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Organochlorine Pesticide Residues in Herbal Ayurvedic Preparations

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The use of plant materials for preparations of ayurvedic tonics has been known since ancient periods (Anonymous 1974). The use of digestive candies and health tonics are at great demand in India. Further, the enormous use of organochlorine pesticides (OCPs) in developing countries has been of serious concern because of their persistant nature. The presence of OCPs in terrestrial and aquatic environment may lead to toxicological implications (Aruda et al. 1988, Cochieri and Amese 1988, Sarkar and Gupta 1988, Dikshith et al. 1989). Large amount of pesticides are used in agriculture sector and public health programmes every year (Pesticide Manual 1997; Ciers 1998). Continuous use of OCPs lead to their presence in water, soil, air, crop plants and biological tissues. Herbal products prepared using parts of the plants for human consumption used as drugs, tonics, toiletories, cosmetics etc. may also contain significant level of OCP residues. Though, pesticides residue analysis have been done in several food/food products (Raizada et al. 1998). There is hardly any information on herbal ayurvedic preparations. The present study deals with the analysis of OCP residues in commonly used herbal ayurvedic preparations of the local market.

MATERIALS AND METHODS

All the solvents used in extraction and cleanup processes were of HPLC grade procured from M/s Spectrochem Pvt. Ltd. Bombay. Activated charcoal, sodium sulfate, sodium chloride and other chemicals used in the study were procured from E. Merck India Ltd., BDH, Glindia Ltd. etc.

Total twenty five samples of herbal preparations of different brands were collected from local market of Lucknow. These preparations are taken orally.While digestive candy is chewed, the digestive churna is

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swallowed. Chyvanpras is a paste and ashava and aristha are liquid. The major constituents of these products are given in table 1.

Table 1. List of major constituents of herbal preparations Analysed.

Product	Constituents
Digestive Candy	Emblica officinalis, Citrus aurantifolia, Carmun carvi, Zinger officinale, Mango, Piper longum, Piper nigrum, Ammonium chloride, Black salt, Potasium chloride, sugar
Digestive Churna	Sea salt, Black salt, Ammonium chloride, Sugar, Citrus aurantifolia, Carmum carvi, Piper nigrum, Piper longum
Chyvanpras	Milk fat, Sugar, Emblica officinalis, Piper longum, Puerariatuberosa, Sesamum indicum
Ashava (Drakshashava)	Vitis vinifera
Aristha (Dasmularistha)	Aegle marmleos, Oroxylum indicum, Gmelina arborea, Sterospermum suaveolena, Premna latifolia, Desmodium angeteum, Uraria picta, Solanum surattense, Solonum inducum, Tribulus terrestris

Analysis was carried out as per AOAC(1995). Each sample in triplicate was taken (200 ml liquid or 50 grams solid) in 500 ml conical flask mixed with 3x100ml (n-hexane : Acetonitrile mixtrure 1:3) and kept for over night and shaken for 30 min. Organic phase was separated from aqueous phase and collected togather. Aqueous phase was further extracted 3 times with 50ml n-hexane in separatory funnel. Total pool of hexane extract was washed with distilled water 3 times. Washed hexane extract was further passed through charcoal and anhydrous sodium sulfate column. The clean hexane extract was concentrated and made to 5 ml in a clean volumetric flask. Aliquots of above concentrate were injected to precalibrated GC machine equipped with ^{63}Ni electron capture detector.

Table 2. HCH residues (ppb) in herbal preparations.

Preparations	alfa -HCH	beta -HCH	gamma -HCH	delta -HCH	Total HCH
D. Candy	3.1 (0.3-2.3)	11.7 (7.4-16)	3.0 (2.5-3.6)	5.6 (4.3-7.0)	23.0 (16.5-29.6)
D.Churna	4.1 (3.7-7.0)	10.4 (5.4-20.7)	4.6 (1.5-7.0)	3.2 (3.5-7.0)	23.0 (17.0-39.5)
Chyvanprash	1.8 (0.2-5.0)	4.5 (0.4-12.0	1.0 (0.4-5.2)	2.1 (0.3-4.0	9.4 (1.0-20.0)
Ashava	0.6 (0.2-0.9)	1.6 (1.6-2.7)	0.5 (0.4-0.8)	0.7 (ND-0.7)	3.4 (2.8-4.3)
Arishtha	0.6 (0.2-1.1)	4.2 (ND-4.2)	0.7 (ND-0.7)	0.2 (ND-0.2)	5.7 (3.7-16.7)

Table 3. DDT residues (ppb) in herbal preparations.

Preparations	pp-DDE	op-DDT	pp-DDD	pp-DDT	Total DDT
D-Candy	2.1 1.9-2.3)	ND	2.5 (2.0-3.0)	3.5 (3.0-4.0)	8.1 (ND-9.0)
D-Churna	0.9 (0.3-2.0)	ND	0.3 (0.2-0.8)	6.2 (4.0-6.4)	7.5 (ND-7.5)
Chyvanprash	ND	21.2 (2.0-45.0)	9.1 (8-10.0)	24.6 (20.-47)	55.0 (ND-65.0)
Ashava	0.5 (ND-0.5)	6.0 (ND-6.0)	ND	ND	6.5 (ND-6.5)
Arishtha	0.4 (ND-0.5)	0.4 (ND-0.4)	ND	ND	0.8 (ND-0.8)

Average of five samples in triplicate; Values in paraenthesis indicate the range
D = Digestive ; ND= not detected.

A glass column (1.5m x 2mm id) packed with 1.5%OV-17 + 1.95% Qf-1 on 100-120 mesh chromosorb WHP was used. Operation temperature were programmed at 195, 200, 220°C for column, injector and detector respectively. Purified nitrogen gas passing through silicagel and molecular sieves was used as carrier gas at flow rate of 60ml/min. Limit of detection was o. 1 to 0.5 ppb for organochlorine pesticides analysed. Data for GC analysis were further confirmed by TLC. Periodically procedural blanks were used to check cross contamination. Recovery studies with purified samples indicated that overall recovery value exceeded 80%. Identification and quantificxation were accomplished using known amount of external standered recieved from US EPA, Pesticides and Industrial Chemicals repository (MD-8) Research triangle, NC,USA.

RESULTS AND DISCUSSION

It is evident from the results (Table 2 and 3) that residues of HCH were more than residues of DDT in all the preparations analysed except chyvanrash and ashavas where DDT residues (55.0 and 6.5 ppb) was more than that of HCH (9.4 and 3.4 ppb). The contamination of HCH was 23.0 ppb in both and DDT 8.1 ppb and 7.5 ppb in digestive candy and digestive churna respectively. However DDT contamination was maximum in chyvanprash (Table 3). The residue level pattern in digestive candy, digestive churna and aristha was HCH > DDT. Contrary to this in chyvanprash and ashava DDT > HCH. As given in tables the level of beta-HCH was maximum in all the samples. The concentration of gamma-HCH was lower in chyvanprash arishthas and ashavas. However, delta-HCH level was lower in ashavas and aristhas. Report have shown that gamma isomer of HCH is more succeptible to degrade in biological environment (Benezet and Matsmura 1973, Jangnow et al. 1977, Mathur and Saha 1977). It is evident from tables 1 and 2 that the level of both the isomers of DDT was highest in chyavanprash only, DDT undergoes metabolic conversion and dehydrochlorination (Matsumura 1977). The presence of pp'DDE and pp-DDD in present study might be due to such metabolic conversions. OCPs may originate in these herbal preparations from contamination via spillage and volatalisation of residues from pesticide treated soils. Though, the pesticides like HCH and DDT are well known toxic chemicals (WHO 1974, Bulger and Kupfer 1985), their residues levels were very low in all the samples, possibily because of

multifold processing of herbs in preparation of these products. Even their presence in herbal preparations may pose a problem with products being exported. Though organochlorine pesticides are persistant in nature. Even organochlorine pesticides were also detected in human diet (Kannan et al. 1992) and drinking water (Dikshith et al. 1990). Certain physical and chemical conditions like heat treatment, steaming, treatment with supercritical carbon dioxide etc., are known to reduce the level of OCPs (Stahl and Rau 1984; Nash 1984). Further monitoring of organochlorine pesticides in large number of herbal products is needed to conclude something regarding MRL in herbal products etc.

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