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Estimates of Mean Daily Intakes of Persistent Organochlorine Pesticides from Spanish Fatty Foodstuffs

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In many countries pesticide residues in foods are monitored to ensure that public health is not endangered by residue daily intakes in excess of the recommended tolerance levels (van Dokkum and de Vos 1987). In Spain, there is only a total diet study carried out during 1971-72 by Carrasco et al. (1976). In that study, mean daily intakes of 11.5 µg α-HCH, 13.8 µg lindane and 78.4 µg DDTs were calculated. Livestock meat and dairy products were the prime sources of human dietary exposure to organochlorines, since between 60-85% of the mean daily intakes arose from these particular food classes. These percentages are in accordance with the well documented fact that organochlorines predominantly accumulate in the lipid fractions of the human food chain, by which animal fatty foods have become a major route of exposure for humans (Kannan et al. 1992).

Since the current daily intakes of organochlorines in Spain are not known, it was considered necessary to carry out a pesticide survey in several foods that compose an average Spanish diet. To accomplish that: we have determined residues of a list of priority organochlorine compounds in several fatty foodstuffs collected between 1987 to 1990, and prepared in the way in which they would normally be eaten. This study is merely an attempt to estimate the actual intakes, since only a selected number of food classes were investigated and no age-sex group. or seasonal differences were taken into account. In spite of these disadvantages, there are merits to such an approach. Approximate intake figures are available for comparison with toxicologically acceptable intakes and with retrospective studies in Spain and other countries around the world, and they serve to outline the temporal trends in organochlorine contamination that have occurred during the last decades. Also, it may contribute to diminish the consumer's concern about possible health risks involved in the consumption of food products and help to restore confidence in the quality of our food supply.

MATERIALS AND METHODS

In this study, the approach of a limited pesticide survey was followed. The various features of such a study are that it can be used to predict residue intakes by the average person of a particular part of the diet in detail when contamination is mainly restricted to this limited range of foods. Numerous samples of the foods are analyzed for the contaminants of interest, and consumers' intakes are estimated using the consumption and concentration data.

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A total of 359 samples of meat, meat products and sterilized milk were analyzed. The samples of meat and meat products were purchased randomly during 1989 and 1990 in shops and markets or obtained directly from food processing plants and industries, and were prepared according to standard procedures. Seventy-five samples of meat were cooked by either grilling. roasting or pressure cooking. Seventy-six samples of ready-to-eat meat products (including dry-cured fermented sausages, dry-salted and cooked products) were analyzed with no further processing. The data involving dairy products was obtained from 208 samples of sterilized milk collected during 1987-1990 at ordinary commercial outlets from all parts of Spain.

The meat and meat products were analyzed by the method described in detail elsewhere (Conchello et al. 1993; Bayarri et al. 1994), by gas-liquid chromatography using electron capture detector (GLC-ECD) with packed and capillary columns. The analytical procedure for milk samples was carried out as previously reported (Garrido et al. 1994a, 1994b), and organochlorine residues were determined by GLC-ECD. In all samples, the following pesticides were investigated: hexachlorobenzene (HCB). hexachlorocyclohexane (isomers α- and β-, and lindane or γ-isomer of the HCH), dieldrin, DDT and metabolites (DDTs).

Estimates of dietary exposure to organochlorines were calculated from the amount of analyte in the particular food item and the quantity of that food consumed on a daily basis. The data from an official food consumption survey carried out in 1989 by the Spanish Ministry of Agriculture, Fisheries, and Food (MAPA 1990) were used as the basis for the calculation of the daily consumption of the several foods investigated. Thus, the average person in Spain consumed 180 g/day of meat and meat products, and 367 g/day of dairy products. On a daily basis, nearly 25 g fat came from meat and meat products, and 16.5 g fat came from dairy products. From determinations of protein, fat and carbohydrates in the average diet of the average person a mean daily energy intake of 2,819 Kcal was calculated for the year 1989 (MAPA 1990). These figures reflected the pattern of consumption during the late 1980's and therefore they were used to calculate the mean daily intakes of the various chemicals investigated.

RESULTS AND DISCUSSION

The mean concentrations of chlorinated residues found in the samples of each of the food classes investigated are summarized in Table 1 along with the estimated daily intakes of the chemicals. Concentration values are reported as μg/kg on a fat basis, and daily intakes in μg of pesticide per day. Concentrations reported as less than the detection limit (i.e. < 4 μg/kg fat) were treated as half the detection limit. It should be kept in mind that only meat and dairy products were considered herein so the pesticide intakes should be regarded as rough underestimates of the actual values. In Table 1 the FAO/WHO maximum acceptable daily intakes (ADIs) are included for purposes of comparison. For α- and β - isomers of the HCH no ADIs set by FAO/WHO exist so the respective ADIs proposed in Germany by Hapke (1983) were used for interpretation of the data.

Residues of organochlorine compounds were detected in all of the food classes investigated. The fungicide HCB occurred most frequently and was present in 100% of samples of meat and meat products and 58% of milk samples, respectively. Similarly, the insecticide lindane was found in all samples of meat and meat products, whereas only 35% of milk samples showed this residue.

Table 1. Current estimates of daily intakes of organochlorines for the Spanish population (µg/day for an average person of 60 kg body weight). The ADIs (Acceptable Daily Intake) cited here reflect revisions made in 1990.

	Mean pesticide levels (µg/kg fat)			Estimated		
Pesticide	Meat n=75	Meat products n=76	Milk n=208	mean intake µg/day	FAO/WHO ADIs µg/day	%ADI reached
HCB	40	12	19	1.03	36 [a]	2.87%
α-HCH	22	13	43	1.17	300 [b]	0.39%
β-HCH	14	<4	16	0.49	60 [b]	0.82%
Lindane	54	25	12	1.26	480	0.26%
Dieldrin	<4	2.3	26	0.48	6	8.03%
DDTs	14	9	56	1.22	1200	0.10%

[a] FAO/WHO ADI for HCB withdrawn in 1978 (no current ADI for HCB is set)
[b] ADIs for α- and β- isomers of the HCH proposed by Hapke (1983)

The other isomers of the HCH were also more frequently detected in meat samples (α-HCH 76%, β-HCH 40%) than those of milk (30% and 11%, respectively). The DDTs were almost equally detected in meat and meat products samples (88%) than in milk samples (59%). Dieldrin showed major differences in the frequency of occurrence since it was detected in 54% of dairy products but only in 4% of meat and meat products.

The mean daily intakes were far below the respective ADIs. For all chlorinated residues the estimated intakes were less than 1.5 µg/day, with lindane having the highest daily intake that amounted to 1.26 µg/day. This may be due to the fact that lindane is still being used to a limited extent for animal husbandry and agricultural treatments in Spain. The use of the other organochlorines was banned according to European Union Directives in the 1970's and 1980's. For dieldrin and HCB the mean daily intakes reached 8.03% and 2.87% of their respective ADIs; for the remaining compounds much lower percentages were found (0.10% to 0.82% of the ADIs).

Our results were retrospectively compared with data reported by Sanchez-Perez et al. (1982) in a pesticide survey of 969 samples of meat and dairy products carried out by the CNAN, a Spanish national framework of the Ministry of Health and Consume (Table 2). First, the estimated daily intake of HCB (16.58 µg/day) was 16 times greater than that observed in our current study (1.03 µg/day). The intake of 16.58 µg/day amounted to 46.1% of the ADI for HCB. Interestingly, several reports on the high incidence of HCB residues in human adipose tissue and breast milk were reported in Spain at that time (Anonymous, 1990; Gómez-Catalán et al., 1995). The intake of DDTs, 5.22 µg/day, was more than 4 times greater than that of 1.22 µg/day estimated in the current survey. The intake of lindane (1.91 µg/day) has only dropped to 1.26 µg/day in the present study. The intakes calculated for the other HCH isomers have approximately halved in the same period.

Further, we compared our data with the only total diet study ("market basket" approach) published in Spain (Carrasco et al. 1976), in which only data for α-HCH, lindane and DDTs were reported (Table 2). Although a proper comparison is extremely difficult given that the present study is not so comprehensive as that of Carrasco et al., the major trends in organochlorine contamination can be envisaged. Average daily intakes of the HCH isomers have been reduced by 10-fold, whereas that of the DDTs has dramatically dropped by more than 60-fold. No data for dieldrin or HCB were available for comparison.

The number of countries where total diet studies have been performed is relatively small. Some published mean dietary intake figures are given in Table 2. The approximate intake figures for HCB and HCH estimated in the current study were of the same order as those reported in other European countries. The daily intakes of dieldrin and DDTs were at the lower end of the ranges reported in the literature in Europe. Comparing our data with figures reported in the USA, the daily intakes in Spain were higher except for DDTs, which intake in the USA was slightly higher than that estimated in this study.

Table 2. Mean daily intakes µg/day for an average person of 60 kg body weight) of some organochlorine pesticides reported in several developed countries.

Pesticide	Spain This study	Spain 1971-72 *(1)*	Spain 1980-81 *(2)*	Holland 1976-78 *(3)*	Switzerland 1981-83 *(4)*	USA 1990 *(5)*
HCB	1.03	-	16.58	1.0	1.1	0.03
α-HCH	1.17	11.5	-	1.0	-	-
α+β-HCH	1.66	-	3.30	-	1.8	0.04
Lindane	1.26	13.8	1.91	2.0	0.7	0.08
Dieldrin	0.48	-	-	-	0.9	0.10
DDTs	1.22	78.4	5.22	6.0	1.7	1.56

(1) Carrasco et al. 1976; *(2)* Sánchez-Pérez et al. 1982; *(3)* de Vos et al. 1984; *(4)* Wüthrich et al. 1985; *(5)* FDA 1991

Everything considered, we can conclude that the dietary intakes of organochlorines in Spain are of the same order than those observed in most developed countries. For all organochlorines the intakes were well below the existing ADIs, except for dieldrin which intake amounted to 8.03% of the respective ADI. This fact has also been observed by other researchers and is generally attributed to the extremely low ADI (6 µg/day) set up by FAO/WHO. Given the approximate daily intakes for most organochlorines in the order of 1 microgram per day it can be concluded these intakes are probably totally harmless.

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