Persistent Organochlorine Residues in Foodstuffs from India and Their Implications on Human Dietary Exposure

Kurunthachalam Kannan,† Shinsuke Tanabe,*,† Aramandla Ramesh,† Annamalai Subramanian,‡ and Ryo Tatsukawa†

Department of Environment Conservation, Ehime University, Tarumi 3-5-7, Matsuyama 790, Japan, and Centre of Advanced Study in Marine Biology, Annamalai University, Parangipettai 608 502, Tamil Nadu, India

Foodstuffs collected from different regions in India were analyzed for the presence of HCH (BHC), DDT, HCB, aldrin, dieldrin, heptachlor, and PCBs. Significantly high levels of food contamination with HCH, DDT, aldrin, and dieldrin were evident throughout India. Dairy products and livestock meat are the prime sources of human dietary exposure to these chemicals. Concentrations of these organochlorine compounds in a few dairy products were above the maximum residue limits (MRLs) set forth by the FAO/WHO as well as the Ministry of Health of the Indian government. The average daily intakes of HCH and DDT by Indians were estimated to be 115 and 48 μg/person, respectively, which were higher than those observed in most of the developed nations. The dietary intakes of aldrin and dieldrin exceeded the acceptable daily intake (ADI) recommended by the FAO/WHO. Food pollution and dietary intakes of PCBs, HCB, and heptachlor were relatively low in India.

INTRODUCTION

India is now both the largest manufacturer and consumer of pesticides in southern Asia. Despite the proliferation of different types of pesticides, organochlorines such as HCH (1,2,3,4,5,6-hexachlorocyclohexane) and DDT [1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane] still account for two-thirds of the total consumption in the country because of their low cost and versatility in action against various pests. Since these compounds are highly persistent and strongly lipophilic, they tend to accumulate in body fats including breast milk (Yakushiji, 1988). In spite of their toxicity and adverse human health effects, many developing nations continue to use these chemicals to combat the pests of agricultural and public health importance.

A recent investigation by Tanabe et al. (1990) revealed the existence of considerably higher levels of HCH and DDT, viz., 6200 and 1200 ng/g of fat weight, respectively, in human breast milk collected from southern India. They also detected PCBs, a toxic industrial organochlorine compound, in breast milk although at lower levels. Similarly, PCBs have been detected in human adipose tissue and blood from India (Rao and Banerji, 1988, 1989). These observations suggest that the PCBs are widespread even in developing countries.

A few studies indicated previously the contamination of Indian food and feeds by HCH and DDT (Noronha et al., 1980; Dikshith et al., 1989a,b; Battu et al., 1989; Kaphalia et al., 1990). These works were concerned mainly with HCH and DDT in limited cases of foodstuffs from a few locations. No comprehensive information is available covering a wide variety of foodstuffs from different regions in the country. Besides, food pollution by PCBs, HCB, aldrin, dieldrin, and heptachlor is not clearly known. Hence, the present investigation was undertaken to monitor the levels of HCH, DDT, PCBs, HCB, aldrin, dieldrin, and heptachlor in foodstuffs collected from different regions in India. An attempt was also made to estimate the amount of dietary intake of organochlorines by Indians.

MATERIALS AND METHODS

Sampling Locations. Raw food samples were collected during the period from December 13 to 29, 1989, in New Delhi, Bombay, Calcutta, Madras, Chidambaram, and Parangipettai (formerly known as Porto Novo). The first four locations mentioned are metropolitan industrial cities located in the northern, western, eastern, and southern portions of India, respectively, while Chidambaram and Parangipettai are suburban and agricultural areas situated about 250 km south of Madras.

Samples. Chicken (breast muscle and liver), mutton (goat meat and liver), and pork (meat and fat) were collected from slaughterhouses in the respective locations. Fish samples representing different species, namely, Indian mackerel (*Rastrelliger kanagurta*), jawfish (*Otolithus* sp.), catfish (*Clarias* sp., *Arius arius*, and *Arius maculatus*), sciaenid fish (*Protonibea diacanthus*), scombrid fish (*Scomberoides* sp.), catla (*Catla catla*), silver pomfret (*Pampus argenteus*), perch (*Lates calcarifer*), threadfins (*Eleutheronema tetradactylum*), blackbream (*Acanthopagrus* sp.), striped mullet (*Mugil cephalus*), pearl spot (*Etroplus suratensis*), seer fish (*Scomberomorus commersoni*), and prawn (pooled sample of *Metapenaeus monoceros*, *Metapenaeus dobsoni*, and *Penaeus indicus*) were obtained from fish markets. Only the edible portion was used for analysis. Cereals, pulses (chickpea, black gram, green gram, and green peas), oils (sunflower oil, groundnut oil, gingelly oil, coconut oil, and palm oil), spices (fennel, coriander, cumin, pepper, and chilies), and butter were purchased from retail grocery stores at random. Cow and buffalo milk were collected fresh from farmer's holdings. All of the samples except grains, oils, and butter were preserved in 10% formalin and transported to Japan for analysis. Samples were then stored in a refrigerator (4 °C) prior to analysis.

Chemical Analysis. Organochlorines were analyzed according to the method described by Tanabe et al. (1991). Briefly, the method involved extraction with mixed solvents of diethyl ether (300 mL) and hexane (100 mL) using a Soxhlet apparatus. Fat contents were measured from concentrated aliquots of these extracts. The remaining extracts were then transferred to a glass column packed with 20 g of Florisil (Floridin Co.) followed by elution with acetonitrile containing 20% hexane-washed water. In the case of oil and butter samples, a quantity of 1 g was dissolved in 5–7 mL of hexane and taken directly to a Florisil dry column. The eluate from the Florisil column was collected in a separatory funnel containing 100 mL of hexane and 600 mL of hexane-

* To whom correspondence should be addressed.
† Ehime University.
‡ Annamalai University.

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Table I. Concentrations of Organochlorine Pesticides and Polychlorinated Biphenyls (ng/g of Wet Wt) in Foodstuffs from Different Locations in India

food item	n	PCBs	ΣHCH[a]	γ-HCH	ΣDDT[b]	HCB	aldrin	dieldrin	heptachlor
cereals	3	0.45	35	4.8	3.5	0.03	1.3	0.75	0.08
		(0.21–0.64)[c]	(27–39)	(3.9–5.8)	(1.7–5.4)	(0.01–0.04)	(0.17–2.3)	(0.28–1.4)	(0.04–0.11)
pulses	4	1.2	420	34	20	0.07	1.3	2.1	0.09
		(0.11–2.9)	(5.4–1600)	(1.1–130)	(1.1–40)	(0.02–0.16)	(0.80–1.6)	(0.82–3.3)	(<0.01–0.27)
spices	5	3.0	210	35	62	0.22	2.1	20	0.46
		(0.09–9.8)	(83–410)	(13–71)	(7.5–220)	(<0.01–0.54)	(<0.1–8.4)	(<0.1–41)	(0.13–1.1)
oils	5	11	220	35	21	1.5	19	24	0.45
		(2.6–15)	(6.9–480)	(1.2–100)	(1.8–57)	(0.09–2.8)	(<0.1–47)	(<0.1–47)	(0.08–1.6)
milk	10	0.52	180	4.2	1.4	0.03	0.02	0.09	<0.01
		(0.03–1.7)	(82–490)	(0.60–18)	(0.17–5.2)	(0.01–0.10)	(<0.01–0.03)	(0.01–0.17)	
butter	4	6.0	2800	160	1400	1.7	42	740	0.04
		(2.4–9.3)	(2100–3800)	(87–300)	(780–3000)	(0.86–2.4)	(7.7–140)	(8.9–2900)	(<0.01–0.07)
fishes and prawn	42	3.5	28	2.9	15	0.07	1.3	1.8	0.06
		(0.38–110)	(0.48–380)	(0.15–23)	(0.86–140)	(<0.01–0.55)	(<0.1–6.1)	(<0.1–8.5)	(<0.01–0.50)
meat and animal fat	25	3.6	480	3.2	100	0.61	2.4	8.3	0.09
		(0.43–33)	(3.3–5500)	(0.14–110)	(0.13–820)	(0.02–4.8)	(<0.1–23)	(<0.1–37)	(<0.01–0.91)

[a] ΣHCH = α + β + γ + δ isomers. [b] ΣDDT = o,p'-DDT + p,p'-DDD + p,p'-DDE + p,p'-DDT. [c] Figures in parentheses indicate the range.

Table II. Concentrations of Organochlorine Pesticides and Polychlorinated Biphenyls (ng/g of Fat Wt) in Foodstuffs from Different Locations in India

food item	n	PCBs	ΣHCH[a]	γ-HCH	ΣDDT[b]	HCB	aldrin	dieldrin	heptachlor
cereals	3	100	9700	1500	1400	6.2	440	250	27
		(75–130)[c]	(8000–12000)	(940–2100)	(350–1900)	(3.4–8.2)	(35–820)	(57–480)	(8.2–39)
pulses	4	55	10000	710	1300	3.8	83	120	7.5
		(11–110)	(610–30000)	(73–2400)	(130–2700)	(2.3–7.0)	(30–110)	(61–210)	(0.67–27)
spices	5	91	5400	940	1500	5.6	53	590	15
		(4.7–360)	(2000–10000)	(250–1800)	(390–5500)	(<0.53–14)	(<3.3–210)	(<3.3–1900)	(1.7–41)
oils	5	11	220	39	21	1.5	19	24	0.45
		(2.6–15)	(6.9–480)	(1.2–100)	(1.8–57)	(0.09–2.8)	(<0.1–47)	(<0.1–47)	(0.08–1.6)
milk	10	49	16000	160	110	1.5	1.5	8.1	<0.06
		(1.7–210)	(1900–61000)	(16–600)	(11–650)	(0.15–3.8)	(<0.4–2.0)	(0.6–40)	(<0.06)
butter	4	7.2	3200	190	1600	2.0	51	870	0.05
		(2.8–11)	(2500–4500)	(100–350)	(920–3500)	(1.0–2.8)	(9–170)	(11–3400)	(<0.01–0.08)
fishes and prawn	42	330	2000	360	1100	9.1	110	130	6.6
		(25–2500)	(50–18000)	(4.2–2200)	(79–5600)	(0.17–28)	(<1.2–640)	(<7.1–410)	(0.33–31)
meat and animal fat	25	210	2200	140	1000	4.8	130	240	4.0
		(15–2600)	(320–7800)	(5–1100)	(28–5500)	(1.3–16)	(<0.24–1400)	(<0.24–1300)	(0.20–40)

[a] ΣHCH = α + β + γ + δ isomers. [b] ΣDDT = o,p'-DDT + p,p'-DDD + p,p'-DDE + p,p'-DDT. [c] Figures in parentheses indicate the range.

washed water. Subsequently, the concentrated hexane layer was cleaned and fractionated by passing through 1.5 g of silica gel (Wako S-1 gel) packed in a column (12 mm i.d.). The first fraction eluted with 160 mL of hexane contained PCBs, HCB, aldrin, heptachlor, and p,p'-DDE. The second fraction eluted with 20% dichloromethane in hexane (100 mL v/v) comprised HCH isomers, p,p'-DDD, p,p'-DDT, o,p'-DDT, and dieldrin. Each fraction was concentrated, and after 1 mL was retained for the quantification of aldrin and dieldrin, the extracts were subjected to further cleanup with 5% fuming sulfuric acid in concentrated H_2SO_4.

Samples were then injected into a Shimadzu Model GC-9A gas chromatograph equipped with ^{63}Ni electron capture detector and moving needle type injection system (splitless and solvent cut mode). The fused silica capillary column (30 m × 0.25 mm i.d.) coated with DB-1 (100% dimethylpolysiloxane) having a film thickness of 0.25 μm (J&W Scientific) was used for the determination of PCBs. The capillary column consisting of chemically bound DB-1701 (14% cyanopropylphenyl and 86% dimethylpolysiloxane) having the same dimensions and film thickness was employed for the analysis of organochlorine pesticides. The column oven temperature was programmed from 160 to 240 °C at a rate of 2 °C/min. Both injector and detector temperatures were kept at 300 °C. Nitrogen was used both as a carrier and as makeup gas. An equivalent mixture of Kanechlor preparations (KC-300, KC-400, KC-500, and KC-600) with known PCB composition and content was used as a standard. Concentrations of individually resolved peaks of PCB isomers and congeners were summed to obtain the total PCB concentration. Organochlorine pesticides were quantified from individually resolved peak heights with corresponding peak heights of standards. Recoveries of organochlorines by this method with fortified samples were more than 90%. Reported concentrations have not been corrected for the recovery percentage. The detection limit was 0.01 ng/g for all of the organochlorines but 0.1 ng/g for aldrin and dieldrin. For every set of five samples, a procedural blank consisting of all reagents and glassware used during analysis was run to check for interferences and cross contamination.

The dietary intakes of organochlorines were obtained by multiplying the residue level in the food by the amount of that food consumed. Total intake of the residues is then obtained by summing the intakes from all commodities containing the residue concerned. The recommended guidelines of UNEP/FAO/WHO for predicting dietary intake of pesticide residues were followed in the estimation of the organochlorine residue intake (UNEP/FAO/WHO, 1989).

RESULTS AND DISCUSSION

Residue Pattern. The concentrations of organochlorine pesticides and polychlorinated biphenyls in foodstuffs collected from different regions in India are shown in Table I. On the whole, contamination of Indian foods by organochlorine chemicals followed the order HCH > DDT > aldrin and dieldrin > PCBs > HCB > heptachlor. HCH contamination in different groups of foodstuffs can be rated as follows: dairy products > meat > pulses > oils > spices > cereals > fishes. A similar pattern was noticed for DDT and aldrin and dieldrin with a minor variation. In general, oils and dairy products contained higher levels of HCB than other foods while in spices and oils heptachlor dominated. Contamination by PCBs in oils is greater, followed by meat and fishes.

Concentrations of PCBs in foods from different locations invariably revealed its presence at lower levels and uniform contamination. Maximum contamination in oils could be due to the lipophilic nature of organochlorines. Since foodstuffs analyzed herein had a wide range of fat content, the data on lipid weight basis are essential to a comparative evaluation among foods for the potential organochlorine-containing items. When the concentrations were expressed on fat weight basis, fishes ranked first in PCB concentrations followed by meat and animal fat samples (Table II). However, such a pattern was unclear for organochlorine insecticides. These observations suggest that the major source of PCB contamination exists in areas around the aquatic environment and particularly in estuarine and coastal regions of India. This feature is common even in developed nations such as Japan, where Tanabe et al. (1989) estimated the environmental loads of HCH, DDT, and PCBs in the Seto Inland Sea area and found higher loads of the former two insecticides in agricultural soils in contrast to PCBs, which were abundant in coastal regions. Furthermore, our results also suggest the presence of low levels of PCBs in bovine milk, implying that the cattle feeds in India are not significantly contaminated with PCBs. In any case, these observations need attention in view of the rapid industrial growth in developing countries such as India. Cummins (1988) stated that the developing countries hold 15% of the total PCBs stock in the world. Similarly, our earlier studies also documented the contamination with PCBs of foodstuffs from Thailand (Tanabe et al., 1991). However, we could not trace the exact source of PCBs in developing nations. Transformers and similar electrical appliances are the suspected sources. Possible measures should be taken to prevent the further release of PCBs into water bodies from older electrical equipment and any other sources.

Among various organochlorines analyzed in the present study, HCH is the predominantly noticed compound in all of the foods. In other words, food pollution by HCH isomers is found prevalent in all regions in India. This can be explained by the shift in favor of HCH over the other organochlorines for agricultural purposes due to its low cost. Indian breast milk samples examined in our laboratory also revealed that the concentrations of HCH exceeded those of DDTs (Tanabe et al., 1990). In fact, earlier workers reported that the DDT concentrations were 18 times greater than those of HCH in human adipose tissue in 1964 (Dale et al., 1965). Subsequently, Siddiqui et al. (1981) documented that the levels of DDT were only 1.5 times higher than those of HCH in human adipose tissues in 1979. In the early 1980s, higher levels of HCH than DDT in maternal blood and placenta of live-born children were reported (Saxena et al., 1983). It can be concluded from these observations that the consumption of both HCH and DDT continues to grow but that HCH has supplanted DDT during recent years. In India, current consumptions of technical HCH and DDT are approximately 36 million and 14 million kg/year, respectively (Singh et al., 1988). These results can be evidenced further from the detection of higher levels of HCH than DDT in air, water, and soils (Ramesh et al., 1989, 1990, 1991) collected from paddy areas of southern India.

The isomer composition of HCH in different groups of foodstuffs from India is shown in Figure 1. It can be seen that α-HCH predominated in all of the farm products, which again indicates the recent usage of HCH for agricultural purposes. In contrast, dairy products, meat, and fishes showed higher composition of β-HCH, which is the isomer most stable toward enzymatic degradation in the animal body.

Next to HCH, DDT levels in Indian foods are apparently high. The concentrations of DDT were comparable to those of HCH in New Delhi and Bombay. On the other hand, HCH levels surpassed those of DDTs in suburban and agricultural areas (Chidambaram and Parangipettai). This characteristic trend is in line with our expectations; HCH is preferentially recommended for the control of agricultural pests, while DDT is used mainly for mosquito control in urban areas (Singh et al., 1988). Of the total annual DDT and HCH consumption, 85 and 16%, respectively, are used for mosquito control in India (Mehrotra, 1985).

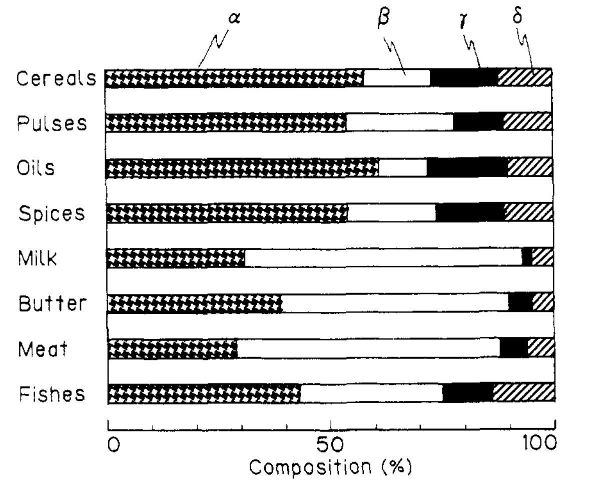

Figure 1. HCH isomer compositions in food samples collected from India.

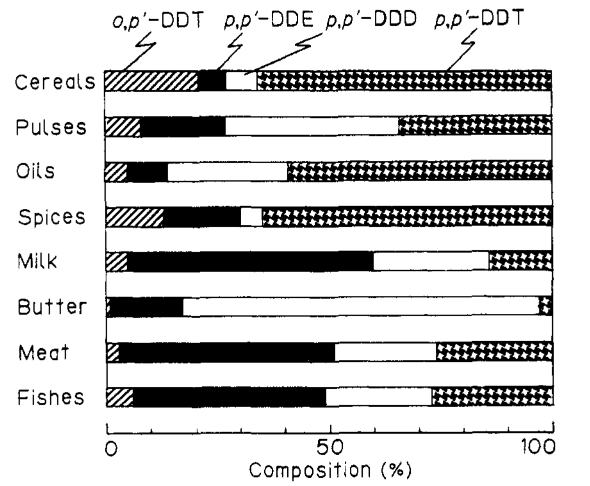

Figure 2. DDT compound compositions in food samples collected from India.

The overall DDT compound composition in different classes of foodstuffs from India is shown in Figure 2. *p,p'*-DDT constituted the maximum proportion in cereals, pulses, oils, and spices. Conversely, *p,p'*-DDE predominated in milk, meat, and fishes, while *p,p'*-DDD predominated in butter. Metabolic transformation of DDT under oxidative conditions led to DDE in animal bodies and thereby accounted for higher levels. It is rather interesting to note the marked difference in the DDT metabolic composition between milk and butter. This change in the proportion of DDT metabolites might be due to the microbial conversion of DDT to DDD during the fermentation (anaerobic conversion) of milk leading to curd. At this juncture, it is worth mentioning that the traditional method of preparation of butter in India consists of an initial fermentation step followed by cream separation. Bacterial fermentation of carbohydrates in milk to form acids resulting in the curdling of milk might have simultaneously favored the conversion of DDT to DDD. Archer (1976) indicated that the microorganisms present in dairy products could reduce DDT and DDE to DDD under anaerobic conditions. Earlier papers also encountered the existence of *p,p'*-DDD as a dominant metabolite, constituting more than 50% of ∑DDT in Indian butter (Dhaliwal and Kalra, 1978; Kalra et al., 1986). Another possibility for the variation in DDT metabolite composition between milk and butter might be due to the difference in DDT component composition in the cattle feed to which the animals are exposed (Fries et al., 1969, 1971). In other words, the animals that produced the milk used for butter production might have been fed with feed mixtures containing a higher proportion of p,p'-DDD and so on.

Among various food commodities analyzed in the present study, milk and butter recorded the maximum levels of HCH and DDT. The pesticidal contamination of milk and milk products, because of their special significance in the human diet, has been viewed with great concern in India from the mid 1960s (Singh and Singh, 1990; Dhaliwal, 1990). The routes by which organochlorine compounds reach dairy products are mainly through the intake of contaminated feed by animals and to a lesser extent by skin adsorption following dermal treatment of animals with spray or dust. Moreover, application of DDT in cattlesheds is commonly practiced in India, because livestock wastes are an important breeding site for mosquitoes, and consequently animals are frequently exposed to DDT. The excretion of HCH residues in the milk of Indian buffalo, after oral and dermal exposure to technical HCH, was studied by Kapoor and Kalra (1988). Since pesticides adhere to fatty tissues in the body, milk, which is rich in fat, has been an ideal storehouse for toxins. When milk is processed into butter, residue levels are magnified. Earlier studies reported the presence of considerable levels of organochlorine pesticides in commercial cattle feed (Dikshith et al., 1989b), wheat straw (Singh et al., 1988), and rice straw (Rajukkannu et al., 1985) in India.

Next to dairy products, animal fat and meat including chicken, mutton, and pork contained higher levels of organochlorine pesticides. The extensive contamination of animal meat with organochlorine insecticides could be due to the higher accumulation ratios of the chemicals in dairy cattle (Noble, 1990). Kaphalia and Seth (1981) revealed that the highest contamination by HCH and DDT was observed in chicken fat followed by mutton and beef collected from Lucknow, India.

With regard to DDT contamination, meat products were followed by spices, fishes, oils, pulses, and then cereals. DDT concentrations in fruits and vegetables from India are lower (Kaphalia et al., 1990). This is because the Agricultural Ministry of the Indian government advises farmers not to use HCH and DDT on vegetables and oilseeds. However, significantly elevated levels of these insecticides were detected in oils. Probably, these insecticides are still being applied to oilseed crops despite the Ministry's recommendation.

Pollution by HCB is minimal in all of the locations, which could probably be due to its smaller usage dose. Since HCB is mainly recommended for seed treatment, large-scale application such as spraying or dusting is not followed. Even the existing amounts of residues in foodstuffs might be contamination from the pesticide formulations, which contain small amounts of HCB as an impurity (Tanabe et al., 1991). The results of this study are in agreement with those of Nair and Pillai (1989), who found that HCB residues in the Indian environment are low. Besides, HCB is a byproduct of various chlorination processes and combustion of industrial products, which might also be a means of entry into the environment.

Significant levels of aldrin and dieldrin were recorded in Indian foods. As observed for HCH and DDT, dairy products contained elevated concentrations of aldrin and dieldrin followed by oils, spices, and meat. The presence of considerable levels of aldrin in commercial cattle feed (Dikshith et al., 1989a) and vegetable oils (Dikshith et al., 1989b) might be the explanation for the higher levels in dairy products. One of the samples of butter from New Delhi showed extremely high levels of aldrin and dieldrin, which indicates that these insecticides are used sporadically in certain regions of India. Even though aldrin is almost completely converted to dieldrin when ingested by cattle (Rumsey and Bond, 1974), the presence of a considerable level of aldrin in one of the butter samples indicates the possibility of postproduction contamination during handling, transport, storage, and marketing. Aldrin and dieldrin are still being used for the control of crop pests in India. As evidence, Bakre et al. (1990) indicated that aldrin 30% emulsifiable concentrate (895 L/annum) was utilized in the catchment area of the Mahala water reservoir, Jaipur, India. Besides being used as an insecticide, aldrin is also reported to be used as a termiticide, mostly in agricultural fields. Aldrin is not recommended in India for direct spray on vegetables. Nevertheless, Lal et al. (1989) detected its presence in most of the vegetables collected from Delhi markets. Due to the extremely higher toxic potential of these compounds, considerable attention must be paid to avoid further contamination in foodstuffs.

The levels of heptachlor were lower in all of the foodstuffs from different regions. Information concerning heptachlor contamination in various ecosystems in India is meager. An earlier investigation by Noronha et al. (1980) reported that heptachlor residues in wheat collected from Bombay markets exceeded the tolerance limit prescribed by FAO/WHO. They also showed the presence of heptachlor in other cereal grains from Bombay. Recently, Bakre et al. (1990) documented the presence of heptachlor in Mahala reservoir water samples in India. Similarly, Sivaswamy (1990) showed that the levels of organochlorine pesticides including heptachlor in sediments from the Madras coast exceeded the safety limit. It is known that heptachlor is metabolized into heptachlor epoxide in humans and animals (Tashiro and Matsumura, 1978). The smaller concentrations of heptachlor in animal meat and fish might be due to its metabolic conversion into heptachlor epoxide. However, we did not determine heptachlor epoxide in this study because of its instability after treatment with 5% fuming H_2SO_4.

We also attempted to evaluate the extent of chlordane contamination in Indian foods, but the levels were too low and <1 ng/g in most of the cases (data not shown). This suggests that the chlordanes are not being used extensively in India.

Tolerance Limits. The results obtained in this study were compared with the recommended tolerance limits of the FDA (for PCBs), the FAO/WHO, and the Ministry of Health (MOH) of the government of India for organochlorine residues in various foodstuffs (Figure 3). To our knowledge there exists no information on the acceptance limits for PCBs in foodstuffs set forth by the government of India. Hence, the tolerance limit suggested by the U.S. FDA was compared; the PCB levels in Indian foods were well below the maximum acceptable limit. Similarly, the Ministry of Health has set forth maximum residue limits (MRL) in foodstuffs only for γ-HCH and not for other isomers. It can be seen in Figure 3 that the dairy products exceeded the prescribed limit even in the case of γ-HCH. γ-HCH levels observed for food grains were close to the MRL of the FAO/WHO and MOH. Likewise, the ΣDDT tolerance limit was surpassed in dairy products, and the country's mean level is close to the MRL of MOH. Meat products showed concentrations below the limits of the FAO/WHO and MOH. As observed for γ-HCH and DDT, dairy products exceeded the tolerance limit of the Indian

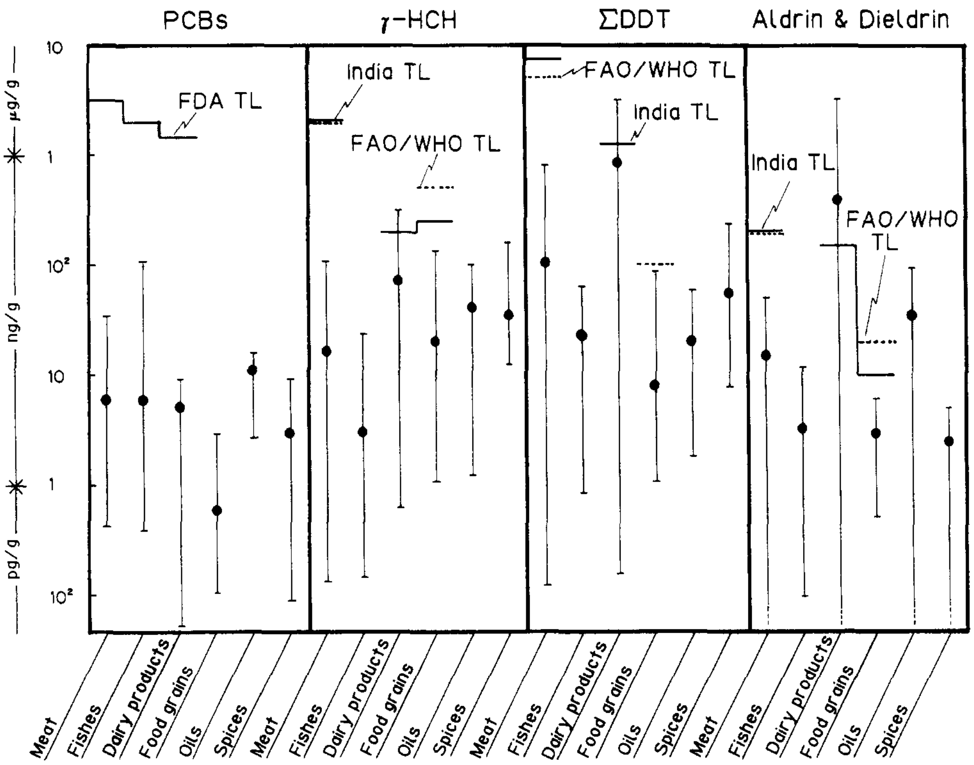

Figure 3. Comparison of organochlorine concentrations (ng/g of wet wt) in Indian foodstuffs with FDA, FAO/WHO, and the Indian government's tolerance limits (TL). The solid jagged lines indicate, in panels other than the first one, Indian TL, while the dashed ones indicate FAO/WHO TL.

government for aldrin and dieldrin in India. Even the national average concentration was far above the attributed limit. Aldrin and dieldrin levels in food grains and meat were close to the maximum acceptible residue limits of the FAO/WHO and MOH. Considering these results, it is clear that the dairy products should be monitored periodically to prevent further contamination in humans. A number of surveys made so far explain that the residue limits of HCH and DDT in Indian dairy products outstripped the FAO/WHO practical residue limit (Dhaliwal, 1990). A 2-year multicentric study sponsored by the FAO through MOH on food contamination monitoring in India revealed that DDT residues exceeded the Codex MRLs in about 10% of the cereals, 60% of the bovine milk, and 66% of the butter samples (Kalra and Chawla, 1985). Our results further suggest an urgent need for control of the usage of these pesticides to minimize their levels in milk and butter.

Dietary Intake. On the basis of information obtained from the National Nutrition Monitoring Bureau, India (personal communication from the Director of the National Institute of Nutrition, Hyderabad, India), regarding the average daily consumption (g/person) of foodstuffs by Indians in 1988–1989 (cereals and millets, 520; pulses, 35; spices and condiments, 15; fat and oils, 15; vegetables, roots, and tubers, 110; fruits, 25; flesh foods, 15; milk and milk products, 90; nuts and oilseeds, 10; sugar and jaggery, 30) and using the values in Table I, we attempted to estimate the average daily intake of these chemicals by Indians; the results are presented in Table III. The residual concentrations recorded by Kaphalia et al. (1990) for ΣHCH and ΣDDT in fruits (2.3 and 2.8 ng/g, respectively) and vegetables (2.8 and 4.5 ng/g, respectively) were also taken into account for this calculation. The numbers of samples of vegetables and fruits analyzed in their study were 59 and 20, respectively. However, we did not consider the daily consumption of water, nuts, sugar, and jaggery for this calculation. This would not have affected the results significantly, because of the smaller

Table III. Comparison of Average Dietary Intake of Organochlorines by Indians with FAO/WHO Intake Limit

organochlorine compound	av daily intake by Indians,[a] µg/person	FAO/WHO daily intake limit, µg/60 kg of body wt
ΣHCH	155	na[b]
γ-HCH	12	600
ΣDDT	48	1200
aldrin and dieldrin	19	6.0
PCBs	0.86	na
HCB	0.13	na
heptachlor	0.07	na

[a] Calculated in the present study. [b] na, data not available.

amount of consumption of these commodities coupled with the expected lower residue levels of chemicals in these commodities. Interestingly, the dietary intake of HCH, DDT, and aldrin and dieldrin is quite high in India (Table III). The contributions of dairy products to the dietary intake of HCH, DDT, and aldrin and dieldrin were 70, 87, and 87%, respectively. The data for HCH and DDT are presented in Figure 4. This is in accordance with our expectations because of the reasonably higher quantity of consumption of dairy products in the total diet and their extremely greater levels of contamination. However, it should be noted that the average daily intakes of HCH and DDT were below the recommended limits of the FAO/WHO. It is worth mentioning here that the dietary intake of aldrin and dieldrin exceeded the acceptable daily intake (ADI) for a person with an assumed body weight of 60 kg (Table III). There have been only a few investigations from India reporting the actual intake of DDT and HCH residues. Gupta et al. (1982), on the basis of "total meal study", highlighted that the average daily intake of DDT and HCH residues by the general population was comparatively high as compared to the average daily intake in many other countries. The results of the total diet studies of Singh (1982) showed that the mean daily dietary intakes of HCH and DDT were 124 and 238 µg/person, respectively, from the vegetarian diet and 133 and 224

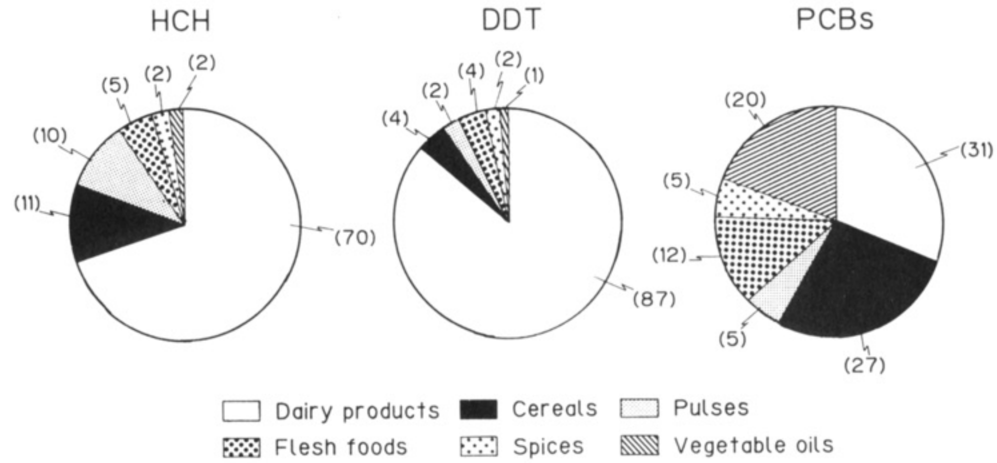

Figure 4. Contribution from different classes of foodstuffs toward the dietary intakes of ΣHCH, ΣDDT, and PCBs by Indians. (Figures in parentheses indicate the percentage.)

μg/person, respectively, from the nonvegetarian diet. They also concluded that the average daily dietary intake of DDT equaled the ADI of 0.005 mg/kg, i.e., 300 μg/person for a man weighing 60 kg. On the other hand, the results of the present study showed higher dietary intake of HCH than of DDT. This reiterates the increased consumption of HCH over DDT in recent years when compared to the early 1980s. A survey conducted in the mid 1980s (Kaphalia et al., 1985) elucidated the higher dietary intake of total HCH (1300 μg/day) than of total DDT (200 μg/day) by an adult Indian. Nevertheless, the mean dietary intake of both DDT and HCH was found to be more than that reported from the United States, the United Kingdom, Canada, and Australia (Kalra and Chawla, 1985). The dietary intakes of PCBs, HCB, and heptachlor in India are relatively low (Table III), which might be due to the minimal contamination of the environment by these compounds in developing countries. A recent investigation by Matsumoto et al. (1988) stated that the total amount of PCBs contained in a day's diet for a Japanese person averaged 2.8 μg/60 kg of body weight, of which about 80% of the contribution was from fishes and other seafoods. In West Germany, Brunn et al. (1989) found that dairy products such as butter, margarine, and sausage were the predominant contributors to the average daily intake of PCBs, where the daily intake rate of 3.4 μg/person was reported. In India, the daily intake of PCBs is 0.86 μg/person, which is apparently lower than those of the developed nations. The source of PCBs intake by Indians was rather unique, where significant contribution was observed from cereals and vegetable oils as well as from dairy products (Figure 4).

CONCLUSIONS

It can be inferred from the present study that the contamination of foodstuffs by HCH, DDT, aldrin, and dieldrin is prevalent throughout India. Dairy products such as milk, butter, and animal meat are the prime sources of human exposure to these chemicals. Residue levels in dairy products exceeded the guidelines set forth by the FAO/WHO and the Ministry of Health of the government of India. Similarly, the dietary intake of aldrin and dieldrin exceeded the ADI recommended by the FAO/WHO. The average daily intake of HCH and DDT was several times higher in India than in most developed nations. Food contamination and dietary intakes of PCBs, HCB, and heptachlor are lower in India. However, in view of the extremely higher toxicity of PCBs and rapid industrial development in India, considerable attention must be paid to avoid further contamination by PCBs. It is also stated that the numbers of samples analyzed for individual foods from each location are small; however, the information obtained through this monitoring effort show that exposures seem to be below the ADIs. Because of the small sample size, further monitoring would have to be done to confirm this impression.

The impact of food contamination with chemical substances on human health is influenced by several factors, including the prevailing socioeconomic as well as the geographical conditions of a country and its inhabitants. As evidence, our earlier study reported that the breast milk of Indian vegetarians contained greater levels of HCH and DDT than those of nonvegetarians and fish eaters (Tanabe et al., 1990). This trend could primarily be due to the higher consumption of dairy products (milk, butter, ghee, etc.) by vegetarians than by nonvegetarians, and greater contamination of these foodstuffs by persistent organochlorines has been found in this study.

It is also worthwhile to indicate that the food contamination in agricultural countries such as India not only is of local concern but also has global significance. In this context, the developed nations which are importing a variety of foodstuffs from the developing countries and concomitantly exporting chemical products and providing technological know-how to the same should also elaborate the countermeasure for preventing global contamination of humans from the tropical point source areas.

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Registry No. α-HCH, 319-84-6; β-HCH, 319-85-7; γ-HCH, 58-89-9; δ-HCH, 319-86-8; *o,p'*-DDT, 789-02-6; *p,p'*-DDD, 72-54-8; *p,p'*-DDE, 72-55-9; DDT, 50-29-3; HCB, 118-74-1; aldrin, 309-00-2; dieldrin, 60-57-1; heptachlor, 76-44-8.