# Technical News Features.

# Changing Practices and Changing Residues in Animal Fats and Plant Oils 1890-1980<sup>1</sup>

R. FRANK, Agricultural Laboratory Service, Ontario Ministry of Agriculture and Food, Guelph, Ontario N1G 2R1, Canada

#### **SUMMARY**

Following World War II and the movement of people from the rural to the urban environment, producers of food have become increasingly dependent on organic chemical substances to control pests affecting the production of food. Because of scientific evidence and public concern, constant changes have occurred in the types of pesticides being used over the last three decades.

The persistent organochlorines which accumulated in fats and oils were removed in the early 1970s. Since that time, residues in animal fats and plant oils have declined markedly and almost disappeared in most cases. No early evidence was found in Ontario that these compounds accumulated in plant oils and so no indepth study was undertaken

With the threat of biological resistance ever looming larger, Integrated Pest Management programs have been investigated and are being implemented to reduce the dependence on chemical pesticides. Current pesticides rarely appear in oil and fats and those that do are controlled below the MRL by following the days to slaughter for livestock and the days to harvest for crops.

In general, pesticide residues in fats and oils produced in the Province of Ontario have declined during the last decade and are continuing to decline. Monitoring for all pesticide residues will continue to ensure that this state of affairs is maintained.

# INTRODUCTION

During the twentieth century, dramatic changes have been observed in the production of food—both to meet the increasing world population and to raise the standard of nutrition and cater to the palate of the more advanced nations. I have chosen the Province of Ontario to illustrate my talk for two reasons: the data bases available are substantial and over the past 25 years I have been intimately involved in these changes. I am sure that the dynamic changes to Ontario agriculture are observable in other provinces of Canada, or the 52 states in the U.S. This talk is intended to introduce the session topic entitled lipophilic

1Presented at the AOCS meeting, Toronto, May 1982.

xenobiotics and pave the way for subsequent presentations today.

## Agricultural Industry, Ontario 1921-76

Although agricultural statistics within the Province are taken every year, national census figures are collected only every five years. Since the latter are considered more reliable and since the 1981 figures are not yet available, the period of review is taken as 1921-1976; however, because of major changes after that date, the 1980 provincial figures are added where available. Between 1921 and 1976, the Ontario population rose from 2.93 to 8.26 million; an almost tripling in 55 years. At the same time the farm population declined from 1.20 to 0.34 million; a drop of almost one quarter of the former level. The farm population therefore declined from 42 to 4.1% of the Ontario population.

This mass exodus of people off the land and to the large urban centres created a cadre of scientists and technicians, who turned their endeavors to improving food production back on the farm, thereby enabling the shrinking farm force to manage increasingly larger farms with greater efficiency, higher yields and better farm returns. Among the urban-based agriculturally orientated businesses that arose were the Fertilizer Industry, the Agricultural Chemicals Industry, the Farm Machinery Industry, the Food Processing Industry and many others. Taking into account all these allied industries, the actual percentage of people involved in the total Food Industry has changed little over this period and is reported to be about 40% of the population.

During this same period, the area of land in concensus farms declined from 9.2 to 6.3 million hectares, while the gross value of produce rose from almost \$0.5 to \$2.8 billion. The value had reached a level of \$4.5 billion by 1980.

#### **Advances in Food Production**

The technical advances have created convulsive waves through the agricultural industry. Since 1921, the area under row crops has risen from 12% of the improved land to become the largest component at 44%. These changes have been accompanied by the appearance of a

large farm segment devoted to a monoculture type of agriculture, with few if any livestock. The shift in management has recently been revealed as not sustainable and so the trend is back to more rotations, more permanent cover and more livestock.

The growth in the hectorage of crops like corn and soybeans has progressively multiplied over the last 50 years. Soybeans were not grown in Ontario until the forties, and their cultivation since that time has expanded to 283,000 hectares in 1980 and now represents a sizeable industry. This increase in row crop production has occurred at the expense of cereal and pasture production.

There have been many urban-based inputs to agriculture that have permitted this increase in land being planted to row crops and at the same time has facilitated substantial increases in yield per unit area. The pertinent technical advances introduced during this period that have given rise to these increments in productivity have been (a) improved cultivars; (b) development of mechanization for all farm operators; (c) chemical control of pests; and (d) improved plant nutrition. This is well illustrated by the figures in potato production where the land area devoted to the crop has shrunk by 76%, while the yield per unit area has increased ninefold and the overall production has increased 216%. Each technical advance is a topic in itself; however, today I wish to concentrate on the chemical energic inputs into this production scenario.

#### Agricultural Chemicals

One of the major reasons for the phenomenal increase in yield per unit area has been the development of agricultural chemicals. Introductions have been made to control all manner of pests from the smallest bacteria to the tallest weed, from the airborne fungal spores to the most voracious insect. Over time, this has allowed one person to build a farm business to handle a continuously increasing hectorage of intensively produced crops. As an example, instead of having continually to cultivate and hoe an intensively produced row crop throughout the growing season, a producer can apply a herbicide treatment but once to control his annual weed problems. This state of affairs did not occur overnight but has evolved over the past 100 years following the first global commercial pesticide application in France when copper sulfate was used on grapes to control mildew. Since then, many advances and retreats have been experienced in the development of chemical aids to food production. A review of this story is the thesis of my main presentation.

#### Pre-World War II

The first pesticide applications in Ontario occurred in 1885, when unbearable losses of apples to insects and diseases prompted growers to use the inorganics copper aceto-arsenite and copper sulfate to relieve the problem. For the next 60 years, i.e., up to the Second World War, growers were dependent on a number of inorganic and botanically derived substances that had a limited usefulness in controlling a small range of pests. Many of these compounds left residues in food of unknown impact to the consumer and many accumulated in soils, with likewise unknown impact on the production of food or on the environment.

# Post-World War II

Immediately after World War II, many organic substances became available for food production. These included the organochlorine insecticides, like DDT, dieldrin and heptachlor and the chlorophenoxy herbicides, like 2,4-D and MCPA. Within the next two decades, a wide variety of chemical groups were researched and active compounds were found that could be used to control pests on a wide range of food commodities. The notable groups were the organophosphorus insecticides, the carbamate herbicides, insecticides and fungicides, the triazine herbicides, and the organochlorine fungicides. More recently, the pyrethroid insecticides have been added.

Chemical energy has now taken the place of manual and mechanical energy, as being the most efficient way of protecting food crops from the ravages of pests. As examples, 96% of beans and 90% of corn crops are treated with herbicides. This has not come about without considerable developmental work and legislative controls. The major Federal and Provincial acts and regulations are discussed below.

### **LEGISLATIVE ACTIONS**

No pesticide can be offered for sale without first passing the registration process. The lead agent in this procedure is Agriculture Canada; however, all pesticides must first meet prerequisites on toxicology laid down by Health & Welfare Canada, and environmental assessment by Environment Canada, and Fisheries and Oceans Canada. The basic requirements involve safeguarding the health of the user, the consumer, the environment, fish and wildlife. The first Pest Control Products Act appeared in the late 1920s and was revised in 1954, and 1971, and thereafter at regular intervals as the need has arisen.

Maximum residue limits (MRL) of each registered pesticide are set forth by Health & Welfare Canada, for both domestically produced and imported foods. The Food and Drug Act first appeared in 1929, was updated to its present format in 1954 and has been revised on a regular basis since then.

At the Ontario level, the Pesticide Act and Regulations were removed from the Public Health Act in 1956 and expanded into a separate act, administered by Environment, Ontario. Since that time, frequent and major changes have occurred in a constantly upgrading process. Provisions in the Act now control the licencing of vendors and applicators of pesticides. The Environmental Protection Act was introduced in 1971, making it an offence to allow a pesticide to be dispersed beyond the borders of the intended target area in such a manner as to be a nuisance or hazard to people or the environment. Many other acts have sections dealing with pesticides from manufacture to end user, from the quality of food to the quality of the environment.

Much of the legislation has resulted from both scientific findings and public concerns. Changes have come about in three ground swells of public pressure or overwhelming scientific evidence over the past 30 years and has been tempered by a growing fourth concern of biological resistance.

Food residues. By the mid-1950s, the use of persistent organochlorine as direct treatment to domestic animals and as topical sprays to animal feed resulted in high levels of residues in animal fats and plant oils. The legislative actions taken removed these types of registered uses in an attempt to stop further accumulations. At the same time the Federal Government established MRL in food for the persistent organochlorines and the principle of acceptable daily intakes (ADI) were developed to safeguard the consumer. These steps set in motion the need for the first toxicology data as a prelude to assessing chronic effects.

Environmental impacts. In the sixties, a growing mass of in-

formation revealed that the persistent organochlorine insecticides were being stored in the fat of nontarget organisms (both domestic and wild mammals and birds) and creating problems in reproduction. The eggshell-thinning phenomenon appeared among raptors and the death of certain mammals and birds occurred in the wild and especially among those at the top of the food chain. Environment Departments were established and the legislative actions that followed ultimately removed all uses of the persistent organochlorines (Table I).

Occupational health hazards. During the seventies, a growing public concern surfaced over the hazards of using pesticides in respect to the user and bystander. New testing procedures and protocols are being developed to improve the assessment of risks to humans and, in particular, side effects and long term effects. Hence, tests for teragenicity, mutagenicity, and oncongenicity are being researched to find suitable systems that can be used in the assessment process.

Not only are pesticides involved in this debate, but also included are industrial compounds like PCB, mirex, and HCB that have biocidal activities. Recently, legislate actions have involved the appearance of worker protection under Occupational Health Acts and the development of standards and criteria for handling toxic substances.

Biological resistance. As a thread weaving through the last 30 years of health and environmental concern, biological resistance has increased at an alarming rate. In some quarters, both agricultural and health officials are highly concerned

about this problem and a flurry of activity has developed to understand the phenomenon better and if possible to slow down the rate of appearance of resident genotypes.

More than 350 species of insects are resistant to one or more insecticides, which previously provided good control. About 250 of these species are agricultural pests. The phenomenon is not confined to insects but occurs among fungal diseases, weeds, rodents and other problem pests. Table II gives a list of some of resistant species and the chemicals involved in Ontario.

#### **CHANGES IN PESTICIDES**

The above four concerns have created dynamic and continuing change in the use of chemicals and the types of pesticides applied. This is well illustrated in the changes by chemical group of those insecticides used in the production of tobacco between 1955 and 1980 (Table III). In 1955, seven persistent organochlorine pesticides were in general use in tobacco production; however, by 1975, none were being applied. To illustrate the impact of these changes on residues in food and the environment, the use of DDT in 1971 is compared with the use of chlorpyrifos in 1978, both applied for cutworm control.

By 1971, a large reservoir of DDT (325,000 kg) had been built up in tobacco soils from 11 years of application amounting to 883,000 kg; small quantities appeared in crops grown on these soils (e.g., 34 kg) and in tobacco (53 kg): an even smaller amount was found in hay crops (4 kg) and woodlots (Table IV). The residue in hay and corn silage even gave rise to smaller amounts in the milk (0.5 kg) and

TABLE I
Legislative Removal of Organochlorine Insecticides in Ontario and Canada

Date	Pesticide	Action		
1969	Aldrin, dieldrin, heptachlor	Agricultural uses cancelled (Ontario)		
1970	DDT, TDE	All except three uses cancelled (Ontario)		
	Endrin	Many uses cancelled (Ontario)		
1971	DDT	Major remaining uses cancelled (Ontario)		
1972	Toxaphene	Rescinded, prescription use (Canada and Ontario)		
1973	DDT '	All agricultural uses cancelled (Ontario)		
1974	HCB	Registration lapsed (Canada)		
1977-78	Endosulfan, Chlordane	Registration lapsed (Canada) Major uses rescinded (Canada)		

TABLE II

Pests of Vegetables Known to Have Become Resistant in Ontario over the Last 20 Years

Pest		Pesticide
Insects	Cabbage maggot Onion maggot	Aldrin, diazinon, heptachlor, thionazin Aldrin, dieldrin, heptachlor, ethion, fensulfothion, carbofuran
	Seed maggot Carrot nest fly Colorado potato beetle	Aldrin, dieldrin Aldrin, dieldrin, heptachlor Endosulfan
Diseases	Apple scab Botrytis Powdery mildew	Dodine, benomyl, thiophanate-methyl Benomyl Benomyl
Mites	European red mite	Dicofol, ovatran, TEPP, mevinphos, dimethoate, azinphosmethyl, ethion genite
	Two spotted spider mite	Dicofol, parathion, malathion, TEPP, mevinphos, dimethoate, azinphosmethyl, ethion
Weeds	Wild carrot Lambsquarters and pigweed	2,4,5-T Atrazine

TABLE III

Changes in Chemical Groups of Pesticides Recommended for Use for Insect Control in Tobacco Production, Ontario, 1955-1980

	Number per group					
Pesticide group	1955	1960	1965	1970	1975	1980
Organochlorine						
long persistence	7	6	3	1	0	0
short persistence	1	1	2	2	1	1
Organophosphorus	1	2	2	5	6	7
Carbamates	0	0	1	1	ī	3
Biologicals	0	0	1	1	1	1
Pyrethroids	0	0	0	0	0	2
Total	9	9	9	10	9	14

TABLE IV

Movement of DDT in the Tobacco Belt,
Following Application to Tobacco Soils

Year	Item	Area (hectares)	DDT (kg)	Percentage
1961-71	Application	216,500	883,000	
1971	All soils	42,000	$325,000^{a}$	37
1971	Tobacco, cured leaf	16,100	53	$6 \times 10^{-3}$
1971	Corn silage	6,000	34	$4 \times 10^{-3}$
1971	Нау	12,000	4.1	$5 \times 10^{-4}$
		Volume kg		
1971	Milk	56 × 10 <sup>6</sup>	0.5	6 × 10 <sup>-5</sup>
	Beef	6 × 10 <sup>6</sup>	0.5	6 × 10 <sup>-5</sup>
		Discharge m <sup>3</sup>		
1971	All streams	$355 \times 10^6$	5.3	6 × 10 <sup>-4</sup>

<sup>&</sup>lt;sup>a</sup>In 1971, kg was applied.

beef (0.5 kg) produced from the area. Soil erosion carried contaminated soil containing 5.3 kg  $\Sigma$ DDT into streams and thence to Lake Eric. This resulted in aquatic fauna accumulating residues in their fatty tissues, discussed later in the paper. After DDT was removed from use in 1971, soil levels have declined slowly with a half life of 8-10 years (Fig. 1).

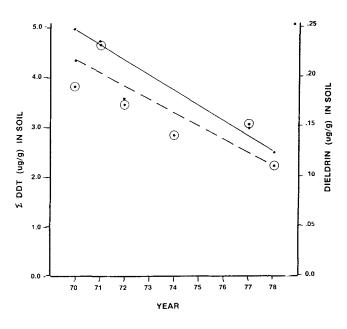


FIG. 1. Decline in  $\Sigma DDT$  and dieldrin residues in soils collected from the tobacco belt, Ontario, 1970-78.

Chlorpyrifos replaced DDT in 1971. In 1978, records were compiled to determine the fate of this insecticide in the tobacco belt. The annual application to 10,900 ha was 22,900 kg. By the end of the growing season, this amount had been reduced to only 59 kg; a loss of 99.7%. Cured tobacco contained only 1-2 µg/kg in the dried tissue, amounting to 84 g in the entire crop. Measurements of chlorpyrifos in stream water between 1975 and 1977 permitted an estimate of the annual losses to Lake Erie to be made and the amount turned out to be less than 1 kg/annum. This loss occurred when storm water runoff carried chlorpyrifos from treated fields to streams within a few days of application. At no other times in the year could chlorpyrifos removal be detected.

#### Pesticides used on Domestic Animals and Residue in Fat

The list of pesticides used on livestock in Ontario to control insect pests appears in Table V, along with the

TABLE V
Pesticides Used on Domestic Animals in Ontario and Maximum Residue Levels Permitted, 1981

Pesticide			Maximum residue level	Withdrawal
Group	Name	Meat species treated	(mg/kg)	(days)
Organochlorine	Lindane	Bovine, porcine	2.0 (Fat)	30
_	Methoxychlor	Bovine	3.0 (Fat)	7
	Toxaphene <sup>a</sup>	Porcine	7.0 (Fat)	
Organophosphorus	Coumaphos	Avian, bovine, porcine	0.5 (Fat)	7
	Crotoxyphos	Bovine	< 0.1 (Meat) <sup>D</sup>	7
	Dichlorvos	Bovine	< 0.1 (Meat)	0
	Fenthion	Bovine, porcine	< 0.1 (Meat)	35
	Malathion	Avian, bovine, porcine	< 0.1 (Meat)	
	Ronnel	Bovine, porcine	7.5 (Fat)	7
	Trichlorfon	Bovine	< 0.1 (Meat)	21
Others	Carbaryl	Avian, bovine, porcine	5.0  (Avian) < 0.1  (Meat)	7
	Rotenone	Bovine	< 0.1 (Meat)	

<sup>&</sup>lt;sup>a</sup>Restricted to prescription use by veterinarians, 1971.

b<0.1 mg/kg is a negligible residue.

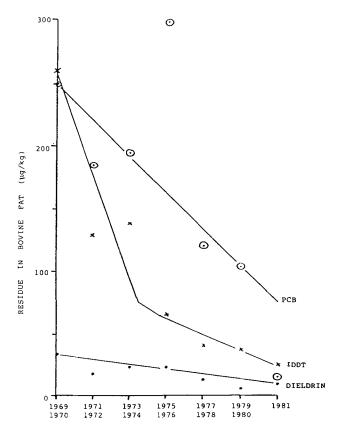


FIG. 2. Residue of  $\Sigma DDT$ , dieldrin and PCB in bovine fats collected between 1969 and 1981 in Ontario.

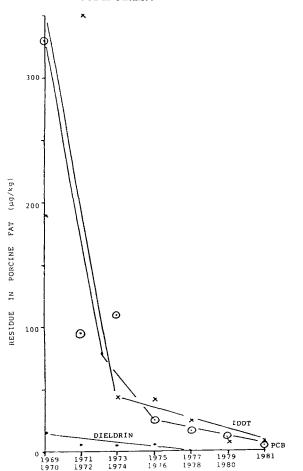


FIG. 3. Residues of  $\Sigma DDT$ , dieldrin and PCB in porcine fats collected between 1969 and 1981 in Ontario.

MRL permitted in fat and meat. Figures 2-4 and Tables VI-VII reveal that the main residues over the last decade have been organochlorine compounds. DDT and dieldrin, as was noted in Table I, were cancelled for all agricultural uses in 1971 and 1969, respectively. Trace amounts still appeared in animal fats up to 1980, presumably coming from uptake of these compounds into animal feeds or from splash onto crops during rainfall. PCB, an industrial fire-retarding chemical, while having no agricultural use, has been used in rural areas to fill electrical transformers; and thereafter the spent oil has been deposited on earthen roads to reduce aerosol dusts. In addition, inputs of PCB into the rural environment have been shown to come from aerial fallout, especially in rain, and this appeared greater in the early 1970s than now.

It is evident from Figures 2-4 that, over the last ten-year period, organochlorine residues have markedly declined in avian, porcine and bovine fat, and negligible residue levels are being approached. Half life disappearance of 2-5 years can be observed for the disappearance of these residues from fat. Tables VI and VII include the residues of other pesticides. Chlordane was used throughout the study period to control soil insects in the production of animal feed. Lindane and the organophosphorus insecticides could have been used directly on live animals to control insects. It should be noted that the residue levels are given in  $\mu g/kg$ ; that is, they fall well below the negligible levels generally designated 100  $\mu g/kg$ .

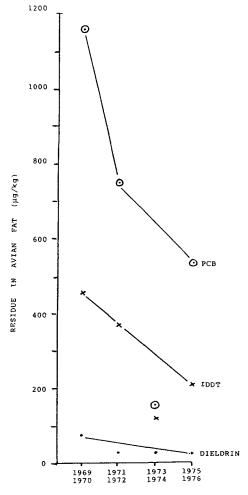


FIG. 4. Residues of  $\Sigma$ DDT, dieldrin and PCB in avian fats collected between 1969 and 1970 in Ontario.

TABLE VI

Residues of Pesticides Being Used During the Study Period in the Production of Animal Feed or Applied Directly to Beef-Producing Animals

Years		Mean content in extractable bovine fat $(\mu g/kg)^{c}$				
	Number of carcasses	Chlordane plus heptachlor epoxide	Lindane	Organophosphorus		
1969-70	835	1	16	_		
1971-72 <sup>a</sup>	143	< 1	2	_		
1973-74	198	< 1	< 1	ND		
1975-76	53	4	32	ND		
1977-78	50	4	< 1	ND		
1979-80	213	6	< 1	ND		
1981	330	7	8	18 <sup>b</sup>		

<sup>&</sup>lt;sup>a</sup>Traces of toxaphene.

TABLE VII
Incidence and Mean Residue of Pesticides and Industrial
Chemicals in the Milk Supply of Ontario, 1977

Compounds		Incidence in samples <sup>a</sup> (%)	Residue in fat (μg/kg) <sup>b</sup> Mean ± SD
Pesticide	Aldrin	0	_
	Chlorodane and metabolites	4.2	< 1
	$\Sigma$ DDT	97	15 ± 20
	Dieldrin	99	11 ± 5
	Endosulfan plus metabolite	7.5	1.8
	Endrin	0	_
	Heptachlor plus epoxide	99	4 ± 2
	Lindane	0	_
	Methoxychlor	0	_
	Organophosphorus	0	_
	Polychlorophenols	0	_
Industrials	НСВ	68	2 ± 3
	Mirex	0	
	PBB	0	_
	PCB	95	35 ± 30

<sup>&</sup>lt;sup>a</sup>Sampling of 308 bulk transporters.

Figure 5 shows a similar trend in the disappearance for organochlorine residues in milk between 1967 and 1977.

### Pesticide Residues and Plant Production

Analysis of fruits and vegetables over the last two years has revealed no residues of the persistant organochlorines, i.e., DDT, dieldrin, heptachlor, etc., as being present in produce offered for sale to the consumer. Only extremely low residues of a few pesticides used were found in the food commodities surveyed. Two oil-producing crops (soybean and corn) have been checked, but no residues have been detected.

Tobacco is the only crop where organochlorine residues have been found from past uses. In the tobacco belt, there is still a substantial reservoir of persistent organochlorine pesticides present in the soil, and these appear to be taken up by plants and to occur in the cured leaf.

# Pesticide Residue in the Environment

Organochlorine residues in lake trout caught in Lake Simcoe, Ontario, have been followed over the last decade. DDT residues have declined markedly when two weight classes were compared (i.e., above and below 4 kg). Following the removal of DDT in 1970 and 1971, residues in the smaller sized lake trout declined to a new level, one-tenth the former level, but had not changed for the larger weight class. By 1980 both classes had declined to the reduced residue level. The decline in PCB residues was evident for the younger age class, but not the older age class.

Residues in yellow perch and coho salmon caught in Lake Erie in 1970 and 1975-76 reveal a marked decline in all three major organochlorine compounds. This was particularly evident with coho salmon, a carnivorous fish feeding at the top of the tropic food chain.

bFenthion.

<sup>&</sup>lt;sup>c</sup>Negligible levels would be 100  $\mu$ g/kg.

<sup>&</sup>lt;sup>b</sup>Mean residue based on all samples analyzed.

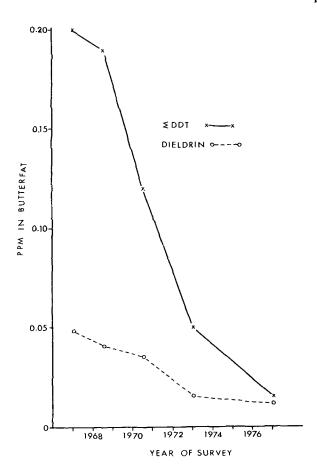


FIG. 5. Decline in  $\Sigma DDT$  and dieldrin residues in cows' milk collected in surveys 1967, 1968, 1970, 1973, and 1977.

# TABLE VIII Residue of Organochlorine in Human Fat and Human Milk

	Content in extractable fats (mg/kg)						
Year	ΣDDT	Dieldrin	Heptachlor epoxide	Chlordine	нсв	PCB	
Human adipo	se tissue						
1969-70	7.6	0.17	ND	NA	NA	1.2	
1973-74	3.7	0.09	ND	NA	0.12	2.3	
1978-79	6.3	0.10	0.06	0.09	0.03	2.5	
Human milk							
1969-70	3.5	0.09	ND	NA	NA	1.0	
1973-74	1.4	0.04	0.04	NA	0.10	1.2	
1978-79	0.7	0.01	0.01	ND	0.01	0,5	

NA: not analyzed

### Pesticide in Residues in Human Tissues

The final assessment of the control of organochlorine residues comes when human tissues are analysed. A monitoring program has been in process in Ontario over the last 13 years and the findings appear in Table VIII. In general, residues in human adipose tissues appear to have reached a plateau in 1979, and are either declining slowly or still rising slightly. On the other hand, organochlorines in human milk declined markedly following their removal from use and this decline has continued over the last decade,