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PESTICIDE RESIDUES CONTAMINATION OF MILK AND DAIRY PRODUCTS. A CASE STUDY: BACAU DISTRICT AREA, ROMANIA

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Abstract. Organochlorine pesticides (OCPs) are highly toxic, persistent organic pollutants (POPs) with adverse effects to the environment and human health. The main way of human exposure to the OCP contamination is through food, especially the food products of animal origin. The aims of this study were to determine the presence and concentration of organochlorine pesticide residues in milk and dairy products samples collected from Bacau district area and to assess human exposure to OCPs through the consumption of these products. A total number of 54 samples of milk and dairy products were analysed for their residual content of hexaclorocyclohexane isomers (HCHs) and dichlorodiphenyltrichloroethane (DDT) and its analogues. The levels of OCP residues were determined by gas chromatography – mass spectrometry (GC/MS) method. The results indicated that all the analysed samples were contaminated with α-HCH, β-HCH and γ-HCH, respectively. In all the analysed samples, DDT and its analogues were non-detectable. The presence of γ-HCH (Lindane) was detected in the range of 0.0042 to 0.2124 (mg kg^{-1} fat) for raw cow milk, of 0.0028 to 0.1408 (mg kg^{-1} fat) for pasteurised cow milk and of 0.0042 to 0.2682 (mg kg^{-1} fat) for sour cream. It could be concluded that organochlorine pesticide residues were detected in raw cow milk and dairy products as they were persistent in the environment due to their slow decomposition rate, long half-life and high stability in the environment. In most cases, the values of detected organochlorine pesticide residues exceeded the maximum residue levels provided by Romanian and international regulations. This study represents an important step towards a more comprehensive understanding of the human health risks associated with OCP exposure via milk and dairy product consumption in Romania.

Keywords: organochlorine pesticide residues, milk, dairy products, food safety.

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AIMS AND BACKGROUND

Milk is considered to be the most balanced food ever found in nature, which contains most nutrients. It is eaten by people of all ages and nationalities, and the quantity varies according to the eating habits and to its availability. Milk and dairy products are an important part of the daily diet of babies, children, elderly and sick people. These products are also the only source of animal protein in the diet of many vegetarians, whose number is continuously increasing all over the world. Milk and the dairy products implicitly can be contaminated with different chemicals in different stages of its production and storage. This contamination can be due to the animal food (the feed) as well as to other parts of the diet such as the water[1–3]. The following types of chemical contaminants can found in milk: pesticide residues, other persistent organic pollutants (POPs) such as PCBs, PCDDs, PCDFs, etc., heavy metals, radionuclides, veterinary drugs and antibiotics, aflatoxins and mycotoxins, nitrites and nitrates, detergents and disinfectants[4–11].

Organochlorine pesticides (OCPs) are highly toxic, persistent organic pollutants (POPs) with adverse effects to the environment and human health. These compounds are considered to have low acute toxicity, but they have high potential of chronic toxicity compared to other types of pesticides such as the organophosphorus pesticides[1,11]. Organochlorine pesticides (OCPs), especially dichloro-diphenyl-trichloroethane (DDT) and hexachloro-cyclohexane (HCHs), together with the cyclodienes such as aldrin, dieldrin, chlordane and heptachlor were found in milk and in dairy products during the last decades and they were mentioned in different studies[12–16]. OCPs, considered to be more persistent, were used on large scale all over the world, especially at the tropics, in order to stimulate the agricultural production by controlling the pests as well as in health to control the vectors that cause diseases such as malaria[8,17]. OCPs are highly stable under most environmental conditions, have long persistence and tendency to accumulate in body tissues and are liposoluble, thus leading to their bioaccumulation through the food chain[12]. The fact that they are liposoluble allows the OCPs to accumulate in the body tissues were they get into a state of balance and bioconcentration; thus the tissues rich in lipids act as OCP deposits. Due to this fact, the concentrations of OCPs in the human and animal body decrease very slowly, even after the contamination sources are eliminated[11,12,18].

Health risks of OCPs. The OCPs have different types and degrees of toxicity. As their residues can be accumulated in the fat tissues which make up the vital organs such as the thyroid, the heart, the kidneys, the liver, mammary gland and the testicles, they can be a major risk for the human health. Many studies showed their effects on the human health, starting with their effects on the cardiovascular and the respiratory systems until genotoxicity[17,19,20]. These residues can also be transferred to the fetus through the umbilical cord and to the baby through breast-feeding[18,21]. Some researches connected the exposure to these pesticides to a high risk of human cancer and some of these pesticides were found to be carcinogenic when tested on animals. Many of the OCPs are now recognised as potentially disturbing on the human endocrine system, even in case of low exposure[22,23]. The International Agency for Research on Cancer (IARC) classified compounds such as dichlorodiphenyltrichlroethane (DDT), mirex, toxaphene, hexachlorobenzene (HCB) or hexachlorocyclohexane (HCH) as possibly carcinogenic to humans (group 2B)[24]. The mechanism on which the toxicity of OCPs is based involves inducing enzymatic activity by means of the free radicals which affect the immune response, the neurologic system, the lipid metabolism and the transport of vitamins and glucose in the blood. It should also be mentioned the fact that certain OCPs have mutagenic, teratogenic and carcinogenic effect not only on humans but also on other biotic communities with different levels of sensitivity[11,25–27]. Numerous studies indicated the affinity of organo-chlorinated compounds for fats, showing evidence of increasing contamination through the food chain[3,5,11–13].

The main way of human exposure to the OCP contamination is through food, especially the food products of animal origin which are rich in fat such as pork, fish, milk and dairy products due to for many people, these types of food represent an important part of the daily diet[28–30]. Previous studies proved that food of animal origin is responsible for over 90% of the average human intake of chlorinated organic compounds[11,12].

According to the present regulations, the use of OCPs or of mixtures including OCPs are no longer allowed in the European Union. Products containing γ-HCH were banned starting with January 1st 2007 and Endosulfan was banned starting with January 1st 2008. In Romania pesticide residues are strictly regulated by sanitary veterinary regulations specific for food of animal and vegetal origin, both from our country and imported, according to the European regulations in this field. When it comes to milk and dairy products, the maximum residue levels specific for OCP residues in these products are mentioned[31,32].

Pesticides get into the vegetables directly through the phytosanitary treatments applied, as well as indirectly, being taken from the water and the soil, wherefrom they contaminate the food of animal origin through the trophic chain. Milk amounts high quantities of OCPs because mammary gland is one of the ways of eliminating these toxic compounds from the animal body. Due to the liposolubility of OCPs, their concentration is higher in sour cream, cheese and especially in butter, the residue level also depending on the age of the animal[33].

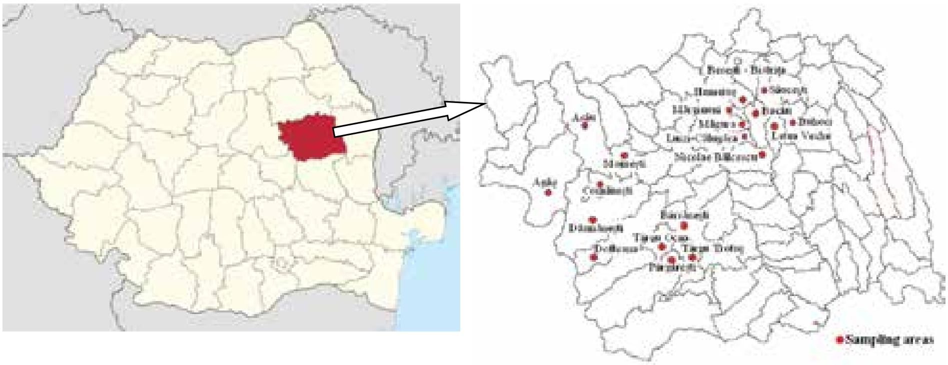
Since OCPs have been used over a long period of time in our country, and since these compounds are very persistent in the environment, they have to be continuously monitored. In Romania, in Bacau County, there are no data available on the levels of OCP residues in milk and dairy products coming from the small producers or independent producers, which constitute an important part of the diet of many adults and children.

The aims of this study were to determine the presence and concentration of organochlorine pesticide residues in milk and dairy products samples collected from Bacau district area and to assess human exposure to OCPs through the consumption of these products. The experimental research had as objectives to determine the physicochemical parameters for milk and dairy products samples, to determine the DDT (dichloro-diphenyl-trichloroethane), HCHs (hexachloro-cyclohexane) residues in raw cow milk, pasteurised cow milk and dairy product samples as well as to quantify the levels of OCP residues in milk and dairy products from Bacau district area and put forward some improvement measures for reducing further contamination.

EXPERIMENTAL

Sample collection. A total number of 54 samples were collected, 18 of which were raw cow milk, 18 others were pasteurised cow milk with 1.80% fat content and 18 were sour cream with 20% fat content. Fresh cow milk, pasteurised milk and sour cream samples were collected from several farms and dairy producers located in Bacau County over a period of three months (February, March, April 2014). The samples were collected from three areas in Bacau County: A1 – the area including the villages around Bacau city, A2 – the area including the villages around Comanesti town, A3 – the area including the villages around Targu Ocna town, respectively. When selecting these areas, were taken into consideration the number of animal farmers and the number of small dairy producers, the difference in feed in these areas and the fact that Comanesti and Targu Ocna areas are close to on of the greatest OCP producers in Romania up to 1990 – the Chemical plant located in Onesti.

Standards and reagents. The standards with 98–99% analytical purity for the types of OCPs analysed were purchased from Sigma-Aldrich. All the reagents used in the determinations had analytical purity and were purchased from Merck Company.



Map of Romania highlighting Bacau County

Map of the Bacau County that highlighting the sampling areas

Fig. 1. Location of the study area

Physicochemical analysis. All the samples were collected, preserved and analysed for relevant physicochemical parameters according to internationally accepted procedures and standard methods[1,4,10,15,18,34].

The chemical parameters (fat, protein, total dry matter and lactose content) of the collected samples were determined by a Fourier transform infrared spectroscopy (FTIR) method, using the Lactoscope FTIR, Delta Instruments, model 2048 device.

The content of non-fat dry matter was determined by the difference between the content of total dry matter and the fat content. The freezing point was determined by milk cryoscopy method using CryoStar I device. The acidity and density were determined according to the methods described in the AOAC (Ref. 34).

Organochlorine pesticide residues analysis. The OCP residues in milk and dairy products were analysed following the analytical procedures described by Avancini[11] and Salem[12].

The concentrations of OCP residues in the samples were determined by a Gas chromatography – mass spectrometry (GC/MS) method using VARIAN model CP-3800 GC device coupled with VARIAN model 320-MS device[35]. The pesticides were identified with retention time and specific ions and were quantified using the external standard method.

The values of the OCP residues determined in this study were compared with the maximum residue levels (MRL) provided by the Romanian and international regulations[31,32].

RESULTS AND DISCUSSION

Physicochemical composition. The physicochemical composition of the milk and dairy product samples analysed is presented in Table 1.

Table 1. Physicochemical parameters of the milk and dairy product samples collected from three areas of Bacau County

Sample I.D. *n* = 6	Fat [R] (%)	Protein[R] (%)	Total dry matter[R] (%)	Non-fat dry Matter[R] (%)	Lactose [R] (%)	Density [R] (g/cm3	Acidity[R] (°Th)	Freezing point[R] (°C)
RCM-A1	3.46–3.62	3.22–3.54	12.87–13.51	9.36–9.89	4.82–4.98	1.028–1.032	17.2–17.5	–0.520–0.530
RCM-A2	3.32–3.72	3.18–3.48	12.78–13.82	9.46–10.10	4.58–4.86	1.029–1.032	17.6–18.0	–0.512–0.524
RCM-A3	3.44–3.81	3.28–3.56	12.96–14.12	9.52–10.31	4.28–4.96	1.028–1.032	17.1–17.5	–0.515–0.531
PCM-A1	1.80	3.25–3.42	10.98–11.46	9.12–9.66	4.34–4.62	1.030–1.032	17.5–18.0	–0.494–0.510
PCM-A1	1.80	3.35–3.52	11.28–11.86	9.48–9.82	4.59–4.78	1.029–1.033	17.2–17.5	–0.484–0.505
PCM-A1	1.80	3.26–3.48	11.02–11.56	9.25–9.76	4.22–4.54	1.028–1.032	17.4–17.8	–0.495–0.512
SC-A1	20.0	2.32–2.60	24.69–25.48	–			55.0–58.0	
SC-A1	20.0	2.27–2.58	24.36–25.42	–			50.0–55.0	
SC-A1	20.0	2.23–2.43	24.32–25.12	–			52.0–55.0	

Sample I.D. – sample identification; *n* – number of samples/each area; [R] – range (the minimum and maximum values are presented in the table); A1 – Bacau area; A2 – Comanesti area; A3 – Targu Ocna area; RCM – raw cow milk, PCM – pasteurised cow milk, SC – sour cream; °Th – Thorner degrees: obtained by titrating 100 ml of milk thinned with 2 parts distilled water, with 0.1 N NaOH, using phenolphthalein as an indicator. The acidity expressed in Thorner degrees represents the number of ml of 0.1 N NaOH solution required to neutralise 100 ml of milk.

Assessment of physicochemical parameters. The values for the fat content were between 3.32 and 3.81% in the raw cow milk samples. This difference in percentage for the fat content may be due to the difference in feeding, management practices and breed of the animals. The obtained values for the fat content are around the standard content required, 3.50% respectively.

The total dry matter content is one of the parameters used to assess milk quality. The concentration range of the total dry matter is from 12.78 to 14.20%. These results showed that the values of total dry matter recorded are within the range required by the standards, between 12.50 and 14.50%, respectively.

The main sugar in cow milk is lactose. The raw cow milk samples contain lactose in amounts of 4.28–4.98%. The values of lactose content are around the standard content required, between 4.6 and 4.7%, respectively.

The freezing point represents one of the parameters used to control milk falsification. The variability limits imposed by the standards for these parameters are between –0.520 and –0.550°C. The values of freezing point are around the standard value required, between –0.512 and –0.5310°C, respectively.

Pesticide residues. The results achieved for the OCP residues in the samples analysed are presented in Tables 2–4.

Assessment of OCPs contamination. Hexachlorocyclohexane (HCH) is used against sucking and biting pests. As smoke, it is used for controlling the pests in grain deposits while as dust it is used to control various soil pests such as flea, beetles and flies. It is well known that hexachlorocyclohexane is a mixture of eight isomers, the one with the strongest insecticide properties being the γ isomer. Five out of these eight isomers are found in the fresh products (α, β, γ, δ, ε) (Ref. 13).

The presence of γ-HCH (Llndane) was detected in 100% of the analysed samples in the range of 0.0042 to 0.2124 (mg kg^{-1} fat) for raw cow milk, of 0.0028 to 0.1408 (mg kg^{-1} fat) for pasteurized cow milk and of 0.0042 to 0.2682 (mg kg^{-1} fat) for sour cream.

The levels of γ-HCH residues in raw cow milk samples were higher than those reported by Radzyminska et al.[4] in milk samples in Poland (< 0.001–0.060 mg kg^{-1} fat) and higher than those reported by Shaker and Esharkawy[5] in raw buffalo milk samples in Egypt (0.180–0.192 mg kg^{-1} fat). The large scale use and the great resistance of lindane to biological and chemical degradation in aerobic conditions can be one of the reasons for its presence in all the samples analysed. The presence of lindane above its maximum residue level in about 78, 50 and 66% of the RCM, PCM, SC samples is a cause of serious concern.

The β-isomer of HCH is the most persistent among the HCH isomers in the environment. Due to the fact that this isomer has longer half life in fatty components, it is eliminated from the human body more slowly than lindane. The obtained results showed that the concentrations of β-HCH were in the range of 0.0018 to 0.0255 (mg kg^{-1} fat) for raw cow milk, of 0.0017 to 0.0152 (mg kg^{-1} fat) for pasteurised cow milk and of 0.0016 to 0.0325 (mg kg^{-1} fat) for sour cream. In about 72, 38 and 55% of the RCM, PCM and SC samples, the presence of β-HCH above its maximum residue level was noticed.

Table 2. Levels of OCP residues in the raw cow milk samples collected in the three areas of Bacau County

Pesticide	MRL	Area					
		A1 – Bacau area (villages near Bacau) $n = 6$, RCM		A2 Comanesti area $n = 6$, RCM		A3 Targu Ocna area $n = 6$, RCM	
		range (mg kg^{-1} fat)	mean ± sd	range (mg kg^{-1} fat)	mean ± sd	range (mg kg^{-1} fat)	mean ± sd
α-HCH	0.004[a]	0.0024–0.0112	0.0057 ± 0.0038	0.0028–0.0142	0.0072 ± 0.0045	0.0031–0.0242	0.0095 ± 0.0082
β-HCH	0.003[a]	0.0024–0.0140	0.0062 ± 0.0043	0.0018–0.0171	0.0072 ± 0.0059	0.0028–0.0255	0.0153 ± 0.0124
γ-HCH	0.010[b]	0.0042–0.1212	0.0544 ± 0.0474	0.0078–0.1682	0.0760 ± 0.0585	0.0049–0.2124	0.1062 ± 0.0716
δ -HCH	0.004[a]	n.d.		n.d.		n.d.	
∑-HCH	–	0.0090–0.1464	0.0663 ± 0.0555	0.0124–0.1995	0.0904 ± 0.0689	0.0108–0.2621	0.1310 ± 0.0922
∑-DDT	0.040[a]	n.d.		n.d.		n.d.	

RCM – raw cow milk; n – number of samples/each area; n.d. – not detected; s.d. – standard deviation; MRL – maximum residue level; [a] – MRL according to EC Regulation No 396/2005 (Ref. 31); [b] – MRL according to EC Regulation No 149/2008 (Ref. 32).

Table 3. Levels of OCP residues in the pasteurized cow milk samples with 1.80 % fat content collected in the three areas of Bacau County

Pesticide	MRL	Area					
		A1 – Bacau area (villages near Bacau) $n = 6$, PCM		A2 Comanesti area $n = 6$, PCM		A3 Targu Ocna area $n = 6$, PCM	
		range (mg kg^{-1} fat)	mean ± sd	range (mg kg^{-1} fat)	mean ± sd	range (mg kg^{-1} fat)	mean ± sd
α-HCH	0.004[a]	0.0017–0.0098	0.0043 ± 0.0029	0.0022–0.0122	0.0050 ± 0.0037	0.0025–0.0142	0.0057 ± 0.0045
β-HCH	0.003[a]	0.0018–0.0088	0.0041 ± 0.0027	0.0022–0.0101	0.0056 ± 0.0034	0.0017–0.0152	0.0068 ± 0.0054
γ-HCH	0.010[b]	0.0034–0.0898	0.0380 ± 0.0368	0.0048–0.1162	0.0521 ± 0.0503	0.0028–0.1408	0.0684 ± 0.0674
δ -HCH	0.004[a]	n.d.		n.d.		n.d.	
∑-HCH	–	0.0069–0.1084	0.0464 ± 0.0424	0.0092–0.1385	0.0627 ± 0.0574	0.0070–0.1702	0.0866 ± 0.0773
∑-DDT	0.040[a]	n.d		n.d		n.d	

PCM – pasteurised cow milk with 1.8% fat content; n – number of samples/each area; n.d. – not detected; s.d.- standard deviation; MRL – maximum residue levelм [a] – MRL according to EC Regulation No 396/2005 (Ref. 31); [b] – MRL according to EC Regulation No 149/2008 (Ref. 32).

Table 4. Levels of OCP residues in the sour cream samples with 20 % fat content collected in the three areas of Bacau County

Pesticide	MRL	Area					
		A1 – Bacau area (villages near Bacau) $n = 6$, SC		A2 – Comanesti area $n = 6$, SC		A3 – Targu Ocna area $n = 6$, SC	
		range (mg kg^{-1} fat)	mean ± sd	range (mg kg^{-1} fat)	mean ± sd	range (mg kg^{-1} fat)	mean ± sd
α-HCH	0.004[a]	0.0029 – 0.0124	0.0055 ± 0.0036	0.0022 – 0.0158	0.0068 ± 0.0061	0.0028 – 0.0189	0.0094 ± 0.0069
β-HCH	0.003[a]	0.0016 – 0.0192	0.0092 ± 0.0078	0.0021 – 0.0263	0.0119 ± 0.0106	0.0024 – 0.0325	0.0159 ± 0.0121
γ-HCH	0.010[b]	0.0068 – 0.1428	0.0816 ± 0.0588	0.0056 – 0.2682	0.1430 ± 0.1105	0.0042 – 0.2128	0.1262 ± 0.0942
δ -HCH	0.004[a]	n.d.		n.d.		n.d.	
∑-HCH	-	0.0113 – 0.0471	0.0217 ± 0.0166	0.0061 – 0.0780	0.0370 ± 0.0305	0.0166 – 0.6238	0.3508 ± 0.2635
∑-DDT	0.040[a]	n.d.		n.d.		n.d.	

SC – sour cream with 20 % fat content; n – number of samples/each area; n.d. – not detected; s.d.- standard deviation; MRL – maximum residue level; [a] – MRL according to EC Regulation No 396/2005 (Ref. 31); [b] – MRL according to EC Regulation No 149/2008 (Ref. 32).

All the samples contained α-HCH residues but their concentration was lower, it was detected in the analysed samples in the range of 0.0024 to 0.0242 (mg kg^{-1} fat) for raw cow milk, of 0.0017 to 0.0142 (mg kg^{-1} fat) for pasteurised cow milk and of 0.0022 to 0.0189 (mg kg^{-1} fat) for sour cream. The levels of α-HCH residues in pasteurised cow milk samples were lower than those reported by Alawi and Al-Hawadi[36] in milk with 1.9% fat content samples in Jordan (0.060 mg kg^{-1} fat) and higher than those reported by Avancini and Silva[11] in pasteurised cow milk samples in Brazil (0.45–2.34 ng g^{-1} fat).

Considering the fact that this isomer is more volatile and less persistent than the other isomers of the HCH, its lower concentrations are justified. The α-HCH above its maximum residue level is present in about 44, 33 and 39% of the RCM, PCM and SC samples.

Dichlorodiphenyltrichloroethane (DDT) is a potent non-systemic insecticide. In all the analysed samples, DDT and its analogues were non-detectable. The reason for the absence of DDT and its analogues in all the samples analysed can be the fact that their use has been banned in agriculture since 1993.

The results achieved for the OCPs tested can have several motivations. On one hand, the use of OCPs for protecting plants has been drastically reduced after 1991–1993, when the HCH and DDT were banned. However, the γ-HCH isomer (lindane) was still used until 2007, but the number of products in which it was allowed was reduced, only several products based on γ-HCH being used until that year – two insecticides and ten insectofungicides (when the γ-HCH was used in mixture with other perticides). On the other hand, the fact that lindavet, a veterinary product containing γ-HCH, allows the transcutaneous transmission of this product into the animal body led to its banning from veterinary use in our country.

CONCLUSIONS

It could be concluded that organochlorine pesticide residues were detected in raw cow milk and dairy products as they were persistent in the environment due to their slow decomposition rate, long half-life and high stability in the environment. In most cases, the values of detected organochlorine pesticide residues exceeded the maximum residue levels provided by Romanian and international regulations. In case of the analysed samples, there was a difference of organochlorine pesticide residue content due to the producers, the geographical area and due to the agricultural crops existing in these areas. The study carried out allows to establish the possible contamination sources with OCPs and the measures that are needed in order to avoid contamination. The results of this study demonstrate that it is necessary to establish pesticide residue monitoring programs in milk and dairy products as well as in cattle feed and in the storage conditions, in order to improve food safety and decrease exposure risks for consumers. Considering the increase in milk and dairy products consumption from independent producers and small factories and the potential risk associated with the estimated exposure, appropriate management strategies should become a priority in the near future due to the accumulation of OCPs in the food chain and the consequent exposure of the population through the food consumption. Based on this study, it is concluded that further measures, such as education of farmers, control of the sale of pesticides and improvement of the organic agriculture and implementation of integrated pest management methods, are necessary for the reduction of pesticide residues in foods and the prevention of exposure to pesticides. This study represents an important step towards a more comprehensive understanding of the human health risks associated with OCP exposure via milk and dairy product consumption in Romania and in Bacau region.

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