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Pesticide Residues in Drinking Water and Cattle Milk. High Time to find an alternative to Chlorinated Hydrocarbons

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Abstract:

"All substances are poisons; there none which is not a poison.
The right dose differentiates a poison and a remedy"

PARACELUS (1493-1541)

Organochlorine pesticides (OCPs) are quite popular in India mainly because of the substantial gains from their use in agriculture as well as in health programmes. Even after having clear cut evidence suggesting that these chemicals have the ability to eliminate entire species from the earth, the annual consumption of pesticides in India is about 85,000 tonnes of which OCPs comprise the bulk. They travel to distant places away from their point of application through the long-distance transport mechanisms and get distributed widely all across the world. Therefore, today the pesticides and in particular OCPs are, perhaps the most ubiquitous of the potentially harmful chemicals encountered in the environment. The problems would further aggravate in the coming year as OCPs being lipophilic and non-biodegradable in nature due to their great chemical solubility, low aqueous solubility and high fat soluble character became concentrated and magnified as they move up in the food chain. Keeping the cost benefit ratio in mind, we have largely overlooked the darker side of these chemicals i.e. unwanted and unwarranted environment pollution caused by the indiscriminate use of pesticides, secondly their biocidal activity is not only restricted to the target organism but extends to non-target organisms as well. All the above resulted in the natural exposure of human beings and further animal experimentation led to the legislation of allowable concentrations of these xenobiotics. This legislation was made with the intention to prevent the exposure of the general human population to harmful levels of these xenobiotics and prevent contamination of Mother Nature. A monitoring study was therefore, conducted in Bikaner, Rajasthan, (India) in which drinking water and cattle milk were analyzed for OCPs. The above study is of special significance for the Indian population, since, Indians have been reported to possess the highest body burden of pesticides. The samples were analyzed for pesticide residues viz- Dieldrin, isomers of HCH, Heptachlor and its metabolites, Endosulfan and DDT and its metabolites by using gas liquid chromatography. Study revealed that samples of drinking water and cattle milk collected from the Bikaner were having the residues of OCPs in them. Presence of these compounds in the drinking water and cattle milk shows that these compounds are quite stable and have a very long residence time, therefore, the environmental pollution caused by them need to be monitored for the safety of the mother nature and human life.

Keywords: Gas chromatograph; Water; Cattle Milk; Bikaner City; Organochlorine pesticide, Residue

Introduction

There is a burly inclination of the present human race to resort to the use of chemicals especially pesticides, to control various unwanted forms of plant and animal life. Substantial gains from the use of these xenobiotics in agriculture as well as in health programmes have been acknowledged by all. Nonetheless, the harmful residues that remain on crops, especially the edible portion, have been a cause of great concern for the safety of the Mother Nature and human health. Besides, the contamination of feed and food commodities, there is a growing fear that pesticides particularly the persistent types get transported into air, water and soil system from point of initial application, thus endangering the normal functioning of non-target organisms which includes human beings as well. Among the pesticides, Organochlorine pesticides (OCPs) which are lipophilic compounds are well known for their long persistence in the environment and in living organism. Because of this many developed countries who can afford even costlier chemicals, have either restricted or banned these pesticides as a measure to check unwarranted environmental pollution. But in developing countries or third world countries such as India, because of the cost benefit ratio OCPs are still, the major pesticides used in agricultural and public health sector. OCPs are reported to be carcinogenic [1]-[8], mutagenic [9],[10] teratogenic[10], [11],immunosuppressive[12]-[14] create endocrine dysfunction such as hypothyroidism or high estrogenic activity [15]-[18], disturb reproductive

processes[19]-[21], growth depressants [22], [23]induces several psychogenic and neurogenic abnormalities in adult stages[24]-[26], and are associated with abortions, premature deliveries, still births and infants with low birth weights [27]-[38].OCPs have been in use in India nearly for a half century now. Even after having clear cut evidence suggesting that these chemicals have the ability to eliminate entire species from the planet, the annual consumption of pesticides in India is about 85,000 tonnes of which organochlorine comprise the bulk [39].

Taking above point into consideration, a continued surveillance on the levels of pesticide pollutants in human food is an important task to ensure the wellbeing of the human health and Mother Nature. It was, therefore, planned to conduct a monitoring study in Bikaner, Rajasthan, India in drinking water and bovine milk were analyzed for the OCPs residues.

Materials and Methods

Materials and Methods will be discussed under following heads:

1. Sampling.
2. OCPs estimation of water and Milk Samples.
3. Statistical Analysis
4. Questionnaire.

1. Sampling

Water and milk samples were collected after doing the survey of the Bikaner city and site selection. As per the Public Health Engineering Department (PHED), Bikaner, water supply of the city is divided into two zones. First zone is the old city (internal part of the city) and second zone is the new city (outer part of the city). In this part of the paper old city zone is taken into consideration and it has four following subdivisions (I-IV).

Old City

I.	-	Beechwal Sub Division
II	-	Stadium Sub Division
III	-	Jail Well Sub Division
IV	-	Nathusar Sub Division

New City zone is also divided into following four sub divisions (V-VIII)

V	-	Mukta Prasad Sub Division
VI	-	J.N.V. Colony Sub Division
VII	-	Industrial Area Sub Div.
VIII	-	Gangasahar Sub Division

As per the PHED information, water supply in the Bikaner city includes mixture of ground water and Indira Gandhi Nehar Pariyojna (IGNP-Canal water).In the old city water constitution carries more percentage of canal water than ground water and in the outer city it contains more percentage of ground water than canal water.

Water samples were collected from the old city and were kept in glass bottles prewashed with (8M) nitric acid and stored at 4 degree C and analyzed within 48 hours. 25 buffalo milk samples were also collected from selected sites of the old city. Milk samples were kept in glass bottles and stored at 4 degree and analyzed within 72 hours of the collection.

2. **OCPs estimation of water and Milk Samples**

Pesticides were extracted and separated from samples by liquid partition and column chromatography so that they could be analyzed by Gas Liquid Chromatography (GLC) and Thin Layer Chromatography (TLC) procedures. All reagents and chemicals used were of analytical grade and checked for any pesticide contamination. The water samples were extracted by a method prescribed by APHA (1980) [40] and milk samples were extracted by the method given by Takie *et al* (1983) [41] with slight modifications depending on the laboratory condition and recovery experiments. All the chemicals used for the extraction of OCPs were of analytical grade. Water samples (500 ml) were extracted twice, first with 25 ml hexane, 15 ml saturation solution of NaCL and then with 25 ml hexane. Clean up was done on the alumina column. Specimens of milk were extracted by the methodology given by Takie and his Coworkers in 1983[38] with little modifications according to the prevailing laboratory conditions. Milk (2.5ml) was extracted by adding 2 ml of acetone and 1 ml of acetonitrile and 8 ml of hexane and the contents were shaken for 3 min. by hand. The upper layer of hexane was recovered and milk was re-extracted by adding hexane, acetone and acetonitrile. Pooled extract of hexane was washed with acetonitrile and 2% aqueous sodium sulphate and hexane. The analysis of pesticide residues in human milk was done on a whole milk basis.The entire final extracts, after cleanup were evaporated to dryness in a rotatory flash evaporator. The dried extracts were dissolved and made up to 2 ml with hexane in a glass vials and stored in a refrigerator till analyzed.

Qualitative and quantitative estimation of OCPs were carried out on Schimadzu model 2010 coupled with personal computer and equipped with a capillary Column HP ultra (US 4293415) 0.52 X 2.5 X 0.32. The results indicate that nearly all the water and milk samples were contaminated with dichlorodiphenyl-trichloroethane (DDT) and its metabolites (DDE and

p,p'-dichlorodiphenyltrichloroethane [DDD]), isomers of hexachlorocyclohexane (HCH; alpha, beta, and gamma), heptachlor and its epoxide, and Aldrin. Purified nitrogen (IOLAR-1) gas was used as the carrier gas and a known volume of sample was injected in the column with the help of the 10µl Hamilton syringe. Different peaks of the samples were identified by comparing their retention times with those of standards. Recovery of analysis was done by fortification experiments and the percentage recovery was 95–98%. Thin layer Chromatography (TLC) was used for confirming the identity of the OCPs already detected by the Gas Chromatograph.The pesticides for which the GC was standardized and were estimated were Dieldrin, isomers of HCH (α, β &γ), metabolites of heptachlor (Heptachlor& heptachlor epoxide), Endosulphan, and DDT (DDE,DDD and DDT).

3. Statistical Analysis

The calculations are based on biological statistics and values are expressed as mean± standard error (S.E.).The difference in the pesticide residue levels between different groups was analyzed with the help of student t test. Significance between the residue levels of different groups was judged at 5 % and 1% levels.

4. Questionnaire

50 questionnaires were filled by the local residents of old part of Bikaner City regarding their health status, dietary habits, age religion etc. Performa of the questionnaire had the followings heads:

1. Name
2. Address
3. Age
4. Parity
5. Socio-economic status
6. Dietary habits
7. Obstetrics history
8. Family history of any disease
9. Medication during pregnancy
10. Addiction to smoke, chewing tobacco, paan masala, supari, alcohol etc.
11. Accidental exposure to pesticides
12. Past and present history of any serious disease and present health condition.
13. Religion
14. Use of pesticide ion the house or nearby fields
15. Other descriptions, if any

Observations and Results

Water Sample Collected from Old City

Values expressed in parts per million (ppm) in the form of range.

Sample No.	HCH	HCH	HCH	Dieldrin	Hepta Chlor	Hepta Chlor epoxide	Endo Sulphan	p'p DDE	p'p' DDD	p'pp DDT
1.	.073-.192	.062-.126	.053-.139	.023-.095	.034-.098	.044-.108	.048-.160	.063-.170	.078-.159	.096-.190

Milk Sample Collected from Old City

Values expressed in parts per million (ppm) in the form of range.

Sample No.	HCH	HCH	HCH	Dieldrin	Hepta Chlor	Hepta Chlor epoxide	Endo Sulphan	p'p DDE	p'p' DDD	p'pp DDT
1.	.123-1.232	.163-1.105	.192-1.392	.157-1.109	.142-1.032	.104-.128	.173-1.213	.172-1.390	.192-1.923	.196-1.732

Abbreviations used in the tables

- n: number of positive samples
- ∑HCH: Total HCH equivalent which is a sum alpha HCH, gamma HCH and beta HCH.
- ∑Heptachlor: Total Heptachlor equivalent which is a sum of Heptachlor and Heptachlor epoxide.
- ∑DDT: Total DDT equivalent which is a sum of DDD, DDE and DDT.
- ppm=parts per million=mg per liter = mg/Liter

S. No.	OCPs detected	Water No. Of samples - 25 Mean ± S. E.		Milk No. Of samples – 25 Mean ± S. E.	
1	α – HCH	0.075 ± 0.0068	(n=25)	0.742 ± 0.0277*	(n=25)
2	β- HCH	0.067 ± 0.0045	(n=24)	0.041 ± 0.0184*	(n=25)
3	γ- HCH	0.059 ± 0.0124	(n=23)	0.058 ± 0.0195	(n=25)
4	Heptachlor	0.042 ± 0.0089	(n=23)	0.168 ± 0.2206*#	(n=25)
5	Dieldrin	0.029 ± 0.0013	(n=25)	0.150 ± 0.0429	(n=25)
6	Heptachlor Epoxide	0.044 ± 0.0649	(n=25)	0.179 ± 0.2184*#	(n=24)
7	DDE	0.874 ± 0.0542*	(n=25)	0.098 ± 0.02211	(n=25)
8	DDD	0.045 ± 0.0152	(n=25)	0.097 ± 0.0029	(n=25)
9	DDT	0.035 ± 0.0079	(n=25)	0.096 ± 0.0054	(n=24)
10	Σ HCH	0.062 ± 0.0174	(n=25)	0.951 ± 0.0291*	(n=25)
11	Σ Heptachlor	0.872 ± 0.0373	(n=25)	0.347 ± 0.4692*	(n=25)
12	Σ DDT	0.121 ± 0.0164*	(n=25)	0.291 ± 0.2284*	(n=25)
13	Endosulphan	0.042+ 0.0081	(n=22)	0.052+ 0.0072	(n=23)

* Statistically Significant P < .05

\# Statistically Significant P < .01

Discussion

The present study reflects the national scene of magnitude of pesticide pollution and also signifies the distribution and accumulation of non-biodegradable lipophilic OCPs in the water and cattle milk commodities on one side and subsequently the vulnerability of the human race on the other side. Despite the fact that the consumption of pesticides in India is still very low, which is about 0.5 kg/ha of pesticides in comparison to 6.60 and 12.0 kg/ha in Korea and Japan, respectively, there has been a widespread contamination of food commodities with pesticide residues, mainly because of non-judicious use of pesticides. 51% of the food commodities in India are contaminated with pesticide residues and out of these, 20% have pesticides residues which are above the maximum residue level values on a worldwide basis. In the present study water and cattle milk samples were collected from different parts of the

Bikaner city of Rajasthan, India, and it was found that mostly all samples have OCPs residues and many of them are statistically significant P< .05 and P< .01. This shows that how these xenobiotics have contaminated our Mother Nature and now human race is facing danger of existence. It can be concluded that the magnitude of pollution is quantitatively enough to contaminate the food and environment and the pesticides reach the human body through various sources mainly by absorption form the gastrointestinal tract through contaminated food chain, are circulated in blood, stored milk and secreted during lactation resulting in sufficient neonatal intake. Since, the pesticides are reported to be carcinogenic, mutagenic, teratogenic, immunosuppressive, induces endocrine dysfunction and high estrogenic activity, disturb the reproductive processes, growth depressants, induces several psychogenic and neurogenic abnormalities in adult stages and are also reported to be associated with abortions, premature deliveries, still births, low birth weight consequences are obvious on the human beings. It also reflects that there is an urgent need to develop less/non persistent and more/total biodegradable pesticides and other means by which we can to reduce the environmental pollution by the pesticides which is not only posing risk to human health but also jeopardising our future generations as well . The above is of special significance for the Indian population, since, Indians have been reported to possess the highest body burden of pesticides.

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