OVERVIEW OF SOME B GROUP CONTAMINANTS (ORGANICHLORINE PESTICIDES AND PCB'S, ORGANOPHOSPHORUS COMPOUNDS, HEAVY METALS AND RADIONUCLIDES) MONITORING DATA IN PRIMARY PRODUCTS OF ANIMAL ORIGIN IN LITHUANIA THROUGHOUT TEN (1999–2008) YEARS PERIOD

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Abstract. The goal of this work was to analyze contamination of Lithuanian primary products of animal origin with some of the persistent organic pollutants (POP's), heavy metals and radionuclides in the ten years period of monitoring. The influence of the year, the type of pollutant and the type of product on the number of tested and non-compliant samples (samples which exceeded the determined maximum residue limit) was analysed. The analyses showed a significant impact of all those factors on the number of tested and non-compliant samples. Before 2004, only residues of chloroorganic pesticides and PCB were tested in Lithuania. From 2004, organic compounds of phosphorus and dioxins were started to be monitored. In poultry, fish and wild game residues of chloroorganic pesticides were found which exceeded the permissible limits; most of them in game meat ($p \leq 0.05$). The remains of chloroorganic pesticides, DDT ($p \leq 0.05$) in particular, were most common among the non-compliant samples of POP's. Significant residues of dioxins were found only in fish. The number of non-compliant cadmium samples was higher than that of lead ($p \leq 0.001$). The specific activity of artificial radionuclides (^{137}Cs and ^{90}Sr) in animal food products was significantly lower than the permissible limit.

Keywords: monitoring, organichlorine pesticides and PcB's, organophosphorus compounds, heavy metals, radionuclides, foods of animal origin.

B GRUPĖS TERŠALŲ (CHLORORGANINIŲ IR FOSFORO ORGANINIŲ PESTICIDŲ, POLICHLORINTŲ BIFENILŲ, DIOKSINŲ, SUNKIŲJŲ METALŲ IR RADIONUKLIDŲ) STEBĖSENOS LIETUVOS MAISTO ŽALIAVOSE 1999–2008 METAIS REZULTATŲ APŽVALGA

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Santrauka. Darbo tikslas buvo įvertinti Lietuvos maisto žaliavų užterštumą patvariaisiais organiniais teršalais (POT), sunkiaisiais metalais bei radionuklidais 1999–2008 metais. Maisto žaliavų užterštumo POT rezultatų analizė atlikta SPSS statistine programa; aprašomosios ir dispersinės analizės metodais analizuota metų, teršalų ir žaliavos įtaka tirtų ir viršijusių leistinas normas mėginių skaičiui. 1999–2003 metais maisto žaliavose nustatyti tik chlororganinių pesticidų ir polichlorintų bifenilų (PCB), o nuo 2004 metų – ir fosforo organinių pesticidų bei dioksinų likučių. Paukštienoje, žuvyse ir žvėrienoje 1999 ir 2000 metais rasta pesticidų, kurių koncentracija viršijo leistiną kiekį. Dažniausiai šių pesticidų rasta žvėrienoje ($p \leq 0{,}05$). Iš visų POT maiste leistiną koncentraciją dažniausiai viršijo chlororganiniai junginiai, tokie kaip DDT ($p \leq 0{,}05$). Dioksinų koncentracija, viršijanti leistiną, nustatyta tik žuvyse. Tyrimų laikotarpiu leistiną švino kiekį viršijančių mėginių skaičius buvo statistiškai patikimai didesnis nei kadmio. Didžiausia ($p \leq 0{,}05$) švino koncentracija nustatyta 1998 metais. Didesnė už leistiną švino ir kadmio koncentraciją nustatyta galvijų ir kiaulių kepenyse bei inkstuose, švino – ir piene. Dirbtinių radionuklidų (cezio, stroncio) savitasis aktyvumas tiriamuoju laikotarpiu gyvūniniuose maisto produktuose leistino kiekio neviršijo.

Raktažodžiai: stebėsena, sunkieji metalai, radionuklidai, POT, chlororganiniai pesticidai, fosforo organiniai junginiai, PCB, dioksinai, maisto žaliavos.

Introduction. European Council Directive 96/23/EC establishes control (monitoring) measures for veterinary drugs, contaminants, dyes, chemical elements, etc. in live animals and animal products. This Directive is implemented in the Lithuanian law by the “Rules on control of certain substances and residues in live animals and animal products” (Rules on monitoring 2003; Corrigendum of the rules 2008). The Directive obliges EU member states to monitor these substances in food producing animals and animal by-products: meat, milk, eggs and honey. This means that samples from live animals on farm and carcasses at slaughterhouses should be taken for testing. Directive 96/23/EC also obliges to perform National residue monitoring programme for the mentioned substances by creating annual monitoring plans. Directive 96/23/EC divides all residues into groups A and B. Group A includes prohibited substances as well as unauthorized substances. Group B includes veterinary drugs and contaminants. Group B represents extremely hazardous chemicals referred to as persistent organic pollutants (POPs), organophosphorus compounds, chemical elements (heavy metals), radionuclides and others. POPs are highly toxic, long-resistant substances; they spread away from the original source and accumulate in the environment and food chains, thus some of them with the negative feedback are strictly regulated. Organochlorine pesticides are highly resistant to chemical, thermal and biological degradation; therefore they are persistent in the nature. They are fat-soluble and can accumulate in animal fat and enter female milk and can cause various diseases and disorders (El-Shahawi et al., 2010). Therefore, since 1970 organochlorine pesticides actually have been out of use in Lithuania, yet so far, their degradation products are detectable in the environment. Instead, agriculture begins to use organophosphorus pesticides. They are less persistent, but are much more toxic to humans.

Heavy metals from Group B are most important for the monitoring. Heavy metals cause toxic effects when they accumulate in excess in the body outmatching the limits of physiological tolerance (Bryan and Langston, 1992). According to the seriousness of heavy metals to living organism they are located in the following sequence: Hg, As, Cu, Cd, Zn, Cr, Mn, Fe, Ti, and Pb. Carcinogenic and mutagenic effects depend on their concentrations, state of oxidation and can occur not immediately, but after a certain time span. Most of the heavy metals enter the human body via diet, less with the water and the air. Some of them accumulate in hair, skin, bones and internal organs.

The territory of Lithuania has been contaminated with artificial radionuclides in 1950–1960 when tests for nuclear weapons were carried out and after the accident at the Chernobyl nuclear power palnt in 1986. The long-living radionuclides (^{90}Sr and ^{137}Cs) are most dangerous. They migrate in the chain feed-animal-human and accumulate in the body. As a result, both neoplastic and non-neoplastic diseases can occur.

Taking into account the harm of Group B contaminants to the environment and human health, any information about their prevalence in the environment, biota, and foodstuffs is essential. Therefore food contamination monitoring program was implemented by the joint efforts of Lithuanian Ministry of Health and State Food and Veterinary Service (SFVS). The Lithuanian monitoring program has been performed by National Food and Veterinary Risk Assessment Institute (NFVRAI) for more than a decade. According to this program, organochlorine pesticide including PcBs, some organophosphorus compounds, dioxins, heavy metals, radionuclides and some other substances have been examined. The monitoring results were being reported annually by the Lithuanian SFVS to European Commission; however the scientific evaluation and overview of these results has never been done. Therefore this study aims to analyse the monitoring results of some Group B contaminants in Lithuania in the time frame 1999–2008.

Methods. Results of monitoring of certain Group B contaminants, which have been reported annually by Lithuanian SFVS to European Commission were used for the analysis. In accordance with the Rules on monitoring of certain substances and residues in animals and animal products, the “Plan for monitoring the residues in live animals and animal products” is drawn at the beginning of each year and this Plan serves as a base for regional inspectors taking samples. The samples are sent to and analysed at the NFVRAI Laboratory Department by validated methods (Table 1). This Department is assigned to carry out the functions of national reference laboratory (NRL) by the Order of Director of State Food and Veterinary Service (SFVS). The monitoring plan is drawn following the common EU calculations on previous year’s production numbers and the number of samples accordingly is divided to every county individually. SFVS presents the Plan for residues monitoring together with all the data and results of the previous year analyses to European Commission (EC) no later than 31st of March (Rules on monitoring 2003). Before 2001, the number of samples was calculated according to production levels of approved manufacturing plants as they were able to pay for the analyses. In 2002, the financing system of residues monitoring was amended and calculation of number of samples was bound to county production levels.

Residues of Group B contaminants are evaluated according to the Lithuanian Hygiene Standard HN 54-2008 "Food products. The maximum permitted pollutant and pesticide residues limits" and Commission Regulation (EC) No 1881/2006 of 19 December, 2006, setting maximum levels for certain contaminants in foodstuffs, and Commission Regulation (EC) No 629/2008 of 2 July 2008 amending Regulation (EC) No 1881/2006 setting maximum levels for certain contaminants in foodstuffs. The Maximum residue limits (MRL) for POPs and PcBs, and heavy metals are presented in Table 1. Radionuclides are evaluated according of Council Regulation (EC) No 733/2008 of 15 July 2008 on the conditions governing imports of agricultural products originating in third countries following the accident at the Chernobyl nuclear power station. Their MRL are shown in Table 1.

Table 1. **Methods of analyses and maximal residue limits (MLR) for Group B compounds in primary food products**

Group B compounds	Matrix (for meat)	Methods	MRL			
			Bovine, pork, sheep/goats, game, poultry meat	Eggs	Milk	Fish
Organochlorine pesticide and PcBs						
Heptachlor	Fat	GC (s), GC-MS (c) (accredited)	200 µg/kg	20 µg/kg	4 µg/kg	10 µg/kg
DDTs	Fat		1 000 µg/kg	50 µg/kg	40 µg/kg	
Alfa-HCH	Fat		200 µg/kg	20 µg/kg	4 µg/kg	
Beta - HCH	Fat		100 µg/kg	10 µg/kg	3 µg/kg	
Lindan	Fat		20 µg/kg	10 µg/kg	1 µg/kg	
Aldrin	Fat		200 µg/kg	20 µg/kg	6 µg/kg	
Dieldrin	Fat		200 µg/kg	20 µg/kg	6 µg/kg	
HCB	Fat		200 µg/kg	20 µg/kg	10 µg/kg	
PCBs	Fat		-	-	-	
Endrin	Fat		50 µg/kg	5 µg/kg	10 µg/kg	
Organophosphorus compounds						
Metidation	Muscle	GC (s), GC-MS (c) (accredited)	20 µg/kg	-	20 µg/kg	-
Metilchlorpirifos	Muscle		10 µg/kg	-	10 µg/kg	-
Diazinon	Muscle		50 µg/kg	-	10 µg/kg	-
Pirazofos	Muscle		20 µg/kg	-	20 µg/kg	-
Heavy metals						
Pb	Muscle	Z-ETA-AAS (c) (accredited)	0,10 mg/kg	-	0,02 mg/kg	0,3 mg/kg
	Kidney		0,5 mg/kg			
Cd	Muscle	Z-ETA-AAS (c) (accredited)	0,05 mg/kg	-	-	0,05 mg/kg
	Kidney		1,0 mg/kg			
Hg	Muscle	Cold steam AAS	-			0,5 mg/kg
Radionuclide						
Sr-90	Muscle	Radiochemical analysis, gamma spectrometry (accredited)	750 Bq/kg	750 Bq/kg	125 Bq/kg	750 Bq/kg
Cs-134/137	Muscle		600 Bq/kg	600 Bq/kg	370 Bq/kg	600 Bq/kg

Table 2. **Levels of significance of monitoring year, types of pollutant and product, and their interactions on the number of samples tested and number of non-compliant samples** (ns, factors were tested and excluded from the model due to P-level ≥ 0.05)

Factors and their interactions	Variables	
	Total number of samples tested	Number of non-compliant samples
Type of pollutant	0.0001	0.038
Type of product	0.001	0.0001
Year	0.0001	0.042
Year x Type of pollutant	0.001	ns
Year x Type of product	0.001	0.0001
Type of pollutant x Type of product	ns	0.022
Number of tested samples x Year	-	0.0001

SPSS statistical package was used to analyse monitoring data. Initial statistical evaluation of data was done using descriptive statistics. Factors having influence on the variables (number of samples taken and non-compliant samples (samples containing residues of contaminants above the MRL or ’, positive’) were evaluated using GLM univariate method. Year of analysis, type of pollutants, and type of food product were analysed and interaction of these factors was determined. Significance of difference between groups (p) was determined using LSD method of comparison. The difference was considered statistically significant when $p \leq 0.1$.

Results and discussion

Analysing the surveillance results of POPs residues in food, the influence of selected factors (the type of pollutant, year of monitoring, and product) and their interactions on the total number of samples tested and non-compliant samples are evident (Table 2).

Since 1998, only organochlorine pesticides and PCBs were foreseen to analyse. In 2004, organic phosphorus pesticide and dioxin residues were added to the group (Table 3). In total N=1933 samples were tested for POPs during the reviewed period; non-compliant pesticide residues were found only in 22 samples (1 % of samples tested). Samples with pesticide exceeding the permitted levels were found only in 1999 and 2000. Although the impact of year factor on the number of non-compliant samples was not very strong and the number of samples with exceeding MRL in food is more casual and often hard-tracked and explained, the decline in the number of non-compliant samples in Lithuania is obvious. We believe that this is related to implementation of relevant legislation on food contaminants monitoring and creation of control mechanisms. In some EU countries that have been carrying out a similar analysis, this trend is also observed (Yaktine, 2006). However occurrences of noticeable, alarming and requiring cross-border solutions non-compliant cases of some food pollution with POPs are observed during this period. Under the scrutinized observations of those cases it was identified that the causes have usually been associated with POPs contaminated pasture/fodder, or animal feed additives or protein additives imported from third countries (Kawser, 2010).

Table 3. **Total number of samples tested and number/percentage of non-compliant samples of pesticides, organophosphorus compounds and dioxins during the ten years monitoring period**

Year	Organochlorine pesticides and PCBs		Organophosphorus compounds		Dioxins	
	Number of samples tested (n)	Non-compliant samples, (n/%)	Number of samples tested (n)	Non-compliant samples (n)	Number of samples tested (n)	Non-compliant samples, (n/%)
1999 [b]	257 [g]	**1/0.4** [c]	-	-	-	-
2000 [c]	140	**21/15** [b-l]	-	-	-	-
2001 [d]	223 [g]	0 [c]	-	-	-	-
2002 [e]	219 [g]	0 [c]	-	-	-	-
2003 [f]	251	0 [c]	-	-	-	-
2004 [g]	263 [a,b,d,e,h]	0 [c]	38	0	3	0
2005 [h]	181 [g]	0 [c]	48	0	47	0
2006 [j]	141	0	44	0	1	0
2007 [k]	136	0	41	0	19	**2/10.5**
2008 [l]	122	0	38	0	5	**2/40**
Total	**1 933**	**22/1.1**	**209**	**0**	**72**	**4/5.6**

Explanation: differences between superscripts [a-l] are statistically significant ($p \leq 0.05$)

None of non-compliant organic phosphorus compound residues were found in food during the period reviewed; most likely due to their rapid degradation in the environment.

Monitoring analysis of the dioxins in foods of animal origin revealed that in high excess levels they were present only in fish samples (Table 4). This is also confirmed by other scientist (Huwe and Larsen, 2005). Analysis revealed that in recent years, the number of non-compliant for dioxins fish samples is high (Table 3). One of presumable causes is considerable contamination of Lithuanian waters, especially the waters of the Baltic Sea, and accumulation of dioxins due to the degradation of many of the 2nd World War ship vestiges (Emelyanov, 2007). High concentrations of these pollutants remain in the soil for quite a long time, and by rain or surface water drainage channels this polluted water flows into rivers and deposits on the bottom (Concha-Grañaa et al., 2006; Chopra, 2010); then with the fish food the pollutants get into the fish body and accumulate in adipose tissue (Eljarrat et al. 2002) throughout their lives. That is why the concentrations of pollutants in fish can be up to a million times higher than in the water. This means that fish with relatively high concentrations of pollutants may live in practically clean water. Another assumption is related to the sampling specifics: 97 percent of samples go for the analyses from the muscle tissue and only 3 percent from other parts of the carcass. As fish for the survey is taken whole, there is a chance for the more accurate determination of contamination. High positive correlation is found between the age and the amount of POP in the organism (Schecter et al., 2001). This explains the higher concentrations of these pollutants in fish targeting adults compared with relatively young fattening animals.

Table 4. **Number of samples tested and number of non-compliant samples of pesticides, organophosphorus compounds and dioxins by animals and type of products**

Pollutant	Dioxins		Organophosphorus compounds		Pesticides and PCBs	
Type of product	Number of samples tested	Number of non-compliant samples	Number of samples tested	Number of non-compliant samples	Number of samples tested	Number of non-compliant samples
Bovine [a]	2 [f]	0 [f]	50	0	3247 [b,g,h,k]	0 [l]
Swine [b]	2 [f]	0 [f]	68	0	2225	0 [l]
Sheep/goats	0 [f]	0 [f]	3	0	36	0 [l]
Horses [d]	0 [f]	0 [f]	-	-	0	0 [l]
Poultry [e]	3 [f]	0 [f]	-	-	714	2 [l]
Fish [f]	14 **[a-e, g-m]**	4 **[a-e, g-m]**	-	-	476	4 [l]
Milk [g]	3 [f]	0 [f]	43	0	2017	0 [l]
Eggs [h]	2 [f]	0 [f]	-	-	2295	0
Rabbits [j]	0 [f]	0 [f]	-	-	8	0 [l]
Farmed game [k]	0 [f]	0 [f]	-	-	1197	0 [l]
Game [l]	0 [f]	0 [f]	-	-	1359	20 **[a-k, m]**
Honey [m]	0 [f]	0 [f]	-	-	172	0 [l]
Total	**26**	**4**	**164**	**0**	**13746**	**26**

*Explanation: differences between superscripts [a-m] are statistically significant ($p \leq 0.05$)

Large accumulation of dioxins in foods of animal origin, particularly beef, dairy products, milk, poultry, fish, pork and eggs, has been reported in other EU countries during the reviewed period. In particular, a very high concentration of the contaminants of this kind was found in the ponds fish fed on feed of animal origin (Schecter et al., 2001).

The analysis of monitoring results by the product type revealed that majority tests for POPs were done in bovine meat, porcine, and eggs ($p \leq 0.05$, Table 4). Non-compliant samples were detected only in poultry, fish and game, most of them in game meat and organs ($p \leq 0.05$). Pesticide residues in game have been reported only in 2000, most samples (10 out of 30 surveyed) were contaminated with DDT ($p \leq 0.05$), far less with alpha-HCB and HCH. In wildlife the pesticide residues monitoring was intermittent and in 1999, 2002 and 2003 game samples were not examined. That is because the numbers of samples taken are proportionally set to number of food producing animals in the country that year; the production of game meat differs greatly among years in Lithuanian and generally is far less in amount than other foods. The significant game contamination by pesticides can be explained by their exceptional spread in nature and stability. We believe that regular spraying of organochlorine pesticides in forests is responsible for high accumulation of these pesticides and their breakdown products in forest vegetation and in the bodies of water. The other important cause - an inappropriate storage of pesticides in Lithuania after the collapse of the Soviet Union. This speculation confirms the quantities of pesticides exceeding the permitted levels in disaster locations – burnt pesticide warehouses. For example residues of DDT in coniferous forest soil samples near pesticide storage warehouse in Old Impilties village (Kretinga District) were found at a concentration of almost four times as high as the MRLs; this is also true about DDE, alpha-and gamma-HCH residues (Ministry of the Environment, 1999). Such increase in the quantity of pesticides in the soil leads to their higher levels in vegetation and water (Lutze et al., 2009), through which pesticides enter the wild animal tissue. The assumption is confirmed by the data of other scientists, proving that the wild animals, hunted in the areas of large chemical industry and chemical waste management companies, are usually contaminated with high concentrations of POPs (Dykuma and Freer, 2004).

Analysing the type of pollutant effect on the number of samples tested and non-compliant samples showed that organochlorine compound residues were found more frequently ($p \leq 0.05$); of which mostly DDT ($p \leq 0.05$, Table 5). DDT was detected only in 1999 and 2000. In 1999, 3 % poultry meat samples of total number tested were positive for DDT; in 2000 – 7 % of game samples. It is known that the concentration of DDT in the soil is directly proportional to its concentration in foods (Passuello, 2010); this explains the largest quantities of contaminated game. Lithuania banned DDT use in 1970. Since then, DDT remains in Lithuanian environment due to slow degradation. It also enters the environment from the current use in other parts of the world. According the global studies, POPs is one of the world's most widespread pollutants. It is almost undetectable only in the sub-Sahara Africa, Pacific Islands and Antarctica (Regionally Based Assessment of Persistent Toxic Substances, 2003). Most contaminated are highly developed agricultural countries. Analyses of European and USA monitoring data on DDT pollutant (FAO/WHO 1993, 1996) showed that DDT residues are found in most of animal production, especially in beef, pork, lamb and cheeses.

Table 5. **Number of samples tested and number/percentage of non-compliant samples by the type of pollutant**

Pollutant	Samples tested (n)	Non-compliant samples (n)	Non-compliant samples (%)
Organochlorine pesticides *****			
Heptachlor / heptachlor epoxide ^{a}	1346^{j-o}	**2** ^{b}	**0,13**
DDTs ^{b}	1534^{j-o}	**11** $^{a, c-x}$	**0,72**
α – HCH ^{c}	1534^{j-o}	**4** ^{b}	**0,26**
β – HCH ^{d}	1495^{j-o}	0	0
Aldrin ^{f}	1200^{j-o}	0	0
Dieldrin ^{g}	1198^{j-o}	0	0
HCB ^{h}	1160^{j-o}	**4** ^{b}	**0,34**
Lindan ^{p}	521^{j-o}	0	0
Endrin ^{r}	521^{j-o}	0	0
PCB/(PCB 28, 52, 101, 138, 153, 180)	527	0	0
Total		**20***	
Organophosphorus compounds *****			
Metidation ^{j}	$181^{a-h, p-x}$	0	0
Methylchlorpyriphos ^{k}	$181^{a-h, p-x}$	0	0
Diazinon ^{l}	$181^{a-h, p-x}$	0	0
Pyrazophos ^{m}	$181^{a-h, p-x}$	0	0
Total	**231**	**0**	**0**
Dioxins ^{i} *	72	**4** ^{b}*****	**5,60**

Explanation: differences between superscripts $^{a-m}$ and asterisks * are statistically significant (p≤0.05)

Non-compliant alpha-HCH was determined in game samples in 2000. It is believed that it resulted from intensive use of HCH pesticides until 1997. The former Soviet Union was the second most important POP user at that time. Enormous quantities of HCH pesticide consumption is likely to be one of the reasons why in the Lithuanian environment and foods of animal origin this pesticide is still found. Other countries show similar survey results: the most polluted samples have been detected in 2002; they are mostly contaminated with DDT, HCH and dieldrin (Regionally Based Assessment of Persistent Toxic Substances, 2003). Speaking about HCH pollution, alpha-HCH is being detected by 50 % more often than the beta and gamma type, usually in sheep/goat (due to the treatment for ectoparasites) and pork samples (Covaci et al., 2004; Bedi et al., 2005).

Monitoring results for heavy metal residues in foods are presented in Table 6. During the reviewing period, we found significantly more non-compliant cadmium samples than those of lead. The same trend prevails in the European Union countries (Commission Staff Working Paper, 1999–2008). A significant increase of both metals in foods monitored was determined in 2006. We can only speculate on the causes of such a distribution of non-compliant samples among the years reviewed: it could be affected by weather conditions and/or soil granulometric composition where fodder was growed (Zitkevičius et al., 2003).

Table 6. **Dynamics of heavy metal residues in raw materials during the ten years period**

	Lead		Cadmium		Mercury	
	Samples tested (n)	Non-compliant samples (n)	Samples tested (n)	Non-compliant samples (n)	Samples tested (n)	Non-compliant samples (n)
1999	149	0	148	0	-	-
2000	60	0	58	0	-	-
2001	80	1^{B}	79	1^{B}	-	-
2002	200	1^{B}	200	9^{A}	-	-
2003	308	2^{B}	306	4^{B}	-	-
2004	284	2^{B}	252	2^{B}	1	0
2005	199	0	192	2^{B}	-	-
2006	205	6^{A}	207	11^{A}	9	0
2007	128	3^{B}	345	4^{B}	5	0
2008	89	0	89	1^{B}	6	0
Total	**1 956**	**15****	1 939	34**	20	0

Explanation: differences between superscripts $^{A-B}$ (p≤0.05) and asterisks ** (p≤0.001) are statistically significant

Assessing the distribution of non-compliant samples among the foods monitored we found that the number of non-compliant game samples was considerably higher (Table 7). This can be explained by the eating grounds of wild animals, which have a significant effect on the accumulation of heavy metals in tissues of animals (Grove and Henny, 2008). Wild animals ration is constituted of plants of different development phases, by which, as well as air pollution, heavy metals enter the animal's body. Plant contamination with heavy metals depends on environmental pollution – under and over plant canopy absorption. It is identified that in different geographical areas soil and atmospheric contamination by chemical pollutants differs greatly. For example, the environmental monitoring data for lead in sandy areas of Vilnius District (the southern part of Lithuania) revealed concentrations not exceeding 10 mg/kg, and in clay-loam soils of some areas of Kelmė, Šilutė and Prienai districts (the western part of Lithuania) they were 22–25 mg/kg (MRL for lead in soil is 100 mg/kg). Some authors indicate that heavy metals concentrations in soils exceed the MLR only near major sources of local pollution (such as military grounds, etc.) and highways (Pignata, 2007; Ozdemir, 2009). Accordingly, increased forage pollution (various chemicals) and habitat induces greater accumulation of chemical elements in animal tissues.

Table 7. **Results of lead and cadmium residues monitoring by the type of product**

Type of products	Lead		Cadmium		Total number of non-compliant samples (n/%)
	Samples tested (n)	Non-compliant samples (n/%)	Samples tested (n)	Non-compliant samples (n/%)	
Beef	336	0	288	**19/0.59**	**19/6.59** ^{B}
Pork	286	0	237	**2/0.84**	**2/0.84** ^{B}
Sheep/goats	7	0	4	0	0
Game	351	**15/4.3**	353	**13/3.7**	**28/8.0** ^{A}
Poultry	99	0	89	0	0
Fish	43	0	36	0	0
Milk	276	0	256	0	0
Eggs	307	0	226	0	0
Honey	38	0	32	0	0
Rabbits	9	0	6	0	0

Explanation: differences between the superscripts $^{A\text{-}B}$ are statistically significant when $p \leq 0.001$

In 2006, mercury was started to be analysed in Lithuania; food materials were free of mercury residues in the period analysed. In the EU countries and in the USA this contaminant is found in foods each year (Jenkins, 2007). The maximum of non-compliant numbers in 2007 was 23 samples with traces of mercury which were found in porcine meat produced in Germany.

Since 1999, tests for **radioactive nuclides** (caesium and strontium) were done only for main food commodities (bovine and porcine meat, poultry, eggs, milk and honey); then gradually the spectrum was expanded for game (2000), rabbit meat (2003), and fish products (2005). A maximum of samples was tested in 2003. The most often strontium concentrations were measured in bovine meat and milk, and those of caesium in bovine and game meat, and eggs. None of the non-compliant samples was found in foods during the reviewed period. We suppose that these results could be affected by currently used sampling method of monitoring data. According to Ladygienė (2001), in order to observe the evolution of artificial radionuclide concentration in the Ignalina NPP exposure zone samples should be taken regularly in the same area for a long time. There are nine sampling sites in 5 regions: Vilnius, Kaunas, Klaipėda, Šiauliai, Panevėžys and 4 of the Ignalina nuclear power plant zone - Zarasai, Ignalina, Utena and Švenčioniai districts.

Conclusions

1. During the 10 years period, cases of non-compliance of POPs in food commodities (mostly in fish and game) in Lithuania were sporadic, mainly determined by their accidental access to food materials.

2. During the period analysed, only pesticides DDT, alpha-HCH, HCB and heptachlor were found. DDT residues in primary foods was detected more frequently ($p \leq 0.05$) than other pesticides. In most cases ($p \leq 0.05$) contamination concerned game samples.

3. During the period of 1999–2008, the number of non-compliant cadmium samples was significantly higher than that of lead. Significantly more non-compliant samples of those elements were found in game.

4. Testing for organophosphorus compounds and dioxins residues in food products was introduced in Lithuania in 2004. Only non-compliant dioxin residues were found in samples of aquatic animals in 2007–2008.

5. In 1999–2008, the activity of artificial radionuclides (^{137}Cs and ^{90}Sr) in animal food products was significantly lower than permitted.

6. The comparison of Lithuanian monitoring data with the other EU countries is challenging and difficult,

because there are no papers published analysing such data scientifically. Assessment of differences/similarities of Lithuanian results and those of other countries is also complicated, because the monitoring is not carried out in all countries, and most studies are being done only in cases of outbreaks occurrence.

Recommendations

1. Analysis of the results of monitoring data in Lithuania showed that it is reasonable to plan numbers of raw food samples for monitoring not only on the scale of production but also according to their contamination. Results of our survey show that in future monitoring plans particular notice should be taken of DDT and dioxins. An introduction of additional non-statutory surveillance scheme in Lithuania would be an option for strengthening the residue control: additional samples calculated according to the samples foreseen in the Plan might be taken in possible critical points of local and imported production chains.

2. Monitoring of pollutants in the raw foods in the European Union has been carried out for a long time, but there are not enough scientific assessment of the results and papers published. We recommend for relevant scientists to perform a similar analysis of the scientific evaluation of the dynamics of pollutants that can be useful to ascertain present and predict future trends. Without such analysis, monitoring of raw food contamination is just one of the EU's costly policy formalities. Analysis of the trend can provide direction for further monitoring, indicate the areas where monitoring should be strengthened, facilitate detailed planning of research in problem areas and the provide measures for consumer's information and protection.

Literature

1. Bryan, G. W., Langston, W. J. Bioavailability, accumulation and effects of heavy metals in sediments with special reference to United Kingdom estuaries: a review. Environmental Pollution, 1992. V. 76(2), P. 89–131.

2. Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. OJ L 364, 20.12.2006, P. 5–24.

3. Bedi, J. S., Gill, J. P. S., Aulakh, R. S., Joia, B. S., Sharma, J. K. Contamination levels of DDT and HCH residues in different goat tissues. Indian Journal of Animal Sciences, 2005. V. 75(1), P. 11–13.

4. Concha-Grañaa, E., Turnes-Caroua, M., Muniategui-Lorenzo, S., López-Mahíaa, P., Prada-Rodrígueza, D., Fernández-Fernández, E. Evaluation of HCH isomers and metabolites in soils, leachates, river water and sediments of a highly contaminated area. Chemosphere, 2006. V. 64(4), P. 588–94.

5. Chopra, A. K., Sharma, M.K., Chamoli S. Bioaccumulation of organochlorine pesticides in aquatic system—an overview. Environmental Monitoring and Assessment, 2010. DOI: 10.1007/s10661-010-1433-4.

6. Commission Staff Working Paper on the Implementation of National Residue Monitoring Plans in the Member States, 1998-2008. Council Directive 96/23/EC http://ec.europa.eu/food/food/chemicalsafety/residues/control_en.htm Accessed 12 May 2010.

7. Covaci, A., Gheorghe, A., Schepens, P. Distribution of organochlorine pesticides, polychlorinated biphenyls and α-HCH enantiomers in pork tissues. Chemosphere, 2004. V. 56(8), P. 757–766.

8. Dykuma, L.D., Freer, R. Dioxins in Wild Game Taken from the Tittabawassee River Floodplain South of Midland, Midland and Saginaw Counties, Michigan. The Dow Wild Game Study, 2004.

9. Emelyanov, E.M. The geochemical and geoecological situation in the areas of Skagerrak and Baltic Sea where chemical munition was dummped. Geologija, 2007. V. 60, P. 59–60.

10. Eljarrat, E, Caixach, J, Rivera, J. Determination of PCDDs and PCDFs in different animal feed ingredients. Chemosphere, 2002. V. 46, P. 1403–1407.

11. FAO/WHO joint meeting on pesticides residues. Geneva. 20–29 September, 1993.

12. Grove, R.A., Henny, C.J Environmental contaminants in male river otters from Oregon and Washington, USA, 1994–1999. Environmental Monitoring and Assessment, 2008. V. 145(1–3), P. 49–73.

13. Huwe, J.K., Larsen, G.L. Polychlorinated dioxins, furans and biphenyls, and polybrominated diphenyl ethers in a U.S. meat market basket and estimates of dietary intake. Environmental Science and Technology, 2005. V. 39, P. 5606–5611.

14. Jenkins, D.G. A Critical Analysis of Illinois' Fish Mercury Monitoring Program, 1974–1998. Environmental Monitoring and Assessment, 2007. V. 131 (1-3), P. 177-184.

15. Yaktine, A.L, Harrison, G.G., Lawrence, R.S. Reducing Exposure to Dioxins and Related Compounds through Foods in the Next Generation. Nutrition reviews, 2006, V. 64(9), P. 403–409.

16. Kawser, A., Yousuf, M., Rezaul, H., Pulakesh, M. Heavy metal concentrations in some macrobenthic fauna of the Sundarbans mangrove forest, south west coast of Bangladesh. Environmental monitoring and assessment, 2010. DOI:10.1007/s10661-010-1651-9.

17. El-Shahawi, M. S., A. Hamza, A. S. Bashammakh and W. T. Al-Saggaf. Anoverview on the accumulation, distribution, transformations, toxicity and analytical methods for the monitoring of persistent organic pollutants. Talanta, 2010. V. 80(5),

P. 1587–97.

18. Lutze, J., Derrick, J., Korth, W., MacLachlan, D.J. Monitoring of pesticides and veterinary drugs in Australian cattle: verification of the residue. Food Additives and Contaminants: Part B, 2009. V. 2(2), P. 99–111.

19. Ozdemir, S., Dundar, M.S., Sengorur, B., Senol, A.S. Lead and zinc content of cows' milk affected by varying traffic density. International Journal of Environment and Pollution. 2009. V. 36 (4), P. 411–417.

20. Passuello, A., Mari, M., Nadal, M., Schuhmacher, M., Domingo, J.L. POP accumulation in the food chain: Integrated risk model for sewage sludge application in agricultural soils. Environmental international, 2010. V. 36(6), P. 577–583.

21. Pignata, M.L., Pla, R.R., Jasan, R.C., Martinez, M.S., Rodriguez, J.H., Wannaz, E.D., Gudino, G.L., Carreras, H.A., Gonzalez, C.M. Distribution of atmospheric trace elements and assessment of air quality in Argentina employing the lichen, Ramalina celastri, as a passive biomonitor: detection of air pollution emission sources. International Journal of Environment and Health, 2007. V. 1(1), P. 29 – 46.

22. Regionally Based Assessment of Persistent Toxic Substances. The collection, assembly and evaluation of data on sources, environmental levels and impacts of persistent toxic substances across the globe. 2003.

23. Schecter, A., Cramer, P., Boggess, K., Stanley, J., Päpke, O., Olson, J., Silver, A., Schmitz, M. Intake Of Dioxins and Related Compounds from Food in the U.S. Population. Journal of Toxicology and Environmental Health, 2001. V. 63, P. 1–18.

24. Zitkevicius, V., Savickiene, N., Abdrachmanovas, O., Ryselis, S., Masteiková, R., Chalupova, Z., Dagilyte, A., Baranauskas, A. Estimation of maximum acceptable concentration of lead and cadmium in plants and their medicinal preparations. Medicina (Kaunas, Lithuania), 2003. V. 39(2), P. 17–121.

Received 15 February 2012
Accepted 11 January 2013