

Organochlorine pesticide residues in bovine milk from León (Spain)

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Abstract

Residue levels of the organochlorine pesticides (α -HCH, lindane, heptachlor-epoxide, aldrin, endrin, dieldrin, o,p'-TDE, p,p'-TDE, p,p'-DDE and p,p'-DDT) were determined in raw bovine milk and compared with the maximum levels allowed by the European Union (EU) in these foods. The highest incidence percentage of the ten insecticides measured was for lindane, followed by α -HCH and aldrin. Moreover, the highest mean residue level was for α -HCH. None of the samples analyzed exceeded the maximum levels allowed by the EU.

Keywords: Organochlorine residues; Pesticides

1. Introduction

Bovine milk is used as an indicator for measuring the persistence of the chemical substances in agriculture which pollute the environment. Milk quality has frequently been used as a standard in order to undertake legislative decisions concerning the use of pesticides.

Taking into account the special place that milk and its derivative products have in human diet, it is important to know if the detected residue levels are below the maximum levels allowed by the EU.

In this paper, levels of organochlorine pesticide residues were investigated in raw bovine milk. The

samples were obtained in several dairy farms of León, in the North-West of Spain, before industrial processing.

2. Materials and methods

Organochlorine pesticide residues were determined in 39 raw bovine milk samples obtained from several farms located in an eminently agrarian area. All samples were collected in 1992 and stored at -20°C until they were analyzed.

Extraction of organochlorine pesticides was made according to the method described by Suzuki et al. [1]; clean-up of hexane extracts was carried out according to the method provided by Association of Official Analytical Chemists

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(AOAC) [2]. Cleaned-up extracts were completely evaporated under a gentle stream of nitrogen and 1 ml of hexane was added for reconstitution.

Gas chromatography with electron capture detection was utilized for the identification and quantification of residues (Hewlett-Packard with a Ni⁶³ ECD and a computer).

The glass column used was 6 feet long, with an 0.25-inch internal diameter containing a 10% coating of DC-200 on 80–100 mesh Chromosorb W HP (AOAC, 1990) [2]. When necessary, a second column packed with 1.5% of OV-17/1.95% of QF-1 on 100/120 mesh Chromosorb W (AW/OMCS) was used for confirmation of the identity of the organochlorine residues. The operating temperatures were: detector 300°C, inlet 220°C and columns, 180°C (15 min) and 190°C (20 min), with an intermediate temperature increase of 10°C/min. The carrier gas was 5% argon-methane at a flow rate of 21 ml/min and the sample volume injected was 3 μ l.

All solvents were of pesticide residue analysis grade and the purity of every reagent was carefully checked.

The peaks were identified by comparison of retention times with those in corresponding standards: α -HCH, lindane, heptachlor-epoxide, aldrin, endrin, dieldrin, o,p'-TDE, p,p'-TDE, p,p'-DDE and p,p'-DDT (Dr. Ehrenstorfer GmbH, Scharlau). Residue concentrations in the samples were obtained by comparing peak areas from samples with those showed by a mixed standard solution containing all insecticides at the following concentrations (ppm): 0.070 (α -HCH), 0.090 (lindane), 0.062 (heptachlor-epoxide), 0.060 (aldrin), 0.081 (endrin), 0.048 (dieldrin), 0.075 (o,p'-TDE), 0.072 (p,p'-TDE), 0.095 (p,p'-DDE), and 0.165 (p,p'-DDT). Duplicate analysis was performed on all samples.

Recovery from fortified samples of milk were: 91% for α -HCH; 87.5% for lindane; 98.2% for heptachlor-epoxide; 95.3% for aldrin; 90% for dieldrin; 87.5% for endrin; 90% for o,p'-TDE; 84% for p,p'-TDE; 98.1% for p,p'-DDE and 84.5% for p,p'-DDT.

3. Results and discussion

The number of positive samples as well as the in-

Table 1

Mean, range (in ppm/wet weight), incidence (in %) of organochlorine pesticides and number of positive samples in raw bovine milk

	\bar{x} (range)	Incidence (%)	Number of positive samples
α -HCH	0.0028 (0.0015–0.0039)	43.59	17
Lindane	0.0022 (0.001–0.024)	66.67	26
Heptachlor Epoxide	0.0024 (0.0005–0.0086)	20.51	8
Aldrin	0.0037 (0.0005–0.01)	23.10	9
Dieldrin	0.0035 (0.0006–0.012)	17.95	7
Endrin	N.D.*	0	0
o,p'-TDE	0.0008 (0.0002–0.0014)	5.13	2
p,p'-TDE	0.0047 (0.0005–0.017)	12.82	5
p,p'-DDE	0.0050 (0.0005–0.0068)	10.26	4
p,p'-DDT	0.0004 (0.0003–0.0007)	10.26	4

*not detected.

cidence, mean and range values of organochlorine residues in 39 raw bovine milk samples are shown in Table 1. The levels are reported on a wet-weight basis (ppm). The highest incidence percentage of

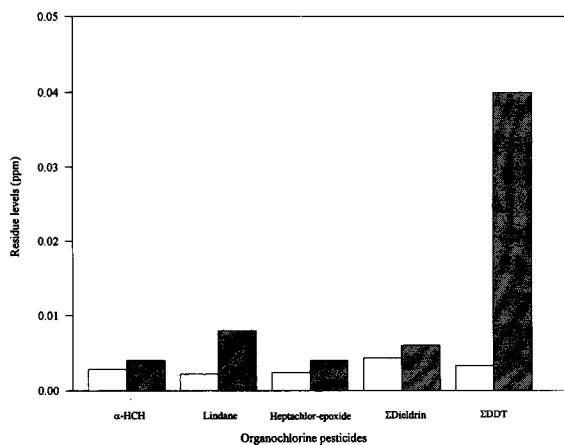


Fig. 1. Average of α -HCH, lindane, heptachlor-epoxide, Dieldrin and DDT detected in bovine raw milk \square compared to EU limits \blacksquare .

the ten insecticides researched was for lindane, 66.7%, followed by α -HCH, 43.6%, and aldrin, 23.1%.

Furthermore, in 61.5% of the samples only two insecticides were present and only one sample contained seven pesticides (2.6% of all samples). Samples containing more than seven compounds were not found. In 30 samples (77%), DDT and related compounds were not found; in only one sample (2.6%), HCHs (α - and γ -isomers) were not detected. The highest mean residue level, 0.005 ppm, was for p,p'-DDE.

In order to compare the results obtained with the maximum levels allowed by the EU in milk, we have included the values of Σ dieldrin (0.0043 ppm) and Σ DDT (0.0033 ppm) in Fig. 1. Σ dieldrin includes aldrin and dieldrin residues. Likewise, Σ DDT refers to the addition of o,p'-TDE, p,p'-TDE and p,p'-DDT.

In comparison with the maximum levels allowed by the EU in whole milk (0.004 ppm for α -HCH; 0.04 ppm for Σ DDT; 0.004 ppm for heptachlor-epoxide; 0.008 ppm for lindane; 0.006 ppm for Σ dieldrin), none of the samples analyzed exceeds the maximum levels established for the pesticides tested.

We have not found any recent references for organochlorine pesticides in bovine milk in Spain; the only work available was carried out 17 years ago. In 1977, Pozo Lora et al. [3], studying pesticide residues in bovine milk collected in the South of Spain, found that the highest incidence percentage was for HCH (α - and β -isomers; 99.2%), followed by p,p'-DDE (97.6%) and heptachlor-epoxide (92.1%). These values were higher than ours, except for lindane whose incidence percentage was 36.8%.

Similar studies in Europe were carried out by Krauthacker et al. [4] who investigated organochlorine pesticides in cow milk and butter samples

obtained between 1985 and 1986 in Croatia. p,p'-DDE was present in all samples, followed by γ -HCH, α -HCH and p,p'-DDT.

Cantoni et al. [5] monitored organochlorine pesticides in several foods available in Italy (ie. milk, cream, butter) and found that milk and cream were the most contaminated foods, although residue levels were within legal tolerances.

Recently, Hernández et al. [6] found a high incidence for DDT and related compounds (100%) and for α -HCH (68.6%) in human breast milk collected in Madrid, Spain. However, the incidence percentage in this study was very low for DDT and related compounds (23.1%) and it was also lower for α -HCH (43.6%).

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