Organochlorine Pesticide Residues in Total Diet Samples from Aragón (Northeastern Spain)

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Samples of different meals of the average diet consumed in Aragon (northeastern Spain) were analyzed by capillary gas chromatography to determine residues of 21 organochlorine pesticides. Only hexachlorobenzene (HCB), γ-hexachlorocyclohexane (lindane), *o*,*p*′-DDD, *p*,*p*′-DDE, *p*,*p*′-DDT, and β-endosulfan were detected in the samples. On the whole, the prevalence of contamination decreased from γ-HCH (21.0%) to DDT (13.2%) to HCB (10.3%) to endosulfan, which was detected only in 0.4% of samples. Fatty meals (egg meals > legumes > fish > meat) were most contaminated, responsible for 90% of the contamination detected. The levels of these organochlorine compounds were well below the respective maximum residue limits set by current European regulations. The average level of contamination of the Aragonese diet was believed to be totally harmless and indicated a clear decrease of the presence of organochlorines in foods as reported in other developed countries.

Keywords: *Total diet; organochlorine pesticides*

INTRODUCTION

The use of organochlorine compounds in agriculture and animal production has led to the contamination of foodstuffs, especially those having a high fat content. Considerable data have been amassed on the presence of organochlorine pesticide (OCP) residues in foods. As legal tolerances of pesticide residues are set for raw materials, foods are usually analyzed unwashed and in the raw state. However, any removal or breakdown of residues due to washing or cooking is not therefore taken into account. In this way, the Joint Meeting of the FAO Working Party of Experts and the WHO Expert Committee on Pesticide Residues expressed the opinion that the use of total diet studies for monitoring levels of pesticide residues should be encouraged (FAO/WHO, 1976). Such studies have been carried out in a limited number of countries so far: the United States (FDA, 1991), New Zealand (Pickston et al., 1985), Switzerland (Wüthrich et al., 1985), Great Britain (Fisher, 1987), and The Netherlands (De Vos et al., 1984; Van Dokkum and de Vos, 1987).

Several previous studies indicated the degree of contamination of Spanish foods by OCPs, but there was only a total diet study published, in which only data for α-HCH, lindane, and DDTs were reported (Carrasco et al., 1976). However, these works were concerned mainly with raw foods, and no comprehensive information is available covering cooked food. Recent studies provide estimates of the average intakes of residues of OCPs from a selected number of ready-to-eat food classes (Herrera et al., 1995) and from the total diet of the Basque country (Spain) (Urieta et al., 1996). Such studies allow the conclusion that the dietary intakes of chlorinated pesticides in Spain are of the same order as those observed in most developed countries.

The present study was carried out to estimate the extent of contamination with organochlorine residues in the diet of Aragon (northeastern Spain) as well as the trend of this contamination in recent years. The reported data were compared with the situation in other developed countries, characterized by the decreasing levels of these compounds as a consequence of the restriction or banning of these insecticides.

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MATERIALS AND METHODS

Sample Collection. In this study, the approach of a market basket survey was followed. A total of 281 samples of constituent meals of the average Aragonese diet were randomly collected between December 1991 and June 1994, from 5 different catering establishments. The foods were previously prepared in a manner ready for consumption. Samples were classified into eight meal groups. Table 1 lists the meals that compose each group and the number of samples analyzed in each one. Only the edible portion was used for analysis.

According to the National Survey of Food Consumption published by the Spanish Ministry of Food and Agriculture (MAPA, 1993), the food consumption figures of Aragón are very similar to those of Spain considered as a whole.

Reagents. All solvents (petroleum ether, ethyl acetate, cyclohexane, and isooctane) were of pesticide residue grade and subjected to a solvent purity test for residue analysis suitability (AOAC, 1990).

Anhydrous granular sodium sulfate (Panreac) was heated in a furnace at 600 °C for 6 h to remove impurities. Reference standards of the organochlorine pesticides were obtained from Supelco and Dr. Ehrenstorfer.

Chemical Analysis. Samples were analyzed for residues of hexachlorobenzene (HCB), hexachlorocyclohexane (α-, β-, and γ-isomers of the HCH), aldrin, dieldrin, endrin, heptachlor, heptachlor epoxide, endosulfan (α- and β- isomers), chlordane (α- and γ-isomers), α-chlordene, *trans*-nonachlor, and DDT and its metabolites DDE and DDD, as both *o*,*p*′- and *p*,*p*′-isomers.

Fat and residues were removed from samples of legumes, potato chips, cereal-based foods, meat, eggs, and fish by dissolving 25−75 g of sample (sample size was adjusted to provide an appropriate amount of fat for cleanup) in petroleum ether (three extractions with 150, 100, and 100 mL, respectively, by blending at high speed 2 min each time), as described in the *Pesticide Analytical Manual* (FDA, 1994). Anhydrous sodium sulfate (100 g) removes water and helps to disintegrate the sample. Fat content was determined according to the method of Lázaro et al. (1995).

Samples of cooked potatoes, vegetables, and fruits (75 g) were extracted by homogenizing with ethyl acetate (200 mL) in the presence of anhydrous sodium sulfate (40 g) (Ministry

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Table 1. Composition of Different Meals and Number of Samples Analyzed

meals	no. of samples	meals	no. of samples
legumes	22	meat	77
lentils	8	beef steak	10
beans	7	roast beef	10
chickpeas	7	fried pork	12
potatoes	20	fried lamb	10
cooked	10	grilled breast of chicken	8
potato chips	10	roasted leg quarter of chicken	9
vegetables	47	hamburgers/meatballs	8
salad	9	cooked sausages	10
string beans	9	eggs	29
green peas	9	potato omelette	9
leaf beet/borage	10	fried eggs	10
stewed vegetables	10	cooked eggs	10
fruits	30	fish	24
apples/pears	8	nonfatty fish	12
oranges/mandarin oranges	11	fatty fish	5
other fruits	11	freshwater fish	7
cereal-based foods	32		
macaroni/spaghetti	8		
cooked rice	8		
paella (Spanish rice dish)	6		
croquettes	10		

of Welfare, Health and Cultural Affairs, The Netherlands, 1988). The solvent was evaporated on a rotary evaporator.

The extracts were redissolved in ethyl acetate−cyclohexane (1:1) so as to give concentrations not exceeding 0.85 g of fat/5 mL and cleaned up by gel permeation chromatography (GPC) on an ABC SP-1000 with a Bio-Beads SX-3 column. Chlorinated hydrocarbon residues were eluted with ethyl acetate−cyclohexane (1:1) at the rate of 5 mL/min. For clean-up of the extracts by gel permeation chromatography, the solutions were placed on the above column and eluted with solvent. The fat was eluted first and discarded (dump fraction, 85 mL), leaving the analytes in the next portion of eluant (collect fraction, 165 mL).

Samples were injected into a Hewlett-Packard 5890 gas chromatograph equipped with ^{63}Ni electron capture detector (ECD) and automatic injector (HP 7673A). Fused silica capillary columns coated with 5% phenyl methyl polysiloxane (007-2, 50 m × 0.25 mm i.d. × 0.25 μm film thickness) and cyanopropylphenyl methyl polysiloxane (007-608, 30 m × 0.53 mm i.d. × 0.80 μm film thickness) were used for determination. An HP 3365 Series II Chemstation with HP Chem software was used for quantitation, and the calculation procedure was based on the external standard method.

Recoveries of organochlorine pesticides according to this method were determined by fortification of corn oil with isooctane solutions of the investigated compounds, as recommended by the Association of Official Analytical Chemists (AOAC, 1990), and ranged from 80 to 110%, in agreement with FDA recommendations (FDA, 1994). Reported concentrations were not corrected for the recovery percentage. The quality of analytical data was assured by participation in two Intercomparison Exercises for Chlorobiphenyls and Organochlorine Pesticides organized by the SOAFD Marine Laboratory, Aberdeen, U.K., and sponsored by the EEC Community Bureau of Reference (BCR).

For every set of 10 samples, a procedural blank consisting of all reagents and glassware used during analysis was run to check for interferences and cross contamination. Detection limits are indicated in Table 2. Since no interfering peaks were observed in the blanks, the detection limit of the pesticides was set to the minimum concentration of each one that had a linear response in the ECD.

A descriptive statistical analysis of the results was made with the use of Statview SE + Graphics (Abacus Concepts, Inc., 1988, Berkeley, CA) for personal computers.

RESULTS AND DISCUSSION

Contamination Pattern. The percentage of detection, mean concentration, and range of OCP residues detected in different food items are summarized in Table 3. Mean levels refer to positive samples.

Table 2. Detection Limits in Samples According to the Two Different Techniques of Extraction

chemical	detection limit A[a]	detection limit B[b]
HCB	1	1
heptachlor epoxide, α-chlordane, α-chlordene, *trans*-nonachlor	2	1
α-HCH, γ-HCH, heptachlor, aldrin, dieldrin, γ-chlordane, α-endosulfan, *o,p′*-DDD, *p,p′*-DDE	3	1
o,p′-DDE	4	1
β-endosulfan, *p,p′*-DDD	5	1
o,p′-DDT	9	1
endrin	13	1
β-HCH	14	1
p,p′-DDT	18	1

[a] A, samples extracted with petroleum ether (ng/g on a lipid basis) (FDA, 1994). [b] B, samples extracted with ethyl acetate (ng/g on a wet weight basis) (Ministry of Welfare, Health and Cultural Affairs, The Netherlands, 1988).

Table 3. Descriptive Analysis of OCP Residues in 281 Samples of Constituent Meals of the Aragonese Total Diet (Results Expressed as Nanograms per Gram on a Wet Weight Basis)

meals (*n*)	descriptive statistics	HCB	γ-HCH	DDT[a]	β-endosulfan
legumes (22)	mean level[b]	0.2	0.3	0.1	nd[c]
	range	0.1−0.5	0.1−0.9	0.1−0.4	
	% of detection	22.7	22.7	22.7	
potatoes (20)	mean level[b]	nd	nd	4.3	nd
	range			4.3	
	% of detection			5.0	
vegetables (47)	mean level[b]	nd	56.1	nd	4.0
	range		1.0−213.3		4.0
	% of detection		8.5		2.1
fruits (30)	mean level[b]	nd	nd	2.0	nd
	range			2.0	
	% of detection			3.3	
cereal-based (32)	mean level[b]	0.2	nd	nd	nd
	range	0.2−0.3			
	% of detection	9.4			
meat (77)	mean level[b]	nd	3.5	3.3	nd
	range		0.2−21.4	1.1−8.8	
	% of detection		37.7	16.9	
eggs (29)	mean level[b]	1.6	3.6	3.4	nd
	range	0.8−9.6	1.3−21.6	2.5−4.4	
	% of detection	62.1	58.6	27.6	
fish (24)	mean level[b]	0.1	0.6	2.6	nd
	range	0.1−0.2	0.3−0.9	0.5−9.9	
	% of detection	12.5	16.7	37.5	
total samples (281)	mean level[b]	1.1	8.2	2.7	4.0
	range	0.1−9.6	0.1−213.3	0.1−9.9	4.0
	% of detection	10.3	21.0	13.2	0.4

[a] DDT, *op′*-DDD + *p,p′*-DDD + *o,p′*-DDE + *p,p′*-DDE + *o,p′*-DDT + *p,p′*-DDT. [b] Mean level of positive samples. [c] Not detected.

Of the 21 different OCPs investigated in this study, only 6 were found above their detection limits (therefore, only data for these residues are reported in tables): HCB, γ-HCH (lindane), *o,p′*-DDD, *p,p′*-DDE, *p,p′*-DDT, and β-endosulfan. On the whole, contamination by organochlorines followed the order γ-HCH > DDT > HCB > endosulfan. The order and pattern of contaminant concentrations varied with the type of meal. As expected from the liposolubility of these residues, fatty meals (egg meals > legumes > fish > meat) were the most contaminated items and responsible for 90% of the contamination detected, followed by vegetables, cereal-based foods, potatoes, and fruits.

The frequency of detection of OCP residues, between 0.4 and 21.0%, was considered low. γ-HCH was most

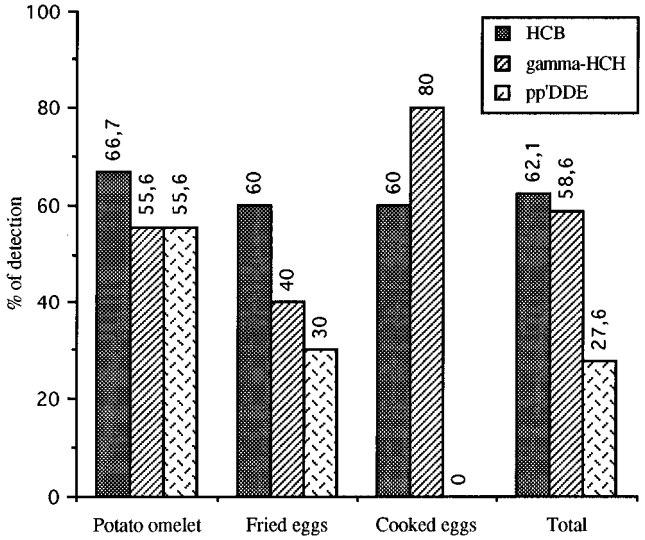

Figure 1. Percentage of detection of organochlorine pesticide residues in egg meals of the Aragonese diet.

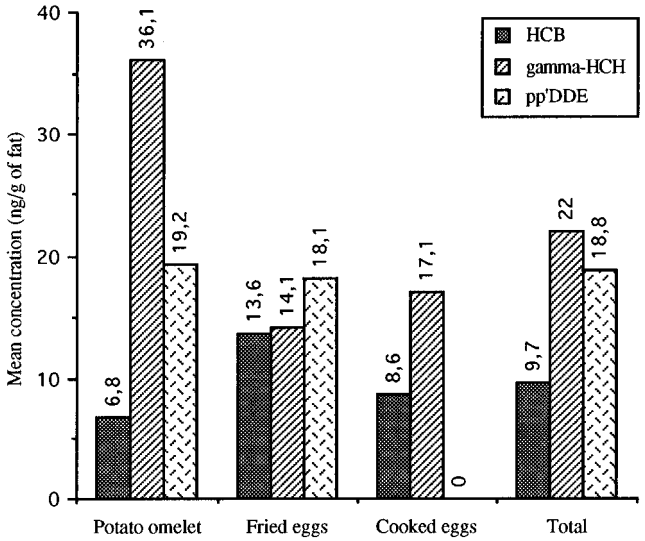

Figure 2. Mean levels (nanograms per gram on a lipid basis) of organochlorine pesticide residues in egg meals of the Aragonese diet.

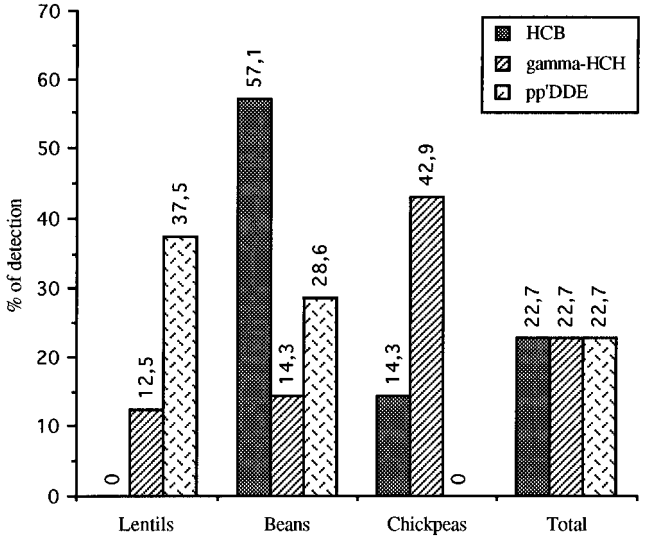

Figure 3. Percentage of detection of organochlorine pesticide residues in legumes of the Aragonese diet.

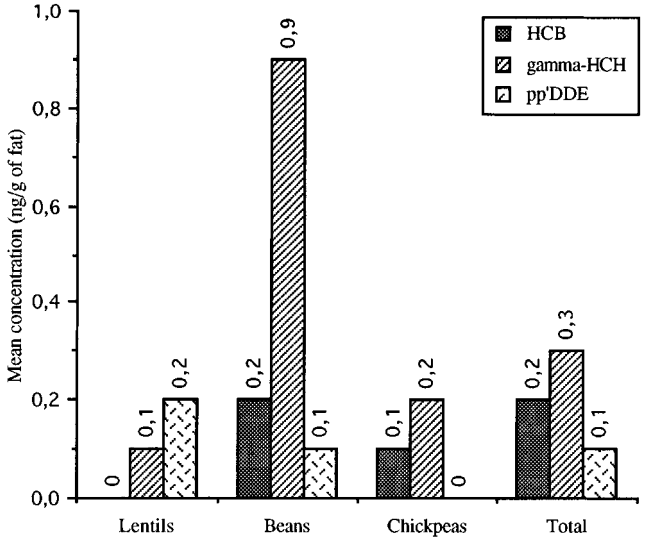

Figure 4. Mean levels (nanograms per gram on a lipid basis) of organochlorine pesticide residues in legumes of the Aragonese diet.

frequently detected (present in 21.0% of samples), followed by DDT (13.2%), mainly in the form of its metabolite *p,p′*-DDE (12.5%), and HCB (10.3%). β-Endosulfan was found only in one vegetable sample (salad). These findings confirm the ubiquitous nature of these highly persistent compounds.

Overall, the residue levels of pesticides determined in the present study were very low (<25.0 ng/g on a wet weight basis) except for γ-HCH residues, which reached a maximum concentration of 213.3 ng/g on a wet weight basis in vegetable-based meals. Mean levels were lower than 10 ng/g on a wet weight basis in all groups for all residues detected except for γ-HCH, which averaged 56.1 ng/g on a wet weight basis in the mentioned group of vegetable-based meals.

HCB contamination was very low, as its mean concentration amounted to 1.1 ng/g on a wet weight basis and its maximum was lower than 10 ng/g on a wet weight basis.

DDT contamination was moderate (2.7 ng/g on a wet weight basis). Among DDT metabolites, *p,p′*-DDE was most frequently detected (12.5%), with mean levels ranging from 0.1 ng/g on a wet weight basis in legumes to 4.3 ng/g on a wet weight basis in potatoes; *o,p′*-DDD and *p,p′*-DDT were detected only in 0.7% of samples. The ratio of DDE to DDT was used to determine whether recent exposure to DDT occurred. In the samples the DDE to DDT ratio was 16, which suggests an earlier usage of DDT, rather than a recent exposure to DDT. The fact that concentrations of DDT were still observed in the samples may be due to long-range atmospheric transport from regions where DDT is still used and to atmospheric deposition.

Figures 1−8 show the contamination pattern of fatty meals. Fish-based meals as a group had higher numbers of DDT metabolites: *p,p′*-DDE (33.3%), *o,p′*-DDD (8.3%), and *p,p′*-DDT (4.2%). *p,p′*-DDE was the main contaminant of this group, especially in fatty fish meals, with a mean level of 143.2 ng/g on a lipid basis. γ-HCH and HCB were found in 12.5 and 16.7% of samples, respectively, and averaged 4.2 and 1.2 ng/g on a lipid basis, respectively.

A high percentage of egg meals was contaminated with residues of HCB (62.1%) and γ-HCH (58.6%) and in a lower proportion with *p,p′*-DDE residues (27.6%). Mean concentrations ranged from 9.7 ng/g on a lipid basis for HCB to 22.0 ng/g on a lipid basis for γ-HCH.

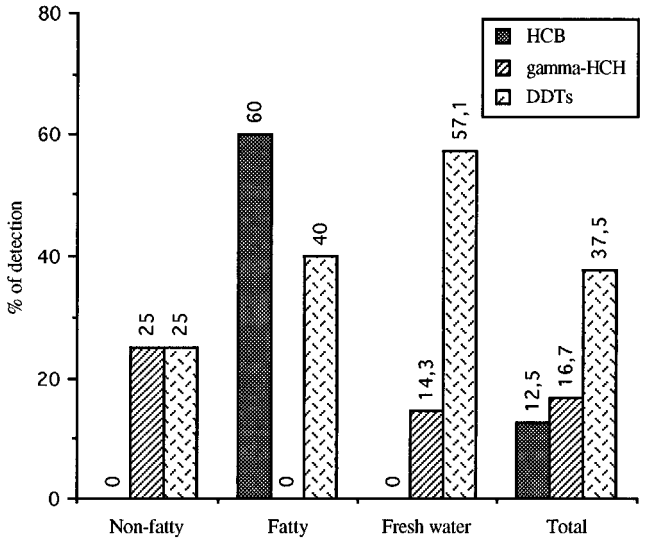

Figure 5. Percentage of detection of organochlorine pesticide residues in fish meals of the Aragonese diet.

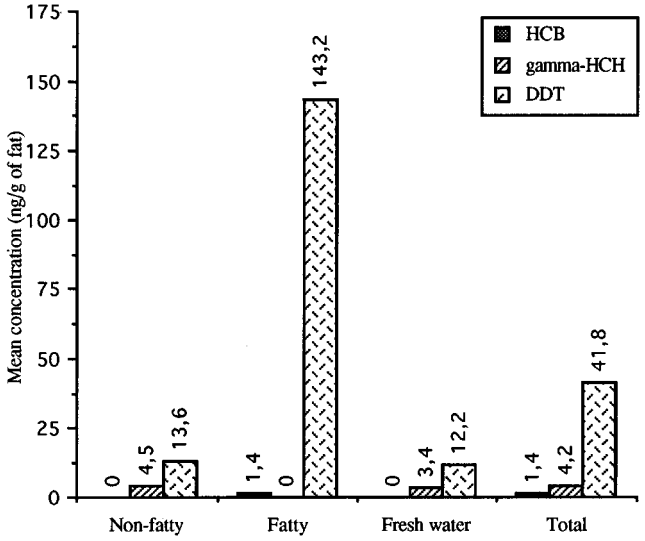

Figure 6. Mean levels (nanograms per gram on a lipid basis) of organochlorine pesticide residues in fish meals of the Aragonese diet.

Meat samples were contaminated with γ-HCH (37.7%) and *p,p′*-DDE (16.9%). Percentages of detection and mean levels of organochlorines were greater for γ-HCH in all groups investigated except for fried lamb samples, in which *p,p′*-DDE was present in 90% with a mean concentration of 16.4 ng/g on a lipid basis.

Legumes were equally contaminated with HCB, γ-HCH, and *p,p′*-DDE. Mean levels did not exceed 1 ng/g on a wet weight basis.

For the remaining groups, it was remarkable that the only presence of residues of HCB was in cereal-based foods, probably related to its fraudulent use as a seed treatment fungicide.

Our results were compared with previous data reported for Spanish foods. Although a proper comparison is extremely difficult given that these studies concerned raw, unwashed items, as previously indicated, the major trends in organochlorine contamination can be envisaged.

In the first total diet study carried out in Spain, Carrasco et al. (1976) reported DDT and HCH as the most frequently detected pesticides with levels from undetectable (<0.001 ppm) to 0.268 ppm. Since then,

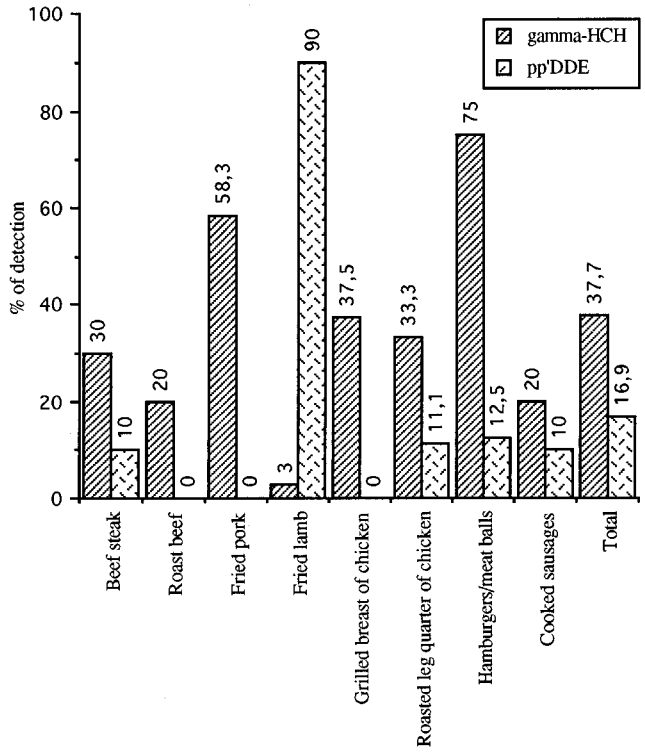

Figure 7. Percentage of detection of organochlorine pesticide residues in meat meals of the Aragonese diet.

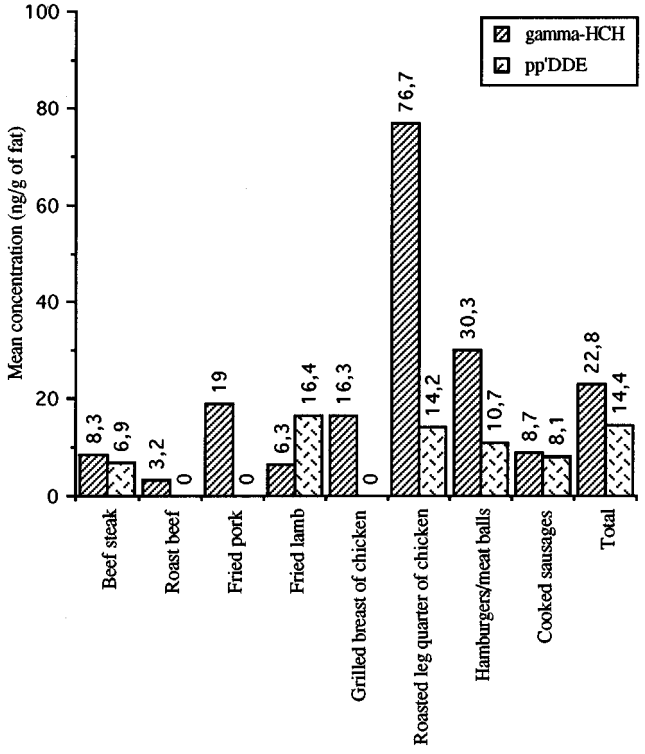

Figure 8. Mean levels (nanograms per gram on a lipid basis) of organochlorine pesticide residues in meat meals of the Aragonese diet.

most authors have reported the high incidence of HCB and HCH (80−100%) and also DDT (50%) residues in a range of Spanish foods: meat and meat products (Pozo et al., 1982; Sánchez-Pérez et al., 1982; Cuñat, 1984; García-Regueiro et al., 1987; Ariño et al., 1992, 1993a,b, 1995; Conchello et al., 1992, 1993a,b; Lázaro et al., 1991; Bayarri et al., 1994; Herrera et al., 1994); fish (Fernández-Aceytuno et al., 1984); and fruits and vegetables

(Arrechedera et al., 1982; Pico et al., 1989). According to these studies, contamination with cyclodienes was quite low.

The percentages of detection of these organochlorines have remained essentially steady during the past decades in Spain, whereas they have declined in most developed countries. In contrast, mean concentrations of organochlorines have decreased over the same period by more than 4-fold, from more than 200 ng/g to less than 50 ng/g, but average levels were still higher than those of industrialized nations (De Vos et al., 1984; Pickston et al., 1985; Wüthrich et al., 1985; Moilanen et al., 1986; Fisher, 1987; van Dokkum and de Vos, 1987; Venant et al., 1989; FDA, 1991; Frank et al., 1990; Cantoni et al., 1991).

Although the pattern of contamination detected in this study is similar to that traditionally observed in Spain, the low incidence of OCPs and the low levels determined confirm this decreasing trend and are comparable to those reported in total diet studies carried out in most developed countries. Such a decline may be probably due to the fact that the use of OCPs was banned according to several European Union Directives in the 1970s and 1980s except for lindane, which is still used for animal husbandry and agricultural treatments.

Urieta et al. (1996), in a total diet study of the Basque country (Spain), reported residues of HCH (α-, β-, and γ-isomers), HCB, dieldrin, endosulfan (α- and β-isomers), DDT, DDD, and DDE at very low levels (always <10 μg/kg on a wet basis), except for lindane. The unexpected high level of lindane residues detected in two samples of the bread group showed that contamination arose from a fraudulent use of this pesticide in a local bakery. No residues were detected in the potato, alcoholic beverage, and nonalcoholic beverage groups.

Tolerance Limits. The results obtained in this study were compared with the recommended tolerance limits set by current European regulations (Directives 86/363/CEE, 93/57/CEE, and 94/29/CEE). The maximum residue limit for HCB is 10 ng/g on a wet weight basis except for meat and eggs, for which it is set to 200 ng/g on a lipid basis. The maximum residue limit for lindane is 100 ng/g on a wet weight basis for legumes and cereals; 1000 ng/g on a wet weight basis for fruits (except for peaches, which is set to 500); and 1000 ng/g on a lipid basis for meat and eggs (except for ovine meat, which is set to 2000). For other vegetables, maximum residue limits range between 500 and 2000 ng/g on a wet weight basis.

Determined levels in the present study were well below the respective maximum residue limits, and none of the samples exceeded the European Union tolerances. However, it should be noted that legal limits are set for raw, unwashed foods. In addition, maximum residue levels are not yet established for OCPs in fish.

Conclusions. It can be inferred from the present study that the levels of OCP residues found in the Aragonese diet analyzed are quite low and present no threat to public health on the basis of current toxicological knowledge. These levels of organochlorines are of the same order as those observed in most developed countries. It is also important to note that the contamination of Spanish foods by persistent chlorinated compounds shows a clear decrease over the past decades. However, it is advisable to continue monitoring the degree of chlorinated contamination of the diet, as the presence of chlorinated residues is still real, although very moderate in extent.

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