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Organochlorines and Organophosphates in Bovine Milk Samples in Allahabad Region

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ABSTRACT: Organochlorine and Organophosphate pesticides are likely to pollute lipid-containing food sources, like milk (3-5% fat), owing to their lipophilicity, thereby jeopardizing the benefits of milk as a health food. Pesticide load in bovine milk may be from the leaching of pesticides persisting in the adipose tissue of the animal, or through direct contamination during milking or improper handling of milk.Bovine milk samples from small-scale rural and urban dairies were collected and investigated for selected array of Organochlorines and Organophosphates. The study revealed that BHC, is still the most common pollutant being present in 75% of the samples, followed by Methyl Parathion (37.5%), Dieldrin and 2,4 DDE (12.5% each). Mean quantity of Methyl Parathion (0.3496 mg/ kg : whole milk basis) was 1.7 times higher than BHC (mean 0.2104 mg/kg: whole milk basis). The mean value was 0.35 times higher than the ADI prescribed by Prevention of Food Adulteration Act, 1992, India, in case of Methyl Parathion, 1.05 times higher for BHC, 2.30 times for Dieldrin and 0.07 times for 2,4 DDE. Rural dairy samples carried higher load of Methyl Parathion which may be attributed to the large scale agricultural practices and unsafe handling in that area.

Key words: Acceptable Daily Intake, Bovine, Milk, Organochlorines, Organophosphates

INTRODUCTION

India, having agriculture based economy, is one of largest insecticide consumers in the world. Moreover two-thirds of the pesticides consumed are Class I and II pesticides (WHO) which are highly toxic (Fig. 1). No wonder a number of studies from India have reported widespread contamination of various food and water sources to be contaminated with these pesticides.

In India, largest pesticide consumption has been in the state of Uttar Pradesh, according to the data of 1995-1996 and 1999-2000, produced by Central Insecticide Board and Registration Committee, India. Pesticides especially the organochlorine group, having high bioaccumulation potential and low degradation rates, has been associated with a number of environmental effects. Due to their persistence they are distributed in air (Strand and Hov, 1996; Wania and Mackay, 1996; Wenzel et al., 1994) water bodies and soil finally making way into the plant and animal biomass (Colborn, *et al.,* 1993). Studies have reported distribution of these pesticides in the surface and sea waters (Seba and Prospero, 1971) with Northern hemisphere being more polluted than Southern Pine leaves (Eriksson, *et al.*, 1989, Hellstrom, *et al.*, 1989, Jensen, *et al.*, 1992; Kylin and Sjodin, 2003; Wenzel, *et al.*, 1994) and lichens have reported to bioaccumulate these toxins and serve as indicators of atmospheric pollution load. These pesticides have been reported to act as endocrine disruptors (DeRosa, *et al.*, 1998, Scippo, *et al.*, 2007) thereby causing disturbances in fertility of wild animals (Jensen, 2000). In humans pesticides have been reported to be anti-androgenic in males causing sterility and

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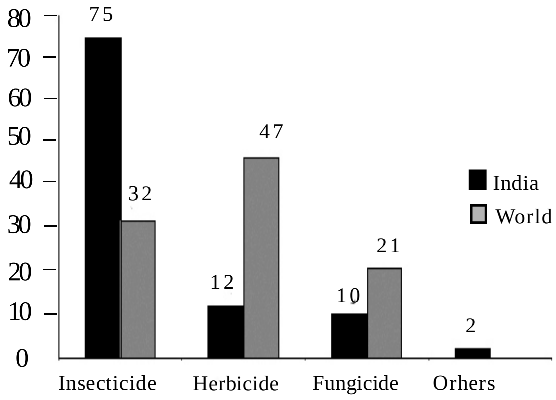

Fig. 1. Comparison of Pesticide consumption of India with the rest of the world (Source: Report of Centre for Science and Environment, India)

carcinogenic in females causing cancers of breast (Reynolds, *et al.*,2004). Indiscriminate use of pesticides and their persistence in environment have led to widespread contamination of food sources such as food grains, vegetables, fruits and animal products. Animal derived products, however, are likely to carry a greater load of pesticides due to their higher lipid content. Bovine Milk, with 3-5% lipid (EPA, 1980) content, therefore, is expected to get contaminated with lipophilic pesticides such as Organochlorines and Organophosphates. The pesticide contamination in bovine milk may be attributed to the food chain contamination of the adipose tissue of the animal and subsequent leaching into milk fat. Secondary contamination is also possible due to direct contamination during milking or improper handling during transportation. Milk constitutes a significant part of human nutrition and is the major contributor of protein in vegetarian diet. Pesticide contamination of milk, therefore, can have far reaching consequences and requires serious monitoring. Pesticides have been proved to have serious hazards to human health (DeRosa, *et al.*, 1998).In the present study, bovine milk samples from small-scale rural and urban dairies in Allahabad District of Uttar Pradesh, India, were collected and investigated for Organochlorines like metabolites of DDT, 2,4 DDE and 4,4 DDE together with Aldrin, Dieldrin, BHC, and Endosulfan. DDT, Dieldrin, BHC and Aldrin are banned in Asia by the Stockholm treaty of 2001(De Almeida and Barretto, 1971). But as their residues are highly persistent and are reported to be sold to unsuspecting consumers, they are still needed to be monitored. Organophosphates investigated included, Methyl Parathion, Dimethoate and Malathion. Organophosphates are not as persistant as Organochlorines but higher quantities due to indiscriminate use of pesticides are suspected and are toxic (Hellstrom, *et al.,* 1989; Jensen, 2000).

MATERIALS & METHODS

Urban dairy milk samples were collected from a dairy located in Allahabad West region while the rural milk samples were taken from individual milk suppliers who collect milk from villages of Allahabad and transport and sell it to consumers in urban Allahabad region. Milk samples were refrigerated at 4 ^{o}C until analyzed, generally within 24 hrs. The milk lipids were extracted following procedure of Environmental Protection Agency Protocol (EPA-1985). The procedure involved denaturation, solvent extraction, centrifugation, extraction of organic layer and concentration. The concentrated extract was run on gas chromatograph for estimation of Organochlorine and Organophosphate insecticides. One µL aliquots of the sample were injected in GC (Perkin Elmer Autosystem XL) equipped with a selective Ni ECD and capillary column-i.d. 0.25 mm film, length 30 m, packed with PE-17, with Nitrogen carrier gas with a flow rate of 3 psi employing a split mode. The oven temperatures were kept at 190-280 °C with a ramp of 5°C/min. The samples were calibrated against 0.1, 1 and 10 ppm standard solutions for all pesticides. Standards were obtained from Facility for Ecological and Analytical Testing, IIT Kanpur. Levels of pesticides in samples were directly determined after calibration using Totalchrom software. Recovery of residues from samples was 70-79% Results were not corrected for recovery.

RESULTS & DISCUSSION

The results presented in Table 1 are reported in mg/kg on whole milk basis.The total mean of Methyl Parathion was 0.3496 mg/kg while the total percentage positive was 37.5%. Acceptable Daily Intake (ADI) listed in the Prevention of Food Adulteration Act, 1992 was used for analysis of the results. On the basis of these limits the mean values of the pesticides reported was 0.35 times higher in case of Methyl Parathion, 1.05 times higher in case of BHC, 2.30 times for Dieldrin and 0.07 times for 2,4 DDE. Rural dairy samples carried higher load of Methyl Parathion which may be attributed to the large scale agricultural practices in that area and hence greater chance of exposure to insecticides. However, urban dairy milk samples also show widespread contamination with Methyl Parathion. Higher levels in rural samples may be due leaching of these lipophilic chemicals from the adipose tissue of cattle or due to secondary contamination from widespread usage in the rural areas. In rural areas Aldrin is used in termite control in houses. Therefore, its metabolite Dieldrin is present in 12.5% of the samples. Due to widespread usage of BHC for insect eradication and seed treatment in agricultural practices and also in termite control it has emerged as the major contaminant. In rural areas, dairies are managed by people who are engaged in agricultural practices hence there is greater chance of secondary contamination. While in urban areas, dairymen are not handling pesticides. The pesticides residues detected in milk are therefore from cattle's adipose tissue. Forage or water contaminated with these pesticides may be the source of intake of these chemicals by animals in urban dairies. Studies on dairy milk samples have been undertaken in India (Eriksson, *et al.*, 1989; http://www.epa.gov), but they have largely focused on Organochlorines. This study clearly shows that Organophosphates also need monitoring as after the imposition of ban on many Organochlorines. Organophosphates are advocated as better alternatives for highly persistant and toxic OCs but due to their low cost and high persistence, better effectiveness and lesser number of applications, OCs are still the preferred group of pesticides among the rural population. Lack of awareness about the toxic potential of OC pesticides among the largely illiterate or less educated rural population is making it difficult for the Government agencies to completely check the use of OCs. Though the levels of these pesticides have shown a considerable drop over the years and it is therefore no residues could be found in the Urban dairy milk samples. Comparison with other studies shows higher level of dieldrin (De Almeida and Barretoo, 1971) and higher percentage of endosulfan (Steffey, *et al.*, 1972). Although aldrin has reported in other studies is completely absent. Previously, Kannan, *et al.*, (1992) have reported highest levels of organochlorine contaminants in Indian foodstuffs. The results are in agreement with this study.

CONCLUSION

Though Organophosphates have emerged as the major pollutant, OCs is also present as contaminant. The mean levels of the OC contaminants exceed the ADI limits. Thus further studies on risk assessment and health effects due to unsafe intake of these toxic pesticides on the local population are warranted. This also calls for conduction of awareness programs on toxic effects and safe handling of pesticides in rural areas which are highly exposed.

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Table 1. OC and OP insecticides detected in rural and urban milk samples in Allahabad.

Pesticides	Rural Dairy Samples		Urban Dairy Samples	
OrganoChlorine	% Positive	Mean	% Positive	Mean
2,4 DDE	12.5	0.0857	-	-
4,4 DDE	-	-	-	-
Aldrin	-	-	-	-
Dieldrin	12.5	0.3446	-	-
BHC	75.0	0.2104	-	-
Endosulfan	-	-	-	-
Organophosphates				
Methyl Parathion	14.29	0.5200	100	0.1792
Malathion	-	-	-	-
Dimethoate	-	-	-	-

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