Original Research

Persistent Organochlorine Pesticide, Lead, Cadmium, Nitrate (V) and Nitrate (III) in Polish Milk and Dairy Products

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Abstract

This study was aimed at determining the effect of the region of origin of dairy products and their type on the concentration of extrinsic chemical compounds (lead, cadmium, sum of DDT, γ HCH, total PCB, nitrates (V) and nitrates III). The experimental material included samples of milk, cream, fresh white cheese and butter originating from the central, western and southeastern regions of Poland.

Statistically significant differences (at a level of $p<0.05$) were found in concentrations of γ HCH, lead and cadmium between products originating from particular regions examined. In analyzing the type of a dairy product, significantly higher contents of Σ DDT were noted in milk (average 0.056 $mg \cdot kg^{-1}$of fat). In turn, a significantly higher level of PCB was determined in fresh white cheese (average 0.047 $mg \cdot kg^{-1}$ of fat). Butter was found to contain the lowest level of cadmium (mean: 0.001, range < 0.01-0.003 $mg \cdot kg^{-1}$) and lead (mean: 0.08, range: 0.05-0.012 $mg \cdot kg^{-1}$), as compared to the other products. The mean contents of the compounds analyzed in the study were lower than those stipulated as permissible in Poland.

Keywords: contaminants, DDT, HCH, PCB, lead, cadmium, dairy products

Introduction

The foods and beverages ingested by humans represent a potentially significant pathway of exposure to many hazardous chemicals. Determining human exposure to environmental contaminants is a key element in evaluating health risks [1].

Milk can serve as a vector for the transmission of non-infectious substances which are potentially toxic to the consumer. These toxins may originate in cow's milk from the ingestion of plants known to contain toxic substances or feeds contaminated with mycotoxins, or residues of pesticides or herbicides. Many review articles have been published on this subject [2-5]. These are chemical substances such as: heavy metals [6-11], organochlorine pesticides and polychlorinated biphenyles [4, 9, 12-15], nitrates and nitrites [16, 17], mycotoxins [10], and antibiotics [2].

The aim of our work was to determine levels of DDT (1.1.1,-trichloro-2.2-bis(4-chlorophenyl)ethane), γ HCH (γ-hexachlorocyclohexane), polychlorinated biphenyls (PCB), nitrates (V) and nitrates (III) in milk and dairy products originating from different regions of Poland. The influence of the origins of dairy products on the contents of examined substances and the differences in their concentrations between particular products have been stated.

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Experimental Procedures

The experimental material included samples of milk, cream, fresh white cheese and butter originating from the central, southeastern and west regions of Poland (Fig. 1). The samples were collected during 2002 and 2003. In each sample we determined concentrations of lead, cadmium, Σ DDT (as a total sum of DDT + DDE + DDD), γ HCH, total PCB (determined as Aroclor), nitrates (V), and nitrates (III). A total of 165 samples of milk and dairy products was examined. The compounds were determined following analytical procedures described previously [18].

The contents of lead and cadmium, after dry mineralization of the biological material and extraction to an organic phase (APDC/MIBK), were measured using atomic absorption flame spectrophotometry with a Unicam 939 AA apparatus, equipped with data station Adax and background correction**.** Organochlorine pesticides (DDT and γ HCH) and polychlorinated biphenyls (PCBs) were extracted from the samples with petroleum ether and acetone together with fat. The extracts were hydrolyzed with sulfuric acid. The identification and quantitative determination of the analyzed compounds were carried out with gas chromatography using a PYE Unicam 4600 apparatus with an EC detector. The contents of nitrates (V) and nitrates (III) were assayed with the spectrophotometric method. Differences in the levels of the compounds between region and products were tested by the use of one-way analysis of variance (ANOVA).

Results

The concentration of Σ DDT, γ HCH, total PCB, lead, cadmium, nitrates (V), and nitrates (III) in all examined products in relation to their origin are presented in Table 1. Organochlorine pesticides and polychlorinated biphenyl levels presented are expressed on a lipid basis. Lead, cadmium**,** nitrates (V), and nitrate (III) levels presented are expressed on a product basis.

No significant differences were found in the contents of Σ DDT (Table 1) between the dairy products examined depending on their origin. In the experimental samples, the sum of DDT ranged from 0.004 to 0.136 $mg \cdot kg^{-1}$ of fat. Statistically significant differences ($p<0.05$) were found in the levels of γ HCH between the products originating from the central region of Poland (mean: 0.004 mg kg^{-1} of fat) and those from western Poland (mean: 0.008 $mg \cdot kg^{-1}$ of fat). The mean concentration of PCB in the products originating from western Poland (0.013 $mg \cdot kg^{-1}$ of fat) was ca. twice as low as that found in the products from the southeastern (0.026 $mg \cdot kg^{-1}$ of fat) and western regions of Poland (0.021 $mg \cdot kg^{-1}$ of fat), yet the differences were not statistically significant.

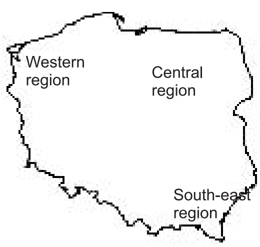

Fig. 1. Map of milk and dairy products collection.

Figs. 2, 3 and 4 show the differences in the levels of DDT, γ HCH, and PCB between particular dairy products. Statistically significant differences were found in the contents of DDT (Fig. 1) between the dairy products examined. Only the level of Σ DDT in cream and white cheese, and in milk and cream was not statistically different. Significantly higher values of Σ DDT were noted in milk (mean: 0.056 $mg \cdot kg^{-1}$ of fat; range: 0.010-0.136). Differences in the concentrations of γ HCH (Fig. 2) between milk, white cheese and butter were not statistically significant ($p<0.05$), in spite of the fact that the range between the milk samples examined was wide. A significantly higher content of PCB (Fig. 3) was determined in fresh white cheese (mean: 0.047 $mg \cdot kg^{-1}$ of fat), as compared with the other dairy products.

Statistically significant differences were observed in the concentrations of lead and cadmium between dairy products originating from different regions (Table 1). Significantly higher contents of cadmium were reported in dairy products from central Poland. However, while the southeastern and central regions have the same measures of mean (0.002 $mg \cdot kg^{-1}$), they have different ranges and they are different in their variability. In both compared cases, the standard deviation data of products from central Poland has more of it. Central Poland's products data introduce higher indicators of variability. The statistical analysis demonstrated significant differences in its concentrations between products from the western region (from 0.001 to 0.007 $mg \cdot kg^{-1}$) and those from central (<0.001to 0.015 $mg \cdot kg^{-1}$) and southeastern Poland (<0.001 to 0.004 $mg \cdot kg^{-1}$).

Figs. 5 and 6 illustrate concentrations of cadmium and lead in particular dairy products. Butter was found to contain a significantly lower (at a level of $p<0.05$) concentration of cadmium (mean: 0.001, range: <0.01-0.003 $mg \cdot kg^{-1}$) as compared to the other products. A similar tendency was observed in the case of lead (mean: 0.08, range: 0.05-0.012 $mg \cdot kg^{-1}$). The differences in its contents were statistically insignificant only between butter and cream. The concentration of metals in dairy products may be affected by both the technological process and the equipment used.

No effect was observed of the origin of dairy products (table 1) on the contents of nitrates (V) and nitrates (III). The mean concentrations of nitrates (V) and nitrates (III) accounted for: 3.39 and 0.35 $mg \cdot kg^{-1}$ (products from the southeastern region), 3.16 and 0.29 $mg \cdot kg^{-1}$ (products from the central region), as well as 2.65 and 0.34 $mg \cdot kg^{-1}$ (products from the western region).

The levels of nitrates (V) and nitrates (III) (Figs. 7 and 8) in fresh white cheese (respectively: 4.09 and 0.45

Table 1. The concentrations of contaminants in milk and dairy products in relation to their origin.

Central region				
		No.of samples	$\overline{x}$ ±SD	Range
Σ DDT	(mg kg^{-1} of fat)	75	0.050 ± 0.029	0.004-0.136
γ HCH	(mg kg^{-1} of fat)	75	0.004^{a} ± 0.003	< 0.001-0.013
PCB	(mg kg^{-1} of fat)	75	0.021 ± 0.042	< 0.001-0.283
Pb	(mg kg^{-1})	75	0.012^{ac} ± 0.007	0.001-0.032
Cd	(mg kg^{-1})	75	0.002^{a} ± 0.002	< 0.001-0.015
NO_3^-	(mg kg^{-1})	60	3.16 ± 1.76	0.19-9.18
NO_2^-	(mg kg^{-1})	60	0.29 ± 0.31	0.01-1.23
Western region				
		No.of samples	$\overline{x}$ ±SD	Range
Σ DDT	(mg kg^{-1} of fat)	30	0.040 ± 0.026	0.005-0.126
γ HCH	(mg kg^{-1} of fat)	30	0.008^{b} ± 0.017	< 0.001-0.060
PCB	(mg kg^{-1} of fat)	30	0.013 ± 0.013	0.001-0.075
Pb	(mg kg^{-1})	30	0.009^{b} ± 0.003	0.003-0.014
Cd	(mg kg^{-1})	30	0.003^{ac} ± 0.002	0.001-0.007
NO_3^-	(mg kg^{-1})	24	2.65 ± 1.76	0.77-6.81
NO_2^-	(mg kg^{-1})	24	0.34 ± 0.28	0.07-0.99
South-east region				
		No.of samples	$\overline{x}$ ±SD	Range
Σ DDT	(mg kg^{-1} of fat)	60	0.044 ± 0.022	0.010-0.107
γ HCH	(mg kg^{-1} of fat)	60	0.005^{ab} ± 0.004	< 0.001-0.018
PCB	(mg kg^{-1} of fat)	60	0.026 ± 0.025	0.002-0.095
Pb	(mg kg^{-1})	60	0.008^{b} ± 0.003	0.004-0.015
Cd	(mg kg^{-1})	60	0.002^{b} ± 0.001	< 0.001-0.004
NO_3^-	(mg kg^{-1})	48	3.39 ± 1.99	0.80-9.17
NO_2^-	(mg kg^{-1})	48	0.35 ± 0.18	0.08-0.89

a,b,c: statistically significant difference at the level <0.05.

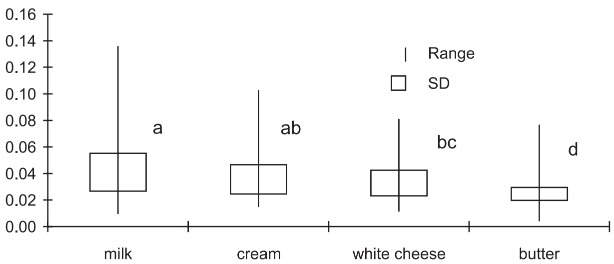

Fig. 2. The concentrations of DDT in milk and dairy products (mg·kg^{-1}of fat basis).
a,b,c,d: statistically significant difference at the level p<0.05.

Fig. 3. The concentrations of γ HCH in milk and dairy products (mg·kg^{-1}of fat basis).
a,b: statistically significant difference at the level p<0.05.

Fig. 4. The concentrations of PCB in milk and dairy products (mg·kg^{-1}of fat basis).
a,b: statistically significant difference at the level p<0.05.

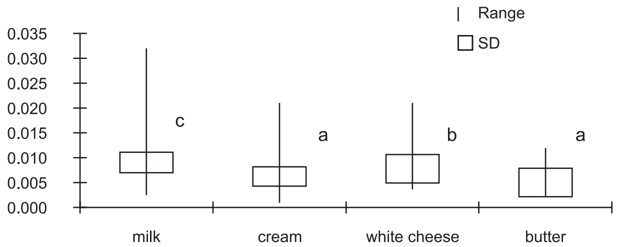

Fig. 5. The concentrations of lead in milk and dairy products (mg·kg^{-1}).
a,b,c: statistically significant difference at the level p<0.05.

Fig. 6. The concentrations of cadmium in milk and dairy products (mg·kg^{-1}).
a,b,c: statistically significant difference at the level p<0.05.

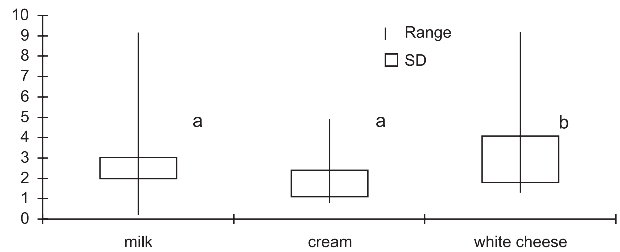

Fig. 7. The concentrations of nitrate (V) in milk and dairy products (mg·kg^{-1}).
a,b: statistically significant difference at the level p<0.05.

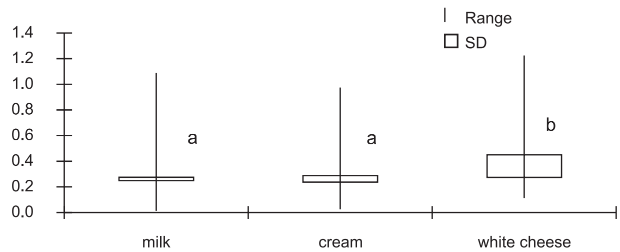

Fig. 8. The concentrations of nitrate (III) in milk and dairy products (mg·kg^{-1}).
a,b: statistically significant difference at the level p<0.05.

mg·kg^{-1}) were significantly higher than in milk (respectively: 3.05 and 0.27 mg·kg^{-1}) and cream (2.41 and 0.29 mg·kg^{-1}).

Discussion of Results

The mean contents of the persistent organochlorine pesticide determined in the study are lower than the results reported over years by other Polish authors. In Poland, the levels of organochlorine compounds in milk and dairy products have been observed to decline [18-23]. According to Smoczyński et al. [20], in 1992 the mean levels of Σ DDT and PCB in milk fat accounted for: 0.137 and 0.141 mg·kg^{-1}, respectively. In 1994, Smoczyński and Pietrzak-Fiećko [22] determined the residues of Σ DDT and γ HCH in samples of milk fat at a level of 0.438 and 0.012 mg·kg^{-1}, respectively, whereas in 2000 Pietrzak-Fiećko et al. [23] assayed the sum of DDT in milk at a level of 0.0014 mg·l^{-1}. Despite this positive trend, some levels of those compounds are still detected. In the 1970s, burial grounds (concrete underground chambers) were built in order to dispose of those compounds, yet they can be subject to damage and evoke secondary contamination of the environment. Hence, the monitoring of the residues of these compounds is indispensable. Nowadays, the highest levels of Σ DDT (0.146 mg·kg^{-1}, 0.156 mg·kg^{-1}) and γ HCH (0.049 mg·kg^{-1}) in milk fat have been reported in Mexico [4.24]. In China [25] and India [3] the residues of Σ DDT in milk fat accounted for: 0.046 and 0.041 mg·kg^{-1}, respectively, whereas the mean level of γ HCH residues in milk fat fluctuated at a similar level, reaching 0.01 mg·kg^{-1} of fat. Literature data lack explicit scientific studies on the effects of technological processes on changes in the concentration of lipophilic organochlorine compounds. Pandit et al. [3] reported on lower levels of Σ DDT in cow's milk (mean: 0.041 mg kg^{-1} of fat) as compared to butter (0.071 mg·kg^{-1} of fat). In contrast, Waliszewski et al. [26] demonstrated higher levels of Σ DDT in milk (mean: 0.159 mg·kg^{-1} of fat), as compared to butter (mean: 0.064 mg·kg^{-1} of fat).

The obtained Σ DDT and γ HCH values are considerably lower than the highest permissible levels of these compounds specified by the Ministry of Health in Poland [27]. According to the national legal regulations, the concentrations, the Σ DDT and γ HCH contents in dairy products with over 2% fat content should not exceed 1 and 0.025 mg·kg^{1} of fat, respectively.

The major cause of the presence of metals in milk is their occurrence in soil. Low concentrations of metals in soil are not reflected in their relatively low contents in plants, since absorption of lead and cadmium by plants is affected, to a great extent, by soil pH. On acidic soil, contaminants penetrate into plants more intensively than on neutral soil. Studies into the quality of soil carried out by the Institute of Cultivation, Fertilization and Soil Science in Puławy [28] as well as by Chemical-and-Agricultural Stations indicate that 1/3 of soils in Poland are excessively acidified – pH<5.5 (necessity of liming). Out of the regions compared, the southeastern region of Poland is characterized by the greatest area (as much as 54%) of acidified soils with pH<5.5. Studies carried out in 1998-2000 on the quality of soils indicate that amongst the regions discussed in the presented paper, the highest concentrations of lead were observed in soils of the western region of Poland, whereas the lowest contents of cadmium occur in southeastern soils and the lowest contents of lead were in the soils of central Poland.

Swarup et al. [6] found a positive correlation between levels of cadmium and lead in the blood of urban and rural cows. The investigated samples of dairy products originated from regions characterized by relatively high agricultural production and a low degree of industrialization. Our results are comparable, and even in some cases a little higher, than results reported by Italian [29], Spanish [8], Slovenian [9] and Lithuanian authors [30.31]. The concentration of lead and cadmium means Poland's monitoring studies amounted to 0.003 and <0.001 mg kg^{-1} respectively [28].

The samples' place of origin had no impact on the values determined for concentrations of nitrates (V) and nitrates (III). Priyo Bintaro et al. [32] demonstrated that the topographic profile did not affect the content of nitrates in milk, whereas it had a significant effect on the composition of food rations fed to cows. Similar findings were reported by Vittozzi [33]. In a study by Slovak authors [34], the level of nitrates (V) in milk ranged from 0.19 to 0.44 mg $NaNO_3 \cdot l^{-1}$, whereas that of nitrates (III) from 0.008 to 0.014 mg $NaNO_2 \cdot l^{-1}$. In cow's milk originating from Indonesia, Priyo Bintoro et al. [32] determined concentrations of nitrates at a level of <3 mg $NO_3^- \cdot l^{-1}$. In turn, Brazilian researchers [17] demonstrated the mean contents of NO_3^- and NO_2^- in fresh milk to reach, on average, 0.831 $mg \cdot l^{-1}$ (range: 0.02-2.1) and 0.082 $mg \cdot l^{-1}$ (range: 0.05-0.18), respectively.

The investigations in this paper reveal low concentrations of lead, cadmium, Σ DDT, γ HCH, PCB, nitrates (V) and nitrates (III) in milk and dairy products. Additionally, from the human health point of view, the low levels of a number of food contaminants cannot be ignored. The fact of their delayed action should be taken into account while estimating health effects and determining acceptable levels of these compounds in food products.

Conclusions

Analyses carried out within the study indicated that the origin of dairy products had no effect on the presence of DDT, PCB, nitrates (V) nor nitrates (III). In the case of γ HCH, lead and cadmium, their statistically significantly different concentrations were reported in products originating from different regions.

In analyzing the type of a dairy product, the lowest concentrations of the compounds examined were found in butter, whereas fresh white cheese was observed to be characterized by significantly higher concentrations of PCB, nitrates (V) and nitrates (III).

The determined concentrations of the compounds examined are substantially lower than those reported in Chinese, Indian and Mexican studies, and comparable with findings of European authors.

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