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A review of research studies into pesticide residues in food in Lithuania

Julijonas PETRAITIS[1], Inga JARMALAITĖ[1], Virginijus VAIČIŪNAS[1], Remigijus UŠČINAS[2], Giedrė JANKOVSKIENĖ[3]

[1]National Food and Veterinary Risk Assessment Institute
J. Kairiūkščio 10, Vilnius, Lithuania
E-mail: julius.petraitis@zmail.lt

[2]Joint stock company "Labtarna"
Kauno 1A, Vilnius, Lithuania

[3]National Public Health Surveillance Laboratory
Žolyno 36, Vilnius, Lithuania

Abstract

The current paper presents the evolution of pesticide research in Lithuania during the period 1970–2010. Analyses were based on single residue methods until 1993. Most of the products were of domestic origin and it was known exactly which pesticides had been used, therefore it was possible to obtain the necessary information about the contamination by single residue methods. Simple and inexpensive methods were used, which allowed five laboratories to analyze 3–5 thousand samples per year. In 1972, a high level of contamination (up to 8.1%) in food of animal origin was determined. This resulted from environmental pollution and feed contamination with organochlorine pesticides. Later this contamination decreased, and animal products no longer posed a major pollution problem. The contamination of food of plant origin was at level 1–3%. A significant increase in imports and the lack of information on pesticides used made the single residue methods unsuitable to ensure effective contamination control. The multiresidue methods were developed. Implementation of multiresidue methods enables a laboratory to determine up to 300 pesticides per sample. A total of 4593 samples of food of plant origin were analysed by multiresidue methods, pesticide residues were determined in 1428 samples (31%), exceedances of maximum residue levels (MRLs) were determined in 120 samples (2.6%). This data are comparable with the pesticide monitoring results obtained in other European countries.

Key words: food, multiresidue method, pesticide, pesticide residues, single residue method.

Introduction

Various materials as pesticides have been used for many years to control pests. The first synthetic organic pesticides were organochlorine compounds, such as dichlorodiphenyltrichloroethane (DDT). It was cheap to produce, very toxic to insects, and much less toxic to mammals. From 1950 to 1970, more than 20 thousand tonnes of DDT were used annually over large areas of the Soviet Union. As a result, dangerously high residue levels were found in both food and human tissue (DDT Factsheet, 1998). In 1970, DDT was banned for use in agriculture, but it remained a widespread and persistent environmental contaminant. Long after banning, DDT together with its breakdown products remained dangerous to human health and the environment and even after 20–30 years their residues were detected in food (Pesticide Residues…, 1996). DDT has not been officially used in Lithuania since 1970; however, organochlorine pesticides were used to protect cotton, wheat, and to fight with malaria in the southern regions (DDT Factsheet, 1998). According to Food safety issue (Pesticide Residues…, 1996), high contamination of cereals with 1.2 mg kg^{-1} 2,4'-DDT and 1.3 mg kg^{-1} 4,4'-DDT was determined in wheat from the Soviet Union.

The control of the pesticide residues in Lithuania covers a forty-year period. The information on the pesticide residues analysis was presented in the conferences (Uščinas et al., 1999; Petraitis et al., 2001; 2007; 2009; 2011; Jarmalaitė et al., 2003; Vaičiūnas et al., 2011; Golubevas et al., 2012). However, there was no published scientific information on pesticide residue analysis in Lithuania during this period. Therefore the objective of this paper was to summarise the experience of Lithuanian chemists in pesticide residue analysis.

Results and discussion

The official food control was established in the Soviet Union and in Lithuania in 1970. Four USSR governmental institutes were working continuously on developing methods for determination of pesticide residues and toxicity evaluation. They developed single residue methods, and established maximum residue limits for the pesticides used in the USSR. Control was conducted by hygiene laboratories of Ministry of Health. The main attention was paid to organochlorine pesticides: DDT and metabolites, kelthane (dicofol), hexachlorobenzene, isomers of 1,2,3,4,5,6-hexachlorcyclohehane, polychlorpinene, heptachlor over the 1970–1975 period. Other important groups were carbamates, organic mercury compounds, and copper residues. During the 1970–1971 period, over 3000 environment (surface and drinking water from wells) and food samples were analysed. High levels of pesticide residues were determined in food of animal origin. The number of food and water samples increased to 4000 (3607 food samples + 398 water samples) in 1972 and to 6000 samples in 1973–1976. In later years, the number of samples decreased to 3000–4000. The number of pesticides analysed was not high, up to 30 compounds mainly organochlorine pesticides, copper and mercury residues. High contamination levels of food of animal origin were determined mostly in fish (22%) and eggs (4.4%). A possible reason for such a high level of contaminated products was environmental pollution and import of wheat for animal feed production from the regions where DDT was used. There were exceedances of MRLs in 1–1.5% of samples of milk and milk products and 0–2.3% in the samples of food of plant origin. More detailed information is presented in Table 1.

Table 1. Results of food analyses in 1972, 1976, 1980, 1983, 1986 and 1993

Foodstuff	No. of samples	Above the MRL	%	No. of samples	Above the MRL	%	No. of samples	Above the MRL	%
		1972			1976			1980	
Fruits and vegetables	1184	27	2.3	1133	5	0.44	796	9	1.3
Cereals and other food of plant origin	279	3	1.1	295	0	0	223	0	0
Milk and milk products	1500	15	1.0	2195	18	0.8	1510	22	1.5
Meat, fish and other food of animal origin	644	52	8.1	855	32	3.7	718	12	1.7
Total food	3607	97	2.7	4478	55	1.1	3247	43	1.3
Water	398	5	1.3	518	0	0	332	3	0.9
		1983			1986			1993	
Fruits and vegetables	918	13	1.4	1733	37	2.1	613	10	1.6
Cereals, oil and other food of plant origin	195	7	3.6	401	0	0	233	1	0.4
Milk and milk products	1226	21	1.7	691	2	0.3	387	2	0.5
Meat, fish and other food of animal origin	691	5	0.7	701	0	0	277	0	0
Total food	3016	46	1.5	3526	39	1.1	1510	13	0.86
Water	222	2	0.9	247	0	0	187	2	1.1

MRL – maximum residue level

Contamination of food of animal origin considerably decreased from 1980 as a result of decreasing environmental pollution and less contaminated feeds by organochlorine pesticides. No exceedance of MRLs was determined in meat, eggs, and fish since 1986. More attention was paid to analysis of food of plant origin and the number of these samples exceeded 2000 per year. Frequency of MRL exceedances was about 1.5–2.0%. The results of analyses of pesticide residues in food in 1983, 1986 and 1993 are presented in Table 1.

Methods for determination of carbamates, organophosphosphorus and other pesticides were implemented in this period to analyse food of plant origin. More than 600 single residue methods were developed for determination of up 100 pesticide residues in different kind of plant matrixes until 1985. The pesticide residues control system was implemented in the Soviet Union for evaluation and statistical analysis of food contamination. This system was well organised for domestic products and was suitable for planned soviet economy. All of the necessary information on what pesticides were used for a particular commodity was provided with the sampling documents making it easier for the laboratories to select the single residue methods. After the year 1990, the main system of agriculture in USSR – collective farms collapsed, but the same pesticides were used for another few years in agriculture. Lithuanian food control system collaborated with the former USSR pesticide residue control system centre in the Ukraine, until 1992. However, this system was not effective to control contamination of imported food of plant origin.

Lithuanian farmers started to use new pesticides purchased from western companies. Most of the activities were aimed to evaluate possible contamination of plant origin domestic products with pesticide residues during field test necessary for registration of a new pesticide formulation in Lithuania. However, the control of imported products of plant origin by single residue methods was ineffective.

Environmental pollution with organochlorine pesticides stipulated accumulation of these pesticide residues in fish, animal and human tissues. Analyses of pesticide residues accumulated in human fat were conducted in 1998. Aldrin, dieldrin, 2,4'-DDE (dichlorodiphenyldichloroethylene), 4,4'-DDE, 2,4'-DDD (dichlorodiphenyldichloroethane), 4,4'-DDD, α- and β-endosulfanes, endrine, α-hexachlorcyclohexane (HCH), lindane, heptachlor, 2,4'-DDT, 4,4'-DDT residues were analysed in 19 samples. The residues of 4,4'-DDE, α-HCH, lindane, were determined in 15 (78%) samples (Uščinas et al., 1999). Similar pesticides were determined in human fat by researchers of other countries. Organochlorine pesticide residues were found in 63.2% of samples in Spain (Molina et al., 2005).

The total level of contamination of food of animal origin was decreasing over the years and since 1994 only a few times MRLs were exceeded in domestic products of animal origin. However, the measurable residues in some commodities were still found in food in Lithuania and other European Union (EU) countries. According to the EU-coordinated programme, 473 butter samples and 559 egg samples were analysed during the calendar year 2009. The measurable residues were found in 19.9% of the butter samples. Alpha-HCH and endosulfan exceeded the MRL in two butter samples. The most frequently found residues in butter were hexachlorbenzene, DDT, methohychlor, dieldrin, HCH and endosulfan. The measurable residues were found in 4.5% of egg samples and in one sample DDT exceeded the MRL, other residues found below the MRL were hexachlorobenzene, lindane, dieldrin, chlordane, chlorpyrifos and cypermetrin (Scientific Report…, 2011). Sixteen butter and 12 chicken egg samples were analysed in Lithuania according to this programme; two butter samples contained α-HCH below the MRL. During the calendar year 2010, no pesticide residues were found in 10 milk and 8 meat samples, analysed in Lithuania. Although most of these pesticides are no longer used in the EU and in most third countries, they are still found in food because of their persistence and tendency to accumulate in the food chain.

A new step to control the pesticide residues in food of plant origin started in 1998. The multiresidue method by capillary gas chromatography (GC) with electron capture and nitrogen phosphorus detectors has been developed in the laboratory of the National Nutrition Centre. Ethyl acetate was chosen as the extraction solvent before the clean-up procedure on gel permeation chromatography column filled with "Bio Beads S-X3" sorbent, while ethyl acetate/cyclohexane 1:1 was used as the mobile phase. This method included 55 pesticides and was used for import control mainly. Thirty samples of imported fruits and vegetables (oranges, mandarins, lemons, apples celery, tomatoes, and others) and wine were analysed in 1999. Pesticide residues were determined in 23 samples; however, most of the results had not been evaluated, as there were no national MRLs for these products. Investigations were expanded in 2000. A total of 200 samples were analysed, of which 114 were of domestic origin. The highest contamination was determined in imported fruits, especially citrus fruit and grapes.

The reorganisation of food control system took place in 2000. State Food and Veterinary Service was established, whose responsibilities included pesticide residue analyses and preparation of a control system. The pesticide residue control in food of plant origin was renewed in 2001 and 221 samples were analysed for 46 pesticide residues. Analysis of domestic samples indicated low contamination of domestic products. Pesticide residues were determined in 6 samples out of 55 (11%) and none exceeded the MRL. The imported samples were contaminated in higher degree, pesticide residues were determined in 49 samples out of 166 (29.5%). MRLs were exceeded in 5 samples out of 166 (3.0%) – cucumbers, head cabbages, apples, peppers and raisins.

Acetone and ethyl acetate/cyclohexane mixture (1 + 1) were used as extraction solvents before gel permeation chromatography clean-up procedure with the mobile phase ethyl acetate/cyclohexane (1 + 1) and additional clean-up with mini-silica column following GC and detection with the electron capture and nitrogen and phosphorus detector, mass spectrometry (MS) detectors. This system allowed determination of 130 pesticide residues in 2006. The new analytical systems: GC/MS and liquid chromatography-tandem mass spectrometry (LC-MS/MS) were implemented in 2007. This allowed the pesticide residue scope to be increased up to 230 in 2008. The QuEChERS (quick, easy, cheap, effective, rugged, safe) is the acronym for a highly beneficial analytical approach that vastly simplifies the analysis of multiple pesticide residues in fruit, vegetables, cereals and processed products thereof. QuEChERS sample preparation approach entailing solvent extraction with acetonitrile, partitioning with magnesium sulphate alone or in combination with other salts followed by clean up using solid-phase extraction. The combination of these extractions – clean-up methods with GC/MS and LC-MS/MS analysis allowed the number of pesticides that can be determined in one sample to be increased up to 274 in 2009, and up to 300 in 2010 and the limits of determination for many residues to be decreased to 0.002–0.01 mg kg^{-1}. However, it is not possible to analyse all of the pesticide residues by multiresidue methods, laboratory has to apply single residue methods for the determination of dithiocarbamates, chlormequat, and glyphosate. The new methods allow determination of pesticide residues with the concentrations below 0.01 mg kg^{-1}.

The monitoring programme of pesticide residues in food products according to EU requirements was prepared and implemented in 2002. More than 60 different commodities both imported and domestic were included in the programme. The programme covers 69 pesticides including some isomers. The contamination of domestic samples was low – 3 samples out of 65 (4.6%) were with residues below the MRL. The number of samples containing pesticide residues was higher in imported products – 109 samples out of 350 (31.1%). MRLs were exceeded in 6 samples out of 350 (1.7%). Percentage of samples obtained by the EU monitoring programme with detectable residues was 37% and frequency of MRLs exceedances reached 5.1% (Monitoring…, 2004).

Similar results were obtained in 2003, except an increase of exceedances of MRLs to 2.6%, depending on the contaminated tea import from China. Exceedances of MRLs were determined in grapefruits – 2 samples, oranges, carrots and peppers – 4 samples. The main residue determined in peppers was endosulfan with quantity between 0.2–0.5 mg kg^{-1}. The European MRLs increased from 0.2 mg kg^{-1} to 0.5 mg kg^{-1}, and there were no exceedances of endosulfan residues later. Percentage of samples with detectable residues obtained by the EU monitoring programme was 37% and frequency of MRLs exceedances reached 5.5% (Monitoring…, 2005).

The level of samples with detectable residues 21.8% and exeedances of MRLs 1.7% was lower in 2004, a similar situation was in 2005 samples with detectable residues 25.9% and exeedances of MRLs 1.5%. Since

2004, the data obtained by the national pesticide monitoring programme have been included in the EU monitoring results data base. Percentage of samples with detectable residues obtained by the EU monitoring programme in 2004 and 2005 was 39.7% and 41.0% and frequency of MRLs exceedances was 4.7% and 4.7%, respectively (Monitoring…, 2006; 2007).

The level of samples with detectable residues 28.0% and exeedances of MRLs had reached 2.3% in 2006, 30.1% and 4.6% in 2007, 21.2% and 2.6% in 2008. Percentage of samples with detectable residues obtained by the EU monitoring programme in 2006, 2007 and 2008 was 42.0, 45.0 and 35.7 % and frequency of MRLs exceedances was 4.4, 4.2 and 3.5 %, respectively (Monitoring…, 2007; 2008; Annual Report…, 2009; 2010). According to USA monitoring programme, percentage of samples with detectable residues was 28.5, 39.7 and 34.9 % in 2006, 2007 and 2008 for domestic products and 28.1, 31.0, and 23.0 % for imported products. Pesticide levels exceeding established MRLs in 2006, 2007 and 2008 were 1.6, 2.3, and 0.9 %, in domestic products and 5.4, 4.7, and 4.7 % in imported samples (Pesticide Monitoring Program…, 2006; 2007; 2008). Brazilian monitoring programme team have reported monitoring data between 2001 and 2010. They found pesticide residues in 48.3% of the samples and less than 3% of the samples that had residue levels above the MRL (Jardim, Caldas, 2012). China's researchers found selected pesticide residues in 37.7% of the samples (Chen et al., 2011). The difference in the results depends on different factors, commodity selected, method of sampling, pesticide residue searched, method of analysis, country of origin. The selective evaluation of pesticide residues in fruits and vegetables from South America was done by Nordic project in 2007. High levels of pesticide residue were determined. 72% of samples contained pesticide residues at or MRL and 8.4% of samples contained pesticide residue above the MRL (Hjorth et al., 2011). In some cases thiabendazole and imazalil were determined; they are used as fungicides to protect citrus fruits and bananas during transportation.

Implementation of modern analytical methods has made it possible to increase the number of pesticides sought per sample to 300 and to decrease the limit of determination to 0.002 mg kg^{-1}. The level of samples with detectable residues increased to 44.6% and exceedances of MRLs to 3.4% in 2009 and to 59.8% of detectable residues with MRL exceedances to 4.2% in 2010. Another reason for increased exceedances of MRLs was that the higher proportion of fruit samples was imported from the third countries. These commodities are usually more contaminated with pesticide residues compared to the products from the EU countries. Percentage of samples with detectable residues obtained by the EU monitoring programme was 37.4% and frequency of MRLs exceedances fell down to 2.6% in 2009 (Scientific Report…, 2011). Results of pesticide residues analyses in Lithuania from 1999 to 2010 are presented in Table 2.

Table 2. Results of pesticide residues analyses in 1999–2010

Year	Samples analysed	With residues below MRL		With residues above MRL		Pesticide residues		Samples origin****		
	No.	No.	%	No.	%	Sought	Found	Dom	EU	Third
1	2	3	4	5	6	7	8	9	10	11
1999 total	30	23	76.7	0	0	55	26	0	17	13
Fruits*	21	18	85.7	0	0	55	22	0	10	11
Vegetables**	4	3	75	0	0	55	21	0	4	0
Wine	5	2	40	0	0	55	2	0	3	2
2000 total	200	57	28.5	3	1.5	55	22	114	48	38
Fruits	53	26	49.1	3	5.7	55	16	10	26	17
Vegetables	51	14	27.5	0	0	55	11	29	10	12
Cereals	75	9	12.0	0	0	55	4	75	0	0
Wine	21	8	38.1	0	0	55	3	0	12	9
2001 total	221	50	22.6	5	2.6	46	21	55	102	64
Fruits	91	22	24.2	2	2.2	46	15	9	33	49
Vegetables	119	28	23.5	3	2.6	46	11	46	62	11
Others***	11	0	0	0	0	46	0	0	7	4
2002 total	415	112	27.0	6	1.5	69	24	65	234	116
Fruits	179	71	39.7	3	1.7	69	19	14	93	72
Vegetables	206	40	19.4	3	1.5	69	12	42	139	25
Cereals	28	1	3.6	0	0	69	1	9	1	18
Others	2	0	0	0	0	69	0	0	1	1
2003 total	495	152	30.7	13	2.6	96	32	78	235	182
Fruits	146	64	43.8	3	2.1	96	25	11	63	72
Vegetables	242	85	35.1	10	3.9	96	15	53	147	42
Cereals	82	3	3.6	0	0	96	2	12	6	64
Others	25	0	0	0	0	96	0	2	19	4
2004 total	468	102	21.8	8	1.7	105	28	199	102	167
Fruits	147	70	47.6	5	3.4	105	23	23	28	86
Vegetables	236	27	11.4	3	1.3	105	14	127	61	48
Cereals	68	2	2.9	0	0	105	1	43	1	24
Others	17	3	17.6	0	0	105	3	6	2	9
Total 1999–2004	**1829**	**496**	**27.1**	**35**	**1.9**			**511**	**738**	**580**

Table 2 continued

1	2	3	4	5	6	7	8	9	10	11
2005 total	466	114	25.9	7	1.5	130	40	187	112	167
Fruits	179	91	50.8	6	3.4	130	33	29	53	97
Vegetables	237	22	9.3	1	0.4	130	16	128	56	53
Cereals	37	1	2.7	0	0	130	1	30	1	6
Others	13	0	0	0	0	130	0	0	2	11
2006 total	436	122	28.0	10	2.3	205	39	135	119	182
Fruits	156	83	53.2	6	3.8	205	27	29	31	96
Vegetables	210	28	13.3	3	1.4	205	18	80	69	61
Cereals	49	4	9.0	1	2.0	205	4	23	8	18
Others	21	7	26.9	0	0	205	6	3	11	7
2007 total	504	156	30.1	23	4.6	223	63	118	154	232
Fruits	205	93	45.4	18	8.8	223	54	29	53	123
Vegetables	233	58	24.5	5	2.1	223	30	63	85	85
Cereals	53	4	7.5	0	0	223	4	24	6	23
Others	13	1	7.7	0	0	223	6	2	10	1
2008 total	529	112	21.2	14	2.6	230	52	113	156	260
Fruits	226	88	38.9	7	3.1	230	37	30	58	138
Vegetables	224	19	8.5	6	2.7	230	28	65	83	76
Cereals	54	3	5.6	1	1.9	230	5	15	6	33
Others	23	2	4.2	0	0	230	4	2	9	12
2009 total	446	199	44.6	15	3.4	325	75	82	144	220
Fruits	265	150	56.6	8	3.0	274	71	5	51	135
Vegetables	112	44	39.3	6	5.4	274	35	18	76	18
Cereals	30	3	14.3	1	3.3	250	1	17	0	13
Others	11	0	0	0	0	274	0	0	9	2
Milk, eggs	28	2	7.1	0	0	48	1	28	0	0
2010 total	383	229	59.8	16	4.2	349	99	94	124	165
Fruits	213	161	75.6	14	6.6	302	97	14	58	141
Vegetables	101	59	58.4	1	1.0	301	52	39	53	9
Cereals	29	7	24.1	0	0	304	14	20	2	7
Others	22	2	9.1	1	4.5	301	12	3	11	8
Milk, meat	18	0	0	0	0	48	0	18	0	0
Total 2005–2010	**2764**	**932**	**33.7**	**85**	**3.1**			**729**	**809**	**1226**

Notes. * – term “Fruits” includes fruits, berries fresh, frozen and dried and nuts; ** – term “Vegetables” – vegetables fresh and frozen, beans, peas, mushrooms, oil plants, infusions, tea and spices; *** – term “Others” – baby and infant food, juice and other processed foods; **** – samples origin “Dom” means domestic (Lithuanian) products, “EU” – products from EU countries, “Third” – products imported from third (non EU) countries. MRL – maximum residue level.

Residues at or below the MRL were found most often in oranges, mandarins, grapes/raisins, pomegranates, peaches, bananas, pears, strawberries and tea. Residues above the MRL were determined in the samples of fruits: grapes/raisins, oranges, lemons, other citrus, plums, pears, apples; in the samples of vegetables: peppers tomatoes, cucumbers, carrots, aubergines, cauliflowers. High exceedances of MRLs were determined in 5 samples of tea from China. More detailed information on samples with pesticide residues exceeding the MRL is presented in Table 3. Exceedances of MRLs in these products were found in other EU countries too. Additionally, exceedances of MRLs were determined in wild fungi from China, melons, beans, spices, okra, spinach, lettuce. The monitoring results presented in the USA reports show high level of pesticide residues in pears, grapes/raisins, citrus fruit, peppers, cucumbers, tomatoes, carrots – similar to the results obtained by Lithuania researchers. Moreover, high level of pesticide residues was determined in imported blackberries, strawberries and other berries as well as mangoes, papaya, pepinos, bitter melon, peas, beans, eggplant, spinach and other vegetables (Pesticide Monitoring Program…, 2006; 2007; 2008). Apples, papayas, sweet peppers and strawberries were among products with the highest percentage of samples with residues above the MRL found in Brazilian pesticide residues monitoring programme (Jardim, Caldas, 2012). The commodities with the highest level of pesticide residues found in China were cabbage, legumes and leaf mustard (Chen et al., 2011).

Table 3. Samples with pesticide residues exceeding the maximum residue levels (MRLs) (2001–2010)

Sampling year	Commodity	Samples found/analysed	Country of origin	Pesticide residue
1	2	3	4	5
2001	apples	1/27	Poland	malathion
	raisins	1/5	Turkey	acephate
	peppers	1/7	Spain	endosulfan
	cucumbers	1/3	The Netherlands	endosulfan
2002	mandarins	2/15	Morocco, Greece	chlorpyrifos, parathion-methyl
	plums	1/5	France	dicofol
	peppers	2/38	Spain	endosulfan
	tomatoes	1/48	The Netherlands	dicofol

Table 3 continued

1	2	3	4	5
2003	grapefruit	2/8	Turkey	chlorpyrifos
	oranges	1/18	Morocco	chlorpyrifos
	peppers	4/70	Spain	endosulfan
	carrots (dried)	1/10	Austria	4,4’-DDE
	tea	5/24	China	fenvalerate
2004	lemons	2/6	Turkey	bromopropylate
	plums	1/2	Unknown	phosalone
	raisins	2/28	Iran	fenvalerate
	tomatoes	3/37	Lithuania	vinclozolin
2005	lemons	1/8	Turkey	parathion-methyl
	apples	1/24	Poland	dimethoate
	raisins	4/32	Iran	fenvalerate
	tomatoes	1/30	Lithuania	vinclozolin
2006	grapes	1/18	Turkey	imazalil
	cauliflower	1/11	Lithuania	dithiocarbamates
	tomatoes	1/22	Spain	endosulfan
	apples	1/25	Lithuania	dimethoate
	pappers	1/14	Spain	amitraz
	plums	1/6	France	chlorothalonil
	wheat	1/13	Lithuania	tebuconazole
	raisins	1/9	Iran	fenvalerate
	orange	1/14	Egypt	profenofos
	lemon	1/14	Turkey	parathion
2007	peaches	5/22	Spain	carbendazim, azinphos-methyl, triadimefon, dichlorvos, captan
	apples	1/18	France	carbendazim
	strawberries	2/24	Poland	endosulfan, carbaril
	carrots	1/15	Lithuania	diazinon
	pomegranate	3/7	Egypt	lambda-cyhalothrin, ethion
	cucumbers	1/16	Lithuania	endosulfam
	oranges	2/29	Egypt	formathion, fenitrothion
	plums	3/8	Hungary (2), Germany	binapacryl, flutriafol, carbaryl
	tomatoes	1/24	Spain	carbendazim
	head cabbage	1/17	The Netherlands	carbendazim
	pears	1/9	The Netherlands	carbaryl
	bananas	1/15	Equador	carbaryl
	tea	1/11	Egypt	bromoxynil
2008	oranges	3/12	Egypt, Spain, Morocco	diazonone, dimethoate, boscalid
	pears	2/13	The Netherlands, Spain	chlorpyrifos, demeton-S-methyl
	pomegranates	2/5	Egypt	ethion
	cucumbers	2/17	Lithuania	demeton-S-methyl, vinclozolin
	tomatoes	1/15	Lithuania	vinclozolin
	bean	1/2	Poland	demeton-S-methyl
	tea	2/8	China	buprofezin, thiazophos
	wheat	1/17	Lithuania	flucitrinate
2009	pomegranate	5/7	Turkey (2), Israel (2), Egypt	acetamiprid, carbendazim, ethion, lambda-cyhalothrin, thiacloprid
	grapes	3/31	Turkey, Italy, USA	imazalil, chlorpyrifos-methyl, tecnazene
	peppers	1/16	Poland	thiophanate-methyl
	aubergines	2/15	Poland, The Netherlands	oxamyl
	peas	1/17	Belgium	azoxystrobin
	tomatoes	1/8	Poland	carbendazim
	lettuce	1/2	The Netherlands	dimethoate
	rice	1/11	Vietnam	acetamiprid
2010	grapes/raisins	8/34	Egypt (2), Iran (2), Macedonia (3), Turkey	dimethoate, chlormequat, procymidone, methomyl, thiodicarb
	pomegranate	2/8	Egypt, Turkey	carbendazim
	oranges	4/32	Egypt	fenitrothion, dimethoate, diazinon
	tea	1/3	India	acetamiprid, monocrotofos
	baby food	1/11	Slovenia	piperonyl butoxide

Many commodities contained more than one residue per product, up to 9 residues in grapes and tea were determined, up to 5–9 residues in citrus fruits – orange, mandarins, lemons, peaches, pears and up 3–5 residues in pomegranates, plums, cucumbers, tomatoes, strawberries. The most frequently detected pesticide residues were imazalil, thiabendazole, chlorpyrifos, maneb group, procymidone, methidathion, lambda-cyhalothrin, carbendazim, iprodione, orthophenylphenol, vinclozolin, endosulfan, pyrimethanil, fenhexamid, prochloraz, cyprodinil, boscalid. The reason of high detection frequency of thiabendazole, imazalil residues is that these pesticides are widely used for post-harvest treatment of citrus fruits and bananas in order to preserve fruit during the transport process which may take several weeks. Residues of these pesticides were found in 70% of the imported citrus fruits and bananas at or below the MRL.

Conclusions

1. A high level of organochlorine pesticide contamination in meat and fish was determined in Lithuania during the period 1970–1976. Contamination gradually decreased after the most dangerous pesticides had been banned.

2. The level of contamination in milk and dairy products was lower, compared with that of meat and fish during the 1970–1976 period.

3. The residues at or below the maximum residue level (MRL) in food of animal origin were found in 10–20% of the samples analysed, exceedance of MRL has not been determined during the last 12 years in Lithuania.

4. Percentage of samples with residues at or below the MRL in food of plant origin ranged from 20% to 60% of the samples analysed, exceedances of MRLs ranged from 1.5% to 4.6%.

5. Grapes/raisins, oranges, pomegranates and tea imported from third countries, were commodities in which pesticide residues were most often determined. Most of these matrixes contained more than one pesticide residues.

6. Contamination of domestic products was not high, pesticide residues exceedances of MRLs were determined in 13 samples out of 1240: apples, cucumbers, tomatoes, cauliflower, carrots and wheat.

7. Imazalil, thiabendazole, chlorpyrifos, maneb group, procymidone, methidathion, lambda-cyhalothrin, carbendazim, iprodione, orthophenylphenol, vinclozolin, endosulfan, pyrimethanil, fenhexamid, prochloraz, cyprodinil, boscalid were the pesticides whose residues were most frequently detected.

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Pesticidų likučių maisto produktuose tyrimų Lietuvoje apžvalga

J. Petraitis[1], I. Jarmalaitė[1], V. Vaičiūnas[1], R. Uščinas[2], G. Jankovskienė[3]

[1]Nacionalinis maisto ir veterinarijos rizikos vertinimo institutas
[2]UAB „Labtarna“
[3]Nacionalinė visuomenės sveikatos priežiūros laboratorija

Santrauka

Straipsnyje pateikta pesticidų tyrimų Lietuvoje raida nuo 1970 iki 2010 m. Pirmuoju laikotarpiu iki 1993 m. tyrimai buvo atliekami pavienių pesticidų likučių nustatymo metodais. Dauguma produktų buvo vietinės kilmės ir žinota, kokie pesticidai naudoti, todėl, taikant paprastus ir pigius pavienių pesticidų likučių nustatymo metodus, buvo galima ištirti didelį kiekį mėginių. Lietuvoje pesticidų likučius tyrė penkios laboratorijos, kurios per metus ištirdavo po 3–5 tūkstančius mėginių. Pirmaisiais metais nustatyta didelis gyvūninės kilmės produktų užterštumas pesticidų likučiais. 1972 m. dalis mėsos ir žuvų produktų, kuriuose pesticidų likučių koncentracija viršijo didžiausią leidžiamą kiekį, sudarė net 8,1 %. Vėliau šis užterštumas smarkiai mažėjo, ir gyvūninės kilmės produktų tarša nebekėlė didesnių problemų. Augalinės kilmės maisto užterštumas labai nekito ir sudarė 1–3 %.
Žymiai padidėjus importui ir trūkstant informacijos apie naudotus pesticidus, pavieniais pesticidų likučių nustatymo metodais jau nebebuvo galima užtikrinti efektyvios taršos kontrolės, todėl buvo diegiami ir tobulinami daugiapakopiai pesticidų likučių nustatymo metodai. Šie metodai viename mėginyje leidžia nustatyti iki 300 pesticidų likučių. Buvo ištirti 4593 augalinės kilmės mėginiai, pesticidų likučių nustatyta 1428 mėginiuose (31 %). Didžiausios leidžiamos koncentracijos viršijimas nustatytas 120 mėginių (2,6 %).

Reikšminiai žodžiai: daugiapakopis pesticidų likučių nustatymo metodas, maistas, pavienių pesticidų likučių nustatymo metodas, pesticidai, pesticidų likučiai.