0.02
		Dimethoate	14	0.0150	0.050	0.002	0.011	0.01
		Phosalone	6	0.0150	0.080	0.002	0.032	0.01
		Nicosulphuron	12	0.0833	0.380	0.030	0.125	0.01
		Propargite	2	0.0250	0.025	0.025	0.000	0.01
		Pirimicarb	6	0.1067	0.140	0.100	0.016	0.01
		Procymidone	5	0.0178	0.040	0.001	0.015	0.01
		Thiacloprid	6	0.0283	0.040	0.010	0.013	0.01
		Thiophanate-methyl	2	0.4000	0.400	0.400	0.000	0.10
Plum	27	Dimethoate	6	0.0147	0.020	0.008	0.006	0.01
		Methamidophos	4	0.0128	0.025	0.002	0.011	0.01
		Propargite	1	0.0250	0.025	0.025	n/a	0.01
Peach	6	Dimethoate	1	0.0250	0.025	0.025	n/a	0.01
Apricot	5	Dimethoate	1	0.0200	0.020	0.020	n/a	0.01
		Methamidophos	1	0.0200	0.020	0.020	n/a	0.01
Pineapple	1	Propargite	1	0.0250	0.025	0.025	n/a	0.01
21	**5206**	**20**	**1194**					

* *St. dev. – standard deviation;*
** *n/a - not applicable.*

The residue concentration values were compared with the MRLs from the European Commission online database [29]. The MRLs are the highest levels of residues expected to be in the food when the pesticide is used according to authorized agricultural practices [26]. All detected concentrations of pesticides, which exceed the MRLs from two to ten times are presented in Table 2. The highest concentration of residues was registered for thiophanate-methyl in two samples of table grapes (0.4 mg/kg). The lowest concentration was recorded for dimethoate in eleven samples of onion (0.0104±0.0040 mg/kg). In these cases, the following measures have been applied: withdrawal of contaminated products from the market; confiscation and destruction of contaminated products; enforcement sampling of the next coming lot; all exceeding MRLs are published at the National Food Safety Agency's website [31].

The most commonly identified pesticide was dimethoate, which was found in 68% of analysed samples of different fruits (such as apples, table grapes, plums, pears) and vegetables (potatoes, onions, cucumbers, tomatoes, sweet peppers, cabbages and other). Twenty residues of different pesticides were registered in 1194 samples of vegetables and fruits. Sweet peppers, tomatoes, apples and table grapes were contaminated with five, six, eight and nine pesticide residues, respectively (Table 2).

Pesticides can be classified by target organism (*e.g.*, herbicides, insecticides, fungicides, rodenticides, acaricides) and chemical structure (group). In this context, thirteen insecticides, four fungicides, two acaricides and one herbicide were detected in the analysed samples. Figure 1 illustrates that 65% of detected pesticides are insecticides (dimethoate, phosalone, pirimiphos-methyl, malathion, diazinon, fenitrothion, methamidophos, methomyl, pirimicarb, acetomiprid, pyridaben, thiamethoxam and thiacloprid), 20% represent fungicides (procimidone, thiophanate-methyl, cymoxanil and dimethomorph), 10% are acaricides (propargite and clofentezin) and 5% correspond to herbicides (nicosulphuron).

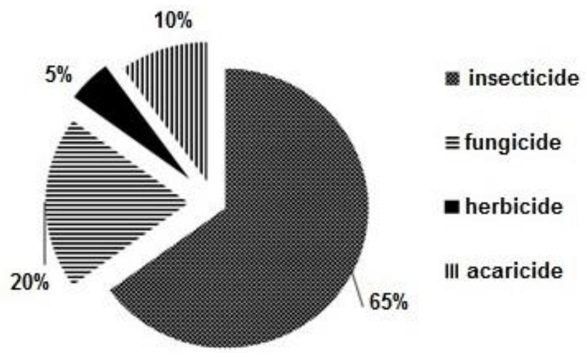

Figure 1. The detected pesticides classified by target organism.

The detected pesticides belong to ten chemical groups: organophosphates, carbamates and thiocarbamates, dicarboximides, benzimidazoles, triazines, urea derivatives, sulphur compounds, neonicotinoids, piridazinones, morpholines (Table 3). Obtained data show that the most commonly identified group was organophosphorous which included dimethoate, phosalone, pirimiphos-methyl, malathion, diazinone, fenitrothion and methamidophos. Dimethoate was found in 18 commodities (85.7%), propargite - in 14 (66.7%) and metamidophos in 5 (23.8%) from 21 different vegetables and fruits.

The analysis of pesticide residues in apple samples revealed one fungicide (dimethomorph from morpholines), five insecticides (four organophosphates: dimethoate, malathion, fenitrothion, diazinon and pyridaben from pyridazinones group), one herbicide (nicosulphuron from urea derivatives) and one acaricide (propargite from sulphur compounds group). Similarly, in samples of table grapes four insecticides (two organophosphates: dimethoate and phosalone; thiacloprid from neonicotinoids group and pirimicarb from carbamates and thiocarbamates groups), one acaricide (clofentezine from triazines groups), two fungicides (thiophanate-methyl from benzimidazoles and procymidone from dicarboximides groups) were detected.

The values of pesticides identified residue levels were used to calculate the EDI expressed in milligram pesticides per kilogram of body weight per day (mg/kg bw/day). The EDI values for pesticide residues found in the analysed vegetables and fruits are the estimation of pesticide exposure calculated according to the international guidelines of the Food and Agriculture Organization, European Food Safety Authority and World Health Organization. Estimation of exposure levels and risk assessment is recommended by the World Health Organization as an important activity as it provides reliable data of dietary intakes of contaminants. The ADI is the estimation of the amount of a substance in food (mg/kg bw/day) that can be ingested daily over a lifetime without appreciable health risk to the consumer [32].

The EDIs have been estimated between 0.000001 and 0.0002 mg/kg of bw/day (Table 4). Calculated values of EDIs are lower than the levels of ADI. Claeys W.L. *et al.* reported that the majority of pesticide residues exposure was 100 times lower than ADI [33]. Mohamed T.A. *et al*. also established that EDIs of pesticides ranged from 0.03% to 40% of the ADIs, depending on pesticide concentration and food consumption in Egypt [34].

Furthermore, the EDI values were used to calculate the hazard index for the identified compounds. The calculated hazard indices ranged from 0.000004 up to 0.15 for the analysed pesticides (Table 4). The highest value of hazard index was calculated for diazinon, being of 0.15. In this analysed sample of apples, the pesticide residues exceeded MRL 1.7 times. Scientists from Pakistan, Syed, J.H. *et al*., also established higher risk index value for diazinon in apple [35].

Table 3

Chemical groups of detected pesticides and commodities.

Identified chemical groups and pesticides	*Number of samples*	*Food commodities*	*Number of commodities*
Organophosphates			
Dimethoate	812	table grapes, pear, plum, apricot, apple, peach, potato, onion, cucumber, tomato, pepper, sweet pepper, cabbage, garlic, cauliflower, salad, melon, watermelon	18
Phosalone	42	table grapes, cucumber, tomato, sweet pepper, pumpkin, eggplant, watermelon	7
Pirimiphos-methyl	2	tomato	1
Malathion	8	cabbage, apple	2
Diazinone	60	apple	1
Fenitrothion	60	apple	1
Methamidophos	31	plum, apricot, tomato, cauliflower, watermelon	5
Carbamates and thiocarbamates			
Methomyl	1	tomato	1
Pirimicarb	6	table grapes	1
Dicarboximides			
Procimidone	6	sweet pepper, table grapes	2
Benzimidazoles			
Tiophanate-methyl	2	table grapes	1
Urea derivatives			
Nicosulphuron	28	table grapes, apple, sweet pepper	3
Cymoxanil	1	potato	1
Triazines			
Clofentezine	3	salad, table grapes	2
Sulphur compounds			
Propargite	95	cucumber, tomato, sweet pepper, cabbage, cauliflower, eggplant, lettuce, asparagus, melon, apple, table grapes, pear, plum, pineapple	14
Neonicotinoids			
Acetomiprid	1	potato	1
Thiamethoxam	2	cabbage	1
Thiacloprid	6	table grapes	1
Piridazinones			
Pyridaben	27	apple	1
Morpholines			
Dimethomorph	1	apple	1
Total no. of samples	**1194**		

Table 4

The estimated daily intake (EDI) and the hazard index (HI) of pesticide residues detected in vegetables and fruits.

Food commodities	*Detected pesticides*	*Mean, mg/kg*	*MRL, mg/kg*	*EDI, mg/kg bw/day*	*ADI, mg/kg bw/day*	*HI*
Potato	Acetamiprid	0.0250	0.01	0.00013	0.025	0.005
	Cymoxanil	0.0200	0.01	0.0001	0.013	0.008
	Dimethoate	0.0129	0.01	0.00007	0.001	0.07
Onion	Dimethoate	0.0104	0.01	0.000007	0.001	0.007
Cucumber	Dimethoate	0.0138	0.01	0.000005	0.001	0.005
	Phosalone	0.0305	0.01	0.00001	0.01	0.001
	Propargite	0.0820	0.01	0.00003	0.03	0.,001

Continuation of Table 4

Food commodities	*Detected pesticides*	*Mean, mg/kg*	*MRL, mg/kg*	*EDI, mg/kg bw/day*	*ADI, mg/kg bw/day*	*HI*
Tomato	Dimethoate	0.0201	0.01	0.000017	0.001	0.017
	Phosalone	0.0149	0.01	0.00001	0.01	0.001
	Imidacloprid	0.0567	0.50	0.00005	0.06	0.0008
	Methamidophos	0.0121	0.01	0.00001	0.001	0.01
	Methomyl	0.0500	0.01	0.00004	0.0025	0.016
	Propargite	0.0370	0.01	0.00003	0.030	0.001
	Pirimiphos-methyl	0.0600	0.01	0.00005	0.004	0.01
Sweet pepper	Dimethoate	0.0146	0.01	0.000004	0.001	0.004
	Phosalone	0.0150	0.01	0.000004	0.01	0.0004
	Nicosulphuron	0.0300	0.01	0.000008	2.0	0.000004
	Propargite	0.0257	0.01	0.000007	0.03	0.0002
	Procimidona	0.0200	0.01	0.000005	0.0028	0.002
Cabbage	Dimethoate	0.0172	0.01	0.00002	0.001	0.02
	Malathion	0.0400	0.02	0.00004	0.03	0.001
	Propargite	0.0175	0.01	0.00002	0.03	0.0006
Pumpkin	Phosalone	0.0307	0.01	0.000008	0.01	0.0008
Eggplant	Phosalone	0.0170	0.01	0.000002	0.01	0.0002
	Propargite	0.0363	0.01	0.000004	0.03	0.0001
Garlic	Dimethoate	0.0171	0.01	0.000001	0.001	0.001
Melon	Dimethoate	0.0200	0. 01	0.000006	0.001	0.006
	Propargite	0.0250	0.01	0.000007	0.03	0.0002
Watermelon	Dimethoate	0.0167	0.01	0.000005	0.001	0.005
	Phosalone	0.0155	0.01	0.000004	0.01	0.0004
	Methamidophos	0.0200	0.01	0.000006	0.001	0.006
Apple	Diazinon	0.0167	0.01	0.00003	0.0002	0.15
	Dimethoate	0.0207	0.01	0.00003	0.001	0.032
	Dimethomorph	0.0200	0.01	0.00003	005	0.0006
	Fenitrothion	0.0145	0.01	0.00002	0.05	0.0005
	Malathion	0.1186	0.02	0.0002	0.03	0.006
	Nicosulphuron	0.0583	0.01	0.000092	2.0	0.00005
	Propargite	0.0548	0.01	0.000087	0.03	0.003
	Pyridaben	0.0781	0.05	0.0001	0.01	0.012
Table grapes	Clofentezine	0.2000	0.02	0.000093	0.02	0.005
	Dimethoate	0.0150	0.01	0.000007	0.001	0.007
	Phosalone	0.0150	0.01	0.000007	0.01	0.0007
	Nicosulphuron	0.0833	0.01	0.00004	2.0	0.00002
	Propargite	0.0250	0.01	0.00001	0.03	0.0004
	Pirimicarb	0.1067	0.01	0.00005	0.035	0.0014
	Procymidone	0.0178	0.01	0.000008	0.0028	0.003
	Thiacloprid	0.0283	0.01	0.00001	0.01	0.0013
	Thiophanate-methyl	0.4000	0.10	0.0002	0.08	0.0023
Pear	Dimethoate	0.0126	0.01	0.000007	0.001	0.007
	Phosalone	0.0215	0.01	0.000012	0.01	0.0012
	Propargite	0.0250	0.01	0.000014	0.03	0.0005
Plum	Dimethoate	0.0147	0.01	0.000008	0.001	0.008
	Methamidophos	0.0128	0.01	0.00007	0.001	0.07
	Propargite	0.0250	0.01	0.000014	0.03	0.0005
Peach	Dimethoate	0.0250	0.01	0.000014	0.001	0.014
Apricot	Dimethoate	0.0200	0.01	0.00001	0.001	0.01
	Methamidophos	0.0200	0.01	0.00001	0.001	0.01
				***$\sum$= 0.002**		****$\sum$= 0.5477**

*$\sum$ - *is the total EDI;*
**$\sum$ - *is the total HI.*

The EDI for diazinon was found less than ADI, which might be explained by the fact that dietary intake of fruits and vegetables per person was estimated according to minimum norms of food included in a living-wage food basket [30], which does not reflect real food consumption pattern. According to these norms, consumption of apples is equal to 0.095 kg/day. EDI in this case is equal to 0.00003 mg/kg bw/day. In Republic of Moldova, the actual consumption of apples by the population is much higher. Although the consumption data used in this study is the most updated, there is a need for data, which reflects real food consumption patterns in the Republic of Moldova. Here, it should be noted as an example the study of Boon, P.E. *et al*. regarding the real data collection on food consumption levels of Dutch infants [36]. The total exposure to the detected pesticide residue, obtained by summing EDI values, is equal to 0.002 mg/kg bw/day. Thus, the calculated hazard index values show that the long-term consumption of tested vegetables and fruits could not pose a health risk for the population of Republic of Moldova as the obtained values for the identified hazard indices were less than one and indicate no risk associated with consumption of such vegetables. Knezevic Z. *et al*. reported that long term exposure of Croatian consumers did not raise health concerns [37]. Ahmed, M.T. *et al.* also concluded that the EDIs of the different pesticides from vegetable consumption are not considered a public health problem [34]. However, exceeding MRLs should be considered as a threat to the population health by higher consumption rates of fruits and vegetables polluted by pesticides. The lack of real consumption data for different group of population is one of the barriers to conduct research concerning the actual exposure to pesticides. It should be noted that pesticides can cause harmful effects over an extended period, usually following repeated or continuous exposure at low levels. Low-dose exposure does not always cause immediate effects, but over time, it can cause very serious illnesses.

Conclusions

The monitoring of pesticide residues in fruits and vegetables has an important role in providing data on the pesticide residues of the fresh products. A total number of 5206 samples from twenty one different vegetables and fruits were collected and analysed during 2009-2017. In 1194 of analysed samples (22.9%) residues of pesticides were found in concentrations exceeding the MRLs. Thirteen insecticides, four fungicides, two acaricides and one herbicide were detected. The EDIs have been estimated between 0.000001 and 0.0002 mg/kg bw/day. Calculated values of EDI are lower than the levels of ADI.

The total exposure to pesticide residues is equal to 0.002 mg/kg bw/day. The calculated hazard indices (risk estimation) ranged from 0.000004 up to 0.15 for the analysed pesticides. The highest value of hazard index was calculated for diazinon, being of 0.15.

The results of our study show that the long-term consumption of vegetables and fruits contaminated with pesticides can pose a health risk for the population of the Republic of Moldova, since the minimum norms used for risk estimation do not reflect real food consumption pattern in the Republic of Moldova.

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