Organochlorine Insecticide Residues in Dairy Milk Samples Collected in Lucknow, India

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Food-chain contamination with persistent pesticide residues is a global concern due to associated human health hazards (Loganathan and Kannan, 1994). Pesticide residues have been reported in food grains, vegetables, fruits and animal products in market samples from time to time (Kaphalia et al, 1990). However, because of their special significance in the human diet, particularly for children, contamination of milk and milk products has been viewed with greater concern (Dhaliwal, 1990). These products constitute about 10% of the total diet among Indians. A survey sponsored by the FAO through the Ministry of Health. Government of India, during 1980s showed DDT and HCH in milk at 0.19-216 and 0.12-40 ug/g fat respectively with more than 60% samples contaminated above the MRL (Kalra and Chawla 1985). More recently, a multicentric study conducted by the Indian Council of Medical Research on samples of dairy milk collected from rural and urban areas of 12 states showed 80% of samples contaminated with DDT and HCH; median values were more than those reported from most other countries (Kalra et al., 1999). Uttar Pradesh, the most populous state in India, comprised of 83 districts with Lucknow, the capital of U.P. alone is habituated by more than 2.5m residents. Although, data for pesticide contamination of milk collected in U.P. is available but in view of its large population a district wide study may better reflect the state with regard to contamination of dairy milk by persistent pesticides. In the present study, milk samples from booths of two different dairies in Lucknow were collected and analysed for the presence of persistent organochlorine insecticide residues to ascertain the extent of contamination in Lucknow and compared with pesticidal contamination of dairy milk in other states of India.

MATERIALS AND METHODS

Milk samples (500 ml in duplicates) from each of the two main dairies (coded as A and B) marketed through milk booths in Lucknow were collected daily for twenty days and immediately transported to laboratory under ice-cold condition and stored at 4°C until analysed, generally within 48 hrs. At the time of processing samples were thoroughly shaken for breaking the fat globules. Extraction was carried out following the method as described by Kapoor et al., (1981) briefly involved extraction with acetone + hexane (1:1, v/v) followed by

treatment with concentrated sulphuric acid for clean-up. The cleaned concentrated extract was used for the estimation of organochlorine insecticides by gas-liquid chromatograph, Nucon 5765, equipped with an electron capture detector (63Ni) and Pyrex glass spiral column 6 m long; 4 mm i.d. packed with a mixture of 1.5% OV-17+1.95% OV-210 on chromosorb-W (80/100). The operating conditions of GLC were: Column temp. 180°C; Injector temp. 250°C; Detector temp. 250°C; Carrier gas was IOLAR grade-1 nitrogen (99.9%) further purified by passing through silica gel and molecular sieve to remove moisture and oxygen respectively; Flow rate 60 ml/min; chart speed 0.5 Cm/min. Recovery of residues from spiked samples was 84-95%. Identification was confirmed by TLC. Results were not corrected for recovery.

RESULTS AND DISCUSSION:

It is clear from the Table 1 that all the samples of milk from Dairy A and Dairy B were contaminated with persistent organochlorine insecticides DDT and HCH. Significantly none of the samples from two dairies has total DDT above the

Table 1. Organochlorine insecticides detected in milk samples of dairy A and

dairy B (ppm) in Lucknow

Pesticides	cides Dairy A				Dairy B			
	%	Range	Mean	Above	%	Range	Mean	Above
	Positive			MRL(%)	Positive			MRL(%)
α-HCH	100	0.005-	0.067	70%	100	0.028-	0.059	60%
		0.127				0.089		
β-НСН	100	0.003-	0.078	80%	100	0.020-	0.068	100%
•		0.139				0.115		
ү-НСН	100	0.007-	0.025	70%	100	0.009-	0.022	90%
•		0.061				0.067		
δ-НСН	100	0.003-	0.007	None	100	0.004-	0.006	None
_		0.015				0.009		
Total	100	0.019-	0.177	70%	100	0.088-	0.155	80%
HCH		0.295				0.277		
p,p'-	90	0.001-	0.005	_	100	0.002-	0.006	_
DDT		0.011				0.010		
o,p'-	100	0.001-	0.003	_	100	0.001-	0.002	-
DDT		0.012				0.005		
p,p'-	100	0.005-	0.007	_	100	0.005-	0.007	_
DDE		0.010			100	0.010		
p,p'-	90	0.001-	0.005	-	100	0.002-	0.007	-
TDE		0.011				0.013		
Total	100	0.010-	0.020	None	100	0.014-	0.023	None
DDT		0.030				0.033		

Data represents the mean of twenty samples each from the two dairies in Lucknow; Total HCH, the sum of α -, β -, γ -, δ - isomers; Total DDT, sum of p,p'-DDT+p,p'-DDE+p,p'-TDE

maximum residue limit (MRL) of 0.05 mg/kg in milk as set by codex alimentarius (FAO, 1986). However, total HCH level was higher than 0.10

mg/kg (MRL) established by government of India under prevention of food adulteration act (Anonymous, 1992), both in samples of Dairy A and Dairy B. The levels of α , β , γ isomers separately were also above their prescribed MRL of 0.05, 0.02 and 0.01respectivley (Table 1). A comparison of the present data with those of our similar study conducted in Lucknow (U.P.) in 1981 (Saxena and Siddiqui 1982) suggest a 50% decline in DDT residue and more than three times increase in HCH level (Table 2) reflecting the usage pattern of DDT and HCH in India. DDT contamination of milk samples collected from Lucknow (present study) was lower than reported from Punjab, Haryana, Madhya Pradesh,

Table 2. A comparison of pesticide residue level in dairy milk from different states in India.

Place/location	Year	DDT	HCH	Reference
Himachal Predesh	1997	0.091	0.037	Kalra et al., 1999
Haryaya	1997	0.022	0.051	Kalra et al., 1999
Madhya Predesh	1997	0.042	0.056	Kalra et al., 1999
West Bengal	1997	0.021	0.170	Kalra et al., 1999
Bihar	1997	0.041	0.179	Kalra et al., 1999
Tamil Nadu	1989	0.02	0.18	Kannan et al., 1992a
Kerala	1997	0.030	0.082	Kalra et al., 1999
Delhi	1992-93	0.017	0.09	Mukherjee & Gopal 1993
Andhra Pradesh	1997	0.207	0.563	Kalra et al., 1999
Punjab	1980-81	0.24	0.04	Singh, 1982
•	1997	0.111	0.067	Kalra et al., 1999
Uttar Predesh	1980-81	0.04	0.05	Saxena & Siddiqui 1982
	1997	0.030	0.234	Kalra et al., 1999
Maharashtra	1991-93	0.020	0.094	Dethe et al., 1995
	1997	0.080	0.049	Kalra et al., 1999
Gujarat	1990	0.063	5.19	Kashyap et al., 1994
•	1997	0.091	0.073	Kalra et al., 1999
Karnataka	1991	0.0229	0.1090	Awasthi & Ahiya 1995
	1997	0.047	0.179	Kalra et al., 1999

Values are in ppm; only mean given; DDT means sum of its isomers and metabolites; HCH means sum of α -, β -, γ -, δ - isomers.

Maharashtra, Gujarat, Bihar, Andhra Pradesh, Karnataka and Kerala but higher than that of Delhi, Tamil Nadu and West Bengal (Table 2) probably indicating the regional differences in the application of this pesticide. Likewise contamination of milk samples with total HCH was much less than that of Gujarat and Andhra Pradesh comparable to Bihar, West Bengal, Karnataka and Tamil Nadu and higher than that of Delhi, Kerala, Maharashtra and Madhya Pradesh (table 2). Evidently, there is a general decline in DDT and a build-up of HCH residues in dairy milk samples from different regions in India.

The organochlorine insecticides DDT and HCH which were banned or severely restricted for use in many countries, constituted the bulk of pesticides consumed

in India for about five decades (Parmar and Dureia, 1990). While agricultural use of DDT was banned, the manufacture and use of technical grade HCH has been banned in India since April 1997 (Kalra et al. 1999). Still dairy milk and its products were shown to have a high incidence of contamination with organochlorine insecticide residues in India (Kannan et al., 1992). As the presence of DDT and HCH in cattle feed in India has already been reported (Battu et al., 1989) it seems to be the main source of organochlorine contamination of dairy milk samples in the present study. Being lipophilic in nature, low amounts of these residues in feed are bioconcentrated resulting in unacceptable levels in dairy milk. In addition, spraying of DDT and HCH to control mosquitoes in cattle sheds of the two dairies may be expected to contribute significantly towards contamination of dairy milk. Breast milk of Indian vegetarians was reported to contain higher amounts of DDT and HCH residues than that of non-vegetarians and fish eaters whereas reverse trend was noticed in several countries in North America and Europe (Kannan et al. 1997) primarily due to greater consumption of dairy products by vegetarians. Data of this study together with those of others (Mukheriee and Gopal, 1993) reporting above MRL levels of DDT, aldrin, dieldrin, and v-HCH in dairy products suggest periodic monitoring of dairy products to prevent excessive exposure of humans to organochlorines. In the context of two dairies in Lucknow that caters to the milk and milk products requirement of a sizable chunk of 2.5 million Lucknow residents, steps to mitigate the cattle feed contamination with organochlorines is the need of the hour

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REFERENCES

Anonynous (1992) Gazette Notification No 262. Government of India.

Awasthi MD, Ahuja AK (1995) Organochlorine Insecticide Residues in Bovine Milk and commercial baby milk powder. J Food Sci Technol 32:420-422.

Battu RS, Singh PP, Joia BS, Kalra RL (1989) Contamination of stored food and feed commodities from indoor use of DDT and HCH in malaria control programmes. Sci Tot Environ 78: 173-178.

Dethe MD, Rane SD, Kale VD (1995) DDT and HCH residues in milk, milk powder, cattle feed and fodder. Ind J Environ and Toxicol 5: 41-43.

Dhaliwal GS (1990) Pesticide contamination of milk and milk products. In Nriagu JO, Simmons MS (eds) Food contamination from Environmental sources. Wiley, New York, pp 357-385.

FAO (1986) Codex Maximum Limits for Pesticide Residues 2nd Edition (Rome: Food & Agriculture Organisation).

Kalra RL, Chawla RP (1985) Pesticidal contamination in foods in the year 2000 AD. Proc Indian Natl Sci Acad B52: 188-204.

- Kalra RL, Kaur H, Sharma S, Kapoor SK et al (1999) DDT and HCH residues in dairy milk samples collected from different geographical regions of India: a multiecentric study. Food Add Contam 16: 411-417.
- Kannan K, Tanabe S, Giesy JP, Tatsukawa R (1997) Organochlorine Pesticides and Polychlorinated Biphenyls in Foodstuffs from Asian and Oceanic Countries. Rev Environ Contam Toxicol 152: 1-55.
- Kannan K, Tanabe S, Ramesh A, Subramaniam A, Tatsukawa R (1992). Persistent Organochlorine residues in food stuffs from India and their implications on human dietary exposure. J Agric Food Chem 40: 518-524.
- Kaphalia BS, Takroo R, Mehrotra S, Nigam U, Seth TD (1990) Organochlorine pesticide residues in different Indian cereals, pulses, spices, vegetables, fruits, milk, butter, desighee and edible oils. J Assoc Off Anal Chem 73: 509-512.
- Kapoor SK, Chawla RP, Kalra RL (1981) A simplified method for determination of DDT and hexachlorocyclohexane residues in milk. J Association Off Anal Chemists 64:14-15.
- Kashyap R, Iyer LR, Singh MM (1994) Evaluation of daily dietary intake of dichlorodiphenyltrichloroethane (DDT) and benzene hexachloride (BHC) in India. Arch Environ Health 49:63-66.
- Loganathan BG, Kannan K (1994) Global Organochlorine contamination trends: an overview. Ambio 27: 187-191.
- Mukherjee I, Gopal M (1993) Organochlorine pesticide residues in dairy milk in and around Delhi. JAOAC Int 76: 283-286.
- Parmar BS, Dureja P (1990) Minimizing Environmental hazards of agrochemicals-Indian Approach and Status. Soc Pest Sci New Delhi. India.
- Saxena MC, Siddiqui MKJ (1982) Pesticide pollution in India: Organochlorine pesticides in milk of women, buffalo and goat. J Dairy Sci 65: 430-434.
- Singh PP (1982) Insecticide residues in foood in Punjab a typical dietary survey. Ph.D. thesis, Punjab Agricultural University, Ludhiana, India.