Banned Organochlorine Pesticides Still in Our Food: The Presence of Organochlorine Pesticide Residues in Milk, Meat, Liver, and Kidney in Jordan Cattle

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ABSTRACT

To assess the potential risks posed by residual organochlorine pesticides (OCPs) to human health, we evaluated the consumption of animal products as the primary route of human exposure to these compounds. In this study, 120 samples of milk, meat, and edible tissues (liver and kidney) were collected from farms and slaughterhouses in Amman, Jordan. Fifteen OCPs, including dichlorodiphenyltrichloroethane (DDT) and its metabolites, hexachlorocyclohexane isomers (HCHs), aldrin, dieldrin, endrin, heptachlor, heptachlor epoxide, and hexachlorobenzene (HCB), were identified and further investigated. These samples were extracted using the Soxhlet method, subjected to Florisil column chromatography, and analyzed using gas chromatography with an electron capture detector (GC-ECD). The results revealed that 40%, 40%, 46.7%, and 33.3% of the examined milk, meat, liver, and kidney samples, respectively, were contaminated with OCPs. This study confirmed that residual OCPs persist in cow-derived food products in Jordan.

INTRODUCTION

An increase in global food demand, especially in developing countries experiencing population increases, has led to extensive use of agrochemicals to enhance food production [1,2]. The extensive use of agrochemicals, such as pesticides, is driven by the need to meet the growing food demand for the increasing global population. The global population is expected to increase by 30% to approximately 9.2 billion in 2050 [3]. Due to the increasing global population and dietary preference for meat and milk products in developing countries, the demand for food production is projected to increase [4].

Pesticides are toxic compounds that vary in both acute and chronic toxicity; therefore, they can be classified according to their hazardous effects. Pesticides, such as formothion, simazine, and dichlorodiphenyltrichloroethane (DDT), are of great concern to both human health and environmental safety. Subsequently, organizations and countries have prohibited their registration and use in agriculture [5,6]. Organochlorine pesticides (OCPs) are a class of pesticides that have been used in the agriculture and public health sector (in the form of antipathogen vector insects) for several decades. The popularity of OCPs is primarily driven by their low cost. In fact, several of these old pesticides are patented.

The major threats posed by OCPs have been attributed to their (1) residual effects, (2) persistence, (3) reduced solubility in water, (4) lipid nature, and (5) bioaccumulation in adipose tissue. These characteristics make it easier for them to be transported and carried over into higher food chain levels. OCP residues have become a significantly contribute to environmental pollution and their noxious effects are prominent in humans and animals [7]. Most OCPs used in Jordan (aldrin, chlordane, dieldrin, endrin, heptachlor, and hexachlorobenzene (HCB)) for controlling agricultural pests have been banned since the 1980s [8]. However, DDT was used up to 1995 for controlling insects (such as mosquitoes) that were of public health concern. Several OCPs have carcinogenic effects and are classified as endocrine-disrupting Keywords: Organochlorine; pesticide residues; cow; milk; meat; liver; kidney; Jordan

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chemicals [9]. OCPs including DDT, HCB, and hexachlorocyclohexane (HCH) are classified in group 2B (carcinogenic in humans) according to the International Agency for Research on Cancer [10,11].

Although most OCPs have been legally banned from commercial use and trade in Jordan, OCP residues are still being detected in different environmental matrices, including animal products [5], human milk [12], eggs, chicken, meat [13], dairy products [9], sheep milk [14], sheep liver and kidney [15], soil [16], fish [17], honey [18], grapes and homemade wine [6], and adipose tissues and fodder [14,15]. Therefore, identifying and monitoring OCPs in various environmental matrices, especially in animal food products, is important for environmental safety and human health. Therefore, this study aimed to characterize the degree of contamination with DDT, HCH, aldrin, dieldrin, endrin, and heptachlor residues in milk, meat, liver, and kidney obtained from cows.

MATERIALS AND METHODS

Sampling

A total of 120 samples (30 samples of each cow product milk, meat, liver, and kidney) were collected from slaughterhouses and farms in Amman, Jordan. Approximately 50 mL of cow milk was collected in clean, dry glass bottles, kept on dry ice during transportation to the laboratory, and stored at -20 °C until analysis. For meat, liver, and kidney, 250 g of each sample was kept separately in polyethylene bags inside a dry-ice box during transportation to the laboratory and stored at 4 °C until analysis within 24 h of arrival to the laboratory. No live animals/human beings were involved in this study; hence, ethics committee approval was not applicable in this case. *Chemicals and Reagents*

All analytical-grade solvents used for pesticide residue analyses were purchased from Riedel de-Hean (Germany). The OCP standards (o,p-DDT, p,p-DDT, o,pdichlorodiphenyldichloroethylene (DDE), p,p-DDE, o,pdichlorodiphenyldichloroethane (DDD), p,p-DDD, α -HCH, β -HCH, γ -HCH, aldrin, dieldrin, endrin, heptachlor, heptachlor epoxide, hexachlorobenzene (HCB), and isodrin as an internal standard) were sourced from Dr. Ehrebstorfer (GmbH, Germany). Analytical-grade anhydrous sodium sulfate (Fluka, Switzerland) was heated at 550 °C for 3 h and kept in a closed container. Florisil, 60–100 mesh, was obtained from Riedel de-Hean (Germany), heated at 550 °C for 12 h, and kept in a closed container. Then, the required amount of Florisil was reheated at 130 °C for 1 h before use.

Analysis of Cow Milk Samples

Determination of Fat Content

First, 10 g of milk samples were thoroughly mixed with 2 mL of 25% ammonia, 25 mL diethyl ether, and 25 mL petroleum ether in a separation flask equipped with a Teflon stopcock. The organic solvent layer was removed, and the extraction procedure was repeated twice. This procedure was followed by pooling and filtration (through anhydrous sodium sulfate) of the fat-containing organic extraction layers. The solvent was evaporated at 30 °C and 200 mbar in a preweighed round-bottom flask. The round-bottom flask with residue was kept in a desiccator overnight and reweighed. The percentage of fat content of the milk was calculated on the basis of the weight difference.

Pesticide Extraction

First, 25 g of Florisil (3% water) was placed in a chromatography column (50 cm × 2 cm with a Teflon stopcock), which containing 100 mL of petroleum ether. The remainder of the petroleum ether was reduced to approximately 50 mL after Florisil sedimentation. Then, 10 g of each milk sample was added to the chromatographic column with 25 g of Florisil. The column was eluted with 300 mL of elution mixture (80% petroleum ether: 20% dichloromethane). The eluate was collected in a 500 mL round-bottom flask and evaporated to near dryness using a rotary evaporator at 35 °C and 12 mbar. The remaining solvent was evaporated under nitrogen stream. The residues were dissolved in 2 mL nhexane containing 0.5 ppm isodrin as an internal standard, and 1 μ L of the final extract was injected into the gas chromatography (GC) column.

Analysis of Cow Meat, Liver, and Kidney Samples

Homogeneous samples (25 g of meat, liver, or kidney) were mixed with 50 g of anhydrous sodium sulfate in a mortar. The mixture was transferred to a thimble and extracted with 250 mL of petroleum ether in a Soxhlet apparatus for 6 h. The extracts were evaporated to near dryness using a rotary evaporator. The remaining solvent was evaporated under a nitrogen stream. The residue was left in desiccators for 30 min, and the fat residue was then weighed to obtain the percentage of fat in the sample. Residual fat was removed from the extracted samples. The extract from the previous step was dissolved in 10 mL of petroleum ether and transferred with a Pasteur pipette to the Florisil column. The column was then eluted with 300 mL of petroleum ether and dichloromethane (80%/20%

v/v). Subsequently, the eluates were evaporated using a rotary evaporator under nitrogen stream. The dried residual eluates were dissolved in 2 mL *n*-hexane containing 0.5 ppm isodrin as an internal standard, and 1 μ L of the final extract was injected into the GC column. *GC Analysis*

OCPs in samples were analyzed using a gas chromatograph (HP 5890) equipped with an ⁶³Ni electron capture detector, as well as an HP-5 capillary column (30 m \times 0.32 mm inner diameter (i.d.) with 0.25 µm film thickness). The carrier gas was helium at a flow rate of 2 mL·min ¹. Argon-methane was the make-up gas and flowed at a rate of 30 mL⋅min⁻¹. Running conditions for GC were an injection temperature of 300 °C and detection temperature of 300 °C. The oven temperature was initially 80 °C (2.2), increased to 175 °C (at a rate of 30 °C·min⁻¹), then to 225 °C (10 °C·min⁻¹), before being held at 225 °C (2 min). The sample injection volume was 1 μ L and the split ratio was 1:25. The identification of OCPs in the samples was made by comparing their retention time with those in the standard mixture with respect to the internal standard retention time under the same injection conditions. Residual OCPs were quantitatively determined using the relative peak area of the sample chromatogram and the relative concentration. The concentration of OCP residues in each sample was reported as mg·kg⁻¹ on a fat basis.

Recovery Tests and Detection Limits

Extraction and cleanup methods were assessed by placing a known concentration of OCPs in blank samples. The spiked samples were analyzed according to the procedures mentioned above. The average recoveries of OCPs were 89.6%–102.3% for milk samples and 88.1%– 97.8% for meat, liver, and kidney samples. The detection limit for each compound was calculated as a signal-tonoise ratio of 3:1 from the chromatogram of the standard mixture after several dilutions. The results show a detection limit ranging from 0.004–0.005 mg·kg⁻¹.

RESULTS

This work is a continuation of previous studies, dating from 1993 to monitor organochlorines in Jordan. During the period of 1993-2003, the Ministry of Environment in Jordan funded four research studies to clarify the extent of OCP contamination in food and to establish a baseline over time. Furthermore, the data from these studies would inform policymakers and serve to provide evidence to direct and propose suitable solutions to the potential OCP hazard in Jordan [17]. Of the 120 samples collected from cows (milk, meat, liver, and kidney), the results revealed that 48 (40%) of the analyzed samples were contaminated with OCP residues. Furthermore, seven (23.3%) of the milk samples exceeded the maximum residue limit (MRL) according to the Food and Agriculture Organization (FAO)/World Health Organization (WHO) Codex Alimentarius [19,20] (Table 1).

 Table 1. Cow milk and meat organochlorine-contaminated samples. DDT, dichlorodiphenyltrichloroethane; HCH, hexachlorocyclohexane; MRL, maximum residue limit.

	Mil	k	Meat			
Pesticides	Codex MRL (mg/kg on Fat Basis)	No. of Samples above MRL	Codex MRL (mg/kg on Fat Basis)	No. of Samples above MRL		
DDT	0.02	1	5	0		
Aldrin and dieldrin	0.006	0	0.2	0		
Lindane (y-HCH)	0.010	3	0.1	0		
Heptachlor	0.006	3	0.2	0		

According to the Codex Alimentarius, residual pesticides exceeded MRL (mg/kg on fat basis).

DISCUSSION

Dichlorodiphenyltrichloroethane (DDT) and Its Metabolites DDT is a carcinogenic chlorinated aromatic hydrocarbon [21]. It was released in 1945 for public sale when insecticides were commonly known as "economic poisons". This chemical enhances agricultural productivity by combating agricultural pests and insects [22]. After the publication of Silent Spring in 1962, in which Rachel Carson documented the harmful effects of long-term use of DDT and other insecticides on human health, the Environmental Protection Agency (EPA) was prompted to prohibit its registration and ban its use in 1972 [23]. As other countries banned DDT in agricultural use, it remained legal in Jordan for controlling insect vectors of public health concern (such as mosquitoes) until 1995 [9]. Due to the chemical properties of DDT, it persists in the environment for long periods of time and can still be found in our food. In this study, 5 (16.7%) of milk, 9 (30%) of meat, 9 (30%) of liver, and 8 (26.7%) of kidney samples, were found to contain DDT. This represents an overall detection of 31 (25.8%) of the total analyzed samples of cow products containing DDT (Table 2). The mean DDT concentrations were 0.016 mg·kg⁻¹ in milk fat, 0.130 mg·kg⁻¹ in meat, 0.107 mg·kg⁻¹ in liver, and 0.170 mg·kg⁻¹ in kidney (Table 2). *o*,*p*-DDT, *o*,*p*-DDE, and *o*,*p*-DDD, were the only metabolites of DDT (*o*,*p*-DDT, *p*,*p*-DDT, *o*,*p*-DDE, *p*,*p*-DDE, *o*,*p*-DDD, and *p*,*p*-DDD) that were not observed at concentration greater than the detection limits. *p*,*p*-DDE was the most abundant metabolite of DDT detected in the samples.

Table 2. Organochlorine pesticides detected in milk, meat, liver, and kidney obtained from cows (mg·kg⁻¹ on fat basis). HCB, hexachlorobenzene; DDE, dichlorodiphenyldichloroethylene; DDD, dichlorodiphenyldichloroethane.

Pesticide	Milk (<i>n</i> = 30)		Meat (<i>n</i> = 30)		Liver (<i>n</i> = 30)			Kidney (<i>n</i> = 30)				
	Frequency	Mean	Range	Frequency	Mean	Range	Frequency	Mean	Range	Frequency	Mean	Range
HCB	n.d.			n.d.			n.d.			n.d.		
α-ΗСΗ	1	0.017		1	0.026		3	0.020	0.009– 0.026	4	0.064	0.010- 0.200
β-НСН	3	0.080	0.014- 0.210	4	0.146	0.014- 0.340	5	0.079	0.006- 0.340	4	0.255	0.018- 0.450
ү-НСН	3	0.040	0.010- 0.080	1	0.067		n.d.			n.d.		
Σ НСН	7	0.055	0.010- 0.210	6	0.110	0.014- 0.340	7	0.066	0.006- 0.34	7	0.180	0.018- 0.450
Heptachlor	3	0.017	0.010- 0.025	2	0.027	0.016- 0.037	2	0.020	0.016- 0.030	n.d.		
Heptachlor epoxide	2	0.019	0.010- 0.029	2	0.050	0.036- 0.070	1	0.07		n.d.		
Σ heptachlor + epoxide	4	0.023	0.018- 0.029	3	0.050	0.036- 0.086	2	0.055	0.030- 0.086	n.d.		
<i>p,p</i> '-DDT	n.d.			2	0.215	0.070- 0.360	2	0.150	0.100- 0.200	2	0.390	0.350- 0.430
o,p'-DDE	n.d.			n.d.			n.d.			n.d.		
<i>p,p</i> '-DDE	4	0.017	0.008- 0.026	5	0.130	0.030- 0.26	4	0.123	0.030- 0.210	6	0.090	0.017- 0.350
p,p'-DDD	1	0.014		2	0.050	0.030- 0.070	3	0.060	0.030- 0.080	3	0.020	0.010- 0.030
Σ DDTs	5	0.016	0.008- 0.026	9	0.130	0.007- 0.360	9	0.107	0.0300- 0.210	8	0.170	0.010- 0.430

n.d., not detected.

The analysis also shows that 4 (13.3%), 5 (16.7%), 4 (13.3%), and 6 (20%) milk, meat, liver, and kidney samples, respectively, were positive for *p*,*p*-DDE, with an overall detection of 15.8% (19/120; Table 2). The quantities of DDT metabolites found, in increasing order, were *p*,*p*-DDE > *p*,*p*-DDD > *p*,*p*-DDT (Table 2). One sample of cow milk had concentrations higher than the MRL for DDT (sum of o,p-DDT, p,p-DDT, o,p-DDE, p,p-DDE, o,p-DDD, and *p*,*p*-DDD; Table 1). The concentration of the sum of DDT metabolites in milk recorded in this study was lower than the concentration in cow milk analyzed in Giza (Egypt) [7]. In India [24,25], Ethiopia [26], and Mexico [27], DDT was detected in samples with a mean of 0.033, 0.041, and 0.159 mg·kg⁻¹ on a fat basis, respectively. In addition, DDT was lower than in the concentrations recorded in liver, kidney, and milk from sheep farmed in Jordan [14,15]. Ahmad et al. (2010) detected DDT quantities in meat (lamb and beef) with mean concentrations of 0.045 mg·kg⁻¹ on a fat basis [13], which is less than that detected in cow meat in the present study. *4.2. Hexachlorocyclohexane (HCH) Isomers*

The common isomers of HCH (α -, β -, γ -, and δ -HCHs) are present in at least 146, 159, 189, and 126 sites, respectively, of the 1662 current or former National Priorities List sites classified by the EPA to identify serious hazardous waste sites in the United States of America (USA) for long-term federal cleanup activities [28]. Despite the 1981 ban on the use of HCHs in Jordan, they are still detectable in dairy milk [9]. In this study, HCH isomers (α , β , and γ) were detected in 7 (23.3%), 6 (20%), 7 (23.3%), and 7 (23.3%) samples of milk, meat, liver, and kidney, respectively (Table 2). The mean values of the residual concentrations (mg·kg⁻¹ fat) of HCHs in the examined samples of milk, meat, liver, and kidney were 0.055, 0.110, 0.066, and 0.180, respectively (Table 2). γ -HCH (lindane) the HCH isomer with the greatest activity and relevance to health outcomes, was detected in three milk samples with a mean concentration of 0.040 mg·kg⁻¹ fat. This exceeded the MRL of 0.010 mg·kg⁻¹ fat (Table 1). Furthermore, γ -HCH was detected in one sample of meat with a mean concentration of 0.067 mg·kg⁻¹ fat (Tables 1 and 2).

The concentration of HCH of milk in this study was lower than that analyzed in both India [29] and Veracruz (Mexico) [27], where it was detected in the samples with a mean of 0.071 and 0.094 mg·kg⁻¹ on a fat basis, respectively. Furthermore, the concentration of HCH in our samples was lower than the concentrations recorded in sheep liver, kidney, and milk found in Jordan [14,15]. Ahmed and Zaki (2009) detected total HCH values in meat (lamb and beef) in Jordan with a mean concentration of 0.050 mg·kg⁻¹ fat [7], which is less than that detected in cow meat during the present study.

4.3. Heptachlor and Heptachlor Epoxide

Heptachlor is a chlorinated dicyclopentadiene insecticide that persists in the environment for long periods of time and accumulates at various levels in the food chain. Its use has been banned or restricted in many countries since the 1980s; however, it is still detected as a contaminant in some food commodities [30]. Heptachlor was observed in three milk samples with an average concentration of 0.017 mg·kg⁻¹ fat. This exceeded the MRL of 0.006 mg·kg⁻¹ fat (Tables 1 and 2). Two meat samples were contaminated with a mean of 0.027 mg·kg⁻¹ fat. This did not exceed the MRL of 0.1 mg·kg⁻¹ fat (Tables 1 and 2). Moreover, heptachlor was detected in two liver samples with a mean of 0.020 mg·kg⁻¹ fat but was not detected in kidney samples (Table 2). Prolonged exposure to this insecticide causes central nervous system toxicity and is associated with damage to the liver [31,32]. Heptachlor was placed in EPA Group 2B, as sufficient evidence of carcinogenicity was found in animals but is inadequate in humans [33,34]. Heptachlor epoxide was also found to be present in 2 (6.7%), 2 (6.7%), and 1 (3.3%) milk, meat, and liver samples with residual concentrations of 0.019, 0.050, and 0.070 mg·kg⁻¹ fat, respectively (Table 2). Alawi and Al-Hawadi (2005) detected heptachlor and heptachlor epoxide in sheep milk in Jordan, with a mean concentration of 0.560 and 1.211 mg·kg⁻¹ fat [14], which is greater than that detected in cow milk during this study. Aldrin, Dieldrin, Endrin, and HCB

Combining hexachlorocyclopentadiene with norbornadiene produces aldrin in a Diels-Alder reaction [35]. The Stockholm Convention on Persistent Organic Pollutants and the European Union (EU) banned its use, while the United States proceeded with de-registration in 1974 [36]. Aldrin, endrin, dieldrin, and HCB were not detected in any samples (Table 2). Alawi and Al-Hawadi (2005) detected aldrin, dieldrin, endrin, and HCB in sheep milk samples from Jordan with a frequency of 48%, 14%, 10%, and 40% and a mean concentration of 0.021, 0.158, 0.684, and 0.161 mg·kg⁻¹ fat [14], respectively. Ahmad et al. (2010) detected aldrin in meat (lamb and beef) in Jordan in one sample with a concentration of 0.470 mg·kg-1 fat and detected HCB in one sample with a concentration of 0.009 mg·kg⁻¹ fat [13].

CONCLUSION

In conclusion, despite the general agreement that meeting food demand without further threats to the environment and human health is vital, this study exposed the existence of residual OCPs that persist above the maximum residue levels in some cow-derived food products in Jordan. Our findings will stimulate debate among policymakers, public health experts, and agriculturists to identify sustainable solutions needed to achieve concentrations of OCPs below the highest levels that are legally tolerated. We are optimistic that future research will stimulate and prioritize further investigation of this topic, accompanied by appropriate policies and institutions to support them.

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