Human health risk from hexachlorocyclohexane and dichlorodiphenyl-trichloroethane pesticides, through consumption of vegetables: estimation of daily intake and hazard quotients

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Abstract

People are encouraged to consume more fresh vegetables due to the high nutritional value and fibre contents in vegetables. At the same time, due to pesticidal residue in vegetables, vegetable consumption is considered as an important route of pesticides uptake by human beings. Pesticides, particularly organochlorines such as Aldrin, heptachlor and isomers of dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) are a major concern because of their persistence, bioaccumulation, toxicity, and long-range environmental transport ability. Hence information on such toxic contaminants in vegetables is very essential, due to human health concern. In order to assess the potential health risks associated with vegetable consumption due to pesticidal residues of DDT and HCH, sixty four samples of commonly available vegetables were collected from a local market in West Bengal, India and analysed for DDT and HCH. In this study, estimated average daily intake of HCHs and DDTs through vegetables was ranged between 5.0×10^{-8} – 9.1×10^{-6} mg kg⁻¹ day⁻¹ with an average value of 2.0×10^{-6} mg kg⁻¹ day⁻¹. Human health risk was assessed in terms of incremental lifetime cancer risk and health hazard index was between 1.7×10^{-8} – 8.8×10^{-6} and 1.5×10^{-7} – 1.7×10^{-2} , respectively. Generated results were compared with maximum residual levels and reference values, were within acceptable limits and suggested no health risk to the human populations. Therefore, an extensive study on vegetables with the inclusion of other persistent organic pollutants and toxic metals should be carried out to know the overall scenario of contamination of vegetables.

Introduction

Dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) are persistent bioaccumulative contaminants that are found ubiquitously in environment and biological matrices, including tissues of fish, birds and mammals.^{1,2} These toxic organic pollutants compounds can travel longer distance by long range atmospheric transport thereby affecting regions far from their release sources³ and therefore listed by the Stockholm Convention as persistent organic pollutants.⁴ The use of DDT and HCH has been banned or severely restricted in several countries, but these are still being produced, exported, and used in some countries due to their low cost and versatile uses.^{5,6}

India is the fourth largest pesticide producer in the world after the US, Japan and China; domestic production of pesticides is approximately 85,000 MT and about 60,000 MT used annually. The organochlorine pesticides and Malathion accounts for about 71% of that used.^{7,8} DDT and technical HCH were earlier extensively used until 1989 and 1997, respectively. After this time, Government of India has withdrawn the use of DDT and HCH in agriculture, but restricted for public health purpose only as per World Health Organization guidelines.⁹

These compounds are much of concern for human health because of their wide variety of toxicity. Human exposure to DDT and HCH has been associated with hormonal-disruption, arthritis, diabetes and development of cancer.¹⁰⁻¹⁴ Robison *et al.*¹⁵ reported that technical DDT and *o,p'*-DDT, the most estrogenic component of technical DDT, can support the growth of estrogen-dependent breast tumors in rats. Humans are exposed to these toxic contaminants, mainly through the consumption of contaminated food and occupational environments. Therefore, health risk assessment due to pesticidal residues has been conducted worldwide.¹⁶⁻¹⁸

Significant level of residues of HCHs and DDTs has been found in the biotic¹⁹⁻²² and abiotic²³⁻²⁸ compartment in the past and also in the recent years in India. Although study on HCH and DDT residues in vegetables have been carried out in India²⁹⁻³⁸ but, quantitative health risk from HCH and DDT in vegetables from India was rarely investigated.³⁹ Majority of people in India are vegetarian and everyday people are being encouraged to consume more vegetables. Vegetables are important components of the human diet due to their high nutritional value and fiber contents. The domestic consumption of vegetables in the urban and rural areas has increased in the recent years. Thus Information on toxic con-

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taminants in vegetables is very essential for protection of human health, because the main non-occupational route of exposure to toxicants is through dietary intake.⁴⁰ This study was undertaken to assess the daily dietary intake of HCH and DDT pesticide through vegetables and their potential health risks in terms of incremental lifetime cancer risk (ILCR) and non-cancer health hazard index (HI) for human beings. For this purpose, primarily we estimated lifetime average daily dose (LADD) of HCH and DDT isomers for human adults through vegetable intake. The LADD is the amount of a chemical intake by a person per kg of body weight per day which may suspect the adverse health effects when absorbed into the body over a long period of time, either through inhalation, ingestion or dermal contact.⁴¹ Secondary, human health risk in terms of upper-bound excess probabilistic ILCR and non-carcinogenic health hazards was estimated.⁴¹ ILCR is the probability of cancer development in a lifetime after a uniform exposure to any carcinogenic chemical, while non-carcinogenic health hazards as HI is the measure of the magnitude of exposure potential or a quantifiable potential for developing non-carcinogenic health effects after averaged exposure period.⁴²

Materials and Methods

Solvents, chemicals and standards

High performance liquid chromatography grade solvents (acetone, methanol, dichloromethane, and hexane) and analytical grade chemicals (sodium sulphate, silver nitrate, potassium hydroxide, activated charcoal and sulphuric acid) were purchased from Merck (Delhi, India). Silica gel 60 (0.063–0.100 mm) was from Supelco (Sigma-Aldrich Co., St. Louis, MO, USA). Silica gel and granular anhydrous sodium sulphate was cleaned in Soxhlet extractor with methanol, dichloromethane and acetone for 8 h each, then stored in air tight desiccator until use. Pesticide standard mixture stock was prepared by diluting the commercial standard solutions [*p,p'*-DDT, *o,p'*-DDT, *p,p'*-dichlorodiphenyldichloroethylene (DDE), *p,p'*-dichlorodiphenyldichloroethane (DDD), α -HCH and γ -HCH] purchased from Supelco (Sigma-Aldrich) and the working standard solution was prepared by diluting the standard stock solutions in hexane.

Sample collection and extraction

The selected vegetables were collected from local markets of West Bengal, India. Vegetables namely, carrot (*Daucus carota* L.), cauliflower (*Brassica oleracea*), eggplant (*Solanum melongena* L.), radish (*Raphanus sativus* L.), spinach (*Spinacia oleracea* L.), and tomato (*S. lycopersicum* L.) collected in clean polyethylene bags, labelled and transported on ice to the laboratory.

In the laboratory, after washing with double distilled water and wiping with tissue paper, vegetables cut into small pieces with a grater and thoroughly mixed. For extraction of pesticides, samples were mixed with anhydrous sodium sulphate and extracted three times with acetone on a mechanical shaker. The acetone extract was filtered with vacuum suction and concentrated to near 20–25 mL using a rotatory vacuum evaporator (Buchi, Essen, Germany). The concentrated extracts were subjected to liquid-liquid partitioning three times with hexane in a separatory funnel. Hexane layer with residues was collected by passing the sample through sodium sulphate and concentrated to ~5 mL.

Extract clean-up for removal of pigments and interferences

Concentrated sample extracts were primarily cleaned to remove pigments. Pigments were removed by passing through a chromatographic glass column containing activated charcoal and anhydrous sodium sulphate. Secondary, multilayered silica gel column chromatography was performed for removal of interfering compounds and other aliphatic compounds. Briefly,

a chromatographic glass column (300×30 mm) was packed from bottom up with 2.5 g activated silica gel, 4.0 g silver nitrate silica gel, 2.5 g activated silica gel, 4.0 basic silica gel, 2.5 g activated silica gel, 12.0 g acid silica and 5.0 g anhydrous sodium sulphate. Preliminary, before loading of sample extract, column was rinsed with 50 mL n-hexane. Subsequently, after loading the sample extract, elution of analytes was carried out using 170 mL hexane, and eluted extract was concentrated under slow stream of purified laboratory grade nitrogen using Rotatory Vacuum evaporator and Turbo Vap (Caliper, Princeton, NJ, USA) to 2.0 mL. The concentrated clean extract was transferred to a sample vial and 1 μ L was manually injected onto a gas chromatograph equipped with an electron capture detector for quantification of HCH and DDT isomers.

Sample analysis and analytical quality control

The identification and quantification of DDTs (*p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE and *p,p'*-DDD), and HCHs (α -HCH and γ -HCH) in clean sample extracts was carried out, using a gas chromatograph (3400cx, Varian Star, Milpitas, CA, USA) equipped with ^{63}Ni electron capture detector. A capillary column (RTX-5) (30 m × 0.25 mm id) coated with a 0.5 μ m of stationary phase (5% diphenyl 95% dimethylpolysiloxane), was used for the separation of the compounds. The column oven temperature was initially maintained at 170°C and programmed to 220°C (at 7°C min⁻¹); temperature was again increased to 250°C (at 5°C min⁻¹) and held for 7.0 min. The temperature of injector and detector was maintained at 250 and 325°C respectively. A purified laboratory grade, nitrogen gas was used as carrier (at 1.0 mL min⁻¹) during all the analysis.

Reference standard solutions were used for instrument calibration and other quality control studies. Concentrations of target compounds were determined with an external standard method, comparing peak area in samples with the standards using. Peak identification was conducted with the accurate retention time of each standard. Before analysis, five levels of calibration curves of standards were prepared with r^2 value of 0.999. Each sample was analyzed in duplicates and average values obtained from duplicate analysis of each sample was used in calculations. For quality assurance/quality control analyses, procedural blanks was analyzed to check for loss and cross contamination (analyte concentrations were below the detection limits). Random duplicate sample analysis (standard deviation <10%), calibration verification (standard deviation <10%), and matrix spiked study was carried out to maintain quality control of analysis. Sample spiked recoveries with known working standard solutions were

assumed to be satisfactory for each compound, and those were in the ranges of 70–110%. Method detection limits were obtained by processing replicate of a sample spiked with a quantity sufficient to produce a detectable response ($s/n > 3$) and multiplying the standard deviation by the t_{students} value for processed replicates. Statistically calculated value of <MDL (explain abbreviation) used for all HCH and DDT isomers was <0.01 ng g⁻¹. Levels below 0.01 $\mu\text{g kg}^{-1}$ (reporting limit) were not used in calculation and considered as zero. Analytical results were not corrected for blank (<MDL) and recoveries (within acceptable range), and reported on wet weight basis. Further, it may be worthy to mention that our laboratory had been participating in proficiency testing exercises conducted by international agencies with satisfactory score of performance for organochlorine pesticides including HCHs and DDTs.

Human health risk assessment

Considering the toxicological effects of HCHs and DDTs, it is important to investigate the potential exposure of human through dietary intakes. In this study, HCHs and DDTs intake through vegetables was estimated as LADD. ILCR and non-cancer health hazard [hazard quotient (HQ)] to humans was assessed using the recommended risk assessment guidance:⁴¹

$$\text{LADD (mg kg}^{-1} \text{ day}^{-1}) = (\text{Cs} \times \text{IR} \times \text{F} \times \text{EF} \times \text{ED}) / (\text{BW} \times \text{AT})$$

$$\text{ILCR} = \text{LADD} \times \text{CSF}$$

$$\text{HQ} = \text{LADD} / \text{RfD}$$

where, Cs is the pesticide concentration in vegetables (mg kg⁻¹), IR is the vegetable intake rate (137 g d⁻¹, single or mixed with others), F is the unit conversion factor, EF is exposure frequency (365 days/year), ED is the life time exposure duration (70 years), BW is the body weight (70 kg), and AT is the averaging time for carcinogens (EF × ED). CSF and RfD is oral cancer slope factor and reference dose respectively, for individual compound (mg kg⁻¹ d⁻¹).⁴³

Results

Residue levels of hexachlorocyclohexane and dichlorodiphenyltrichloroethane

Concentration means and ranges of HCH and DDT residues in different vegetables have been given in Table 1. In this study we studied the two major isomers of HCH (α -HCH and γ -HCH). α -HCH is main component of technical HCH while γ -HCH is the major component of lindane formulation. The observed level of α -HCH and γ -HCH was ranged between 0.05–1.55

$\mu\text{g kg}^{-1}$ and $0.06\text{--}6.61 \mu\text{g kg}^{-1}$ and their mean was 0.56 and $1.37 \mu\text{g kg}^{-1}$ respectively. The average concentrations of DDT isomers was $0.60 \mu\text{g kg}^{-1}$, $1.02 \mu\text{g kg}^{-1}$, $1.09 \mu\text{g kg}^{-1}$ and $0.30 \mu\text{g kg}^{-1}$, respectively for α , p,p' -DDT, o,p' -DDT, p,p' -DDE and p,p' -DDD.

Human health risk estimates

Human health risk assessment was based on assumption that human being exposed to HCH and DDT pesticides through vegetables may cause harmful effects on humans. Human health risk due to exposure to HCHs and DDTs through vegetables was assessed, considering that humans take vegetables all the days in a year during the life span of 70 years. Risk was assessed by estimating the incremental LADD

and subsequently their ILCR. The life time average daily dose is the amount of pollutant intake per kg of body weight per day, which may cause adverse health effects when absorbed into the body over a long period of time. The comparative LADD, ILCR and HI for humans from HCH and DDT exposure through studied vegetables has been shown in Figure 1.

Estimated LADD of individual HCH and DDT isomers and their total (HCHs+DDTs) was presented in Table 2. The estimated LADD of total pesticides from carrot, cauliflower, eggplant, radish, spinach and tomato was ranged between 8.5×10^{-8} – $2.1 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, 5.0×10^{-8} – $4.3 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, 3.1×10^{-7} – $1.3 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, 7.2×10^{-7} – $9.1 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, 6.8×10^{-8} – $2.0 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$ and 4.1×10^{-7} – $2.9 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$,

respectively. The average LADD was $1.1 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, $2.5 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, $7.8 \times 10^{-7} \text{ mg kg}^{-1} \text{ d}^{-1}$, $3.4 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, $1.0 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$ and $1.6 \times 10^{-6} \text{ mg kg}^{-1} \text{ d}^{-1}$, respectively from carrot, cauliflower, eggplant, radish, spinach and tomato.

Human health risk in terms of ILCR from HCHs and DDTs in vegetables has been estimated and presented in Table 3. Total ILCR was varied between 2.7×10^{-8} – 2.6×10^{-6} (mean, 1.2×10^{-6}), 1.7×10^{-8} – 5.3×10^{-6} (mean, 2.9×10^{-6}), 2.8×10^{-7} – 2.2×10^{-6} (mean, 1.3×10^{-6}), 2.3×10^{-7} – 8.8×10^{-6} (3.9×10^{-6}), 2.1×10^{-8} – 1.6×10^{-6} (mean, 1.0×10^{-6}) and 6.5×10^{-7} – 5.2×10^{-6} (mean, 2.8×10^{-6}), respectively from carrot, cauliflower, eggplant, radish, spinach and tomato intakes.

Table 1. Concentrations of hexachlorocyclohexane and dichlorodiphenyltrichloroethane pesticides in different vegetables ($\mu\text{g kg}^{-1}$).

Vegetables (no.) [*]	Pesticides compound concentrations						ΣOCPs	Isomeric ratio		
	α- HCH	γ- HCH	p,p'- DDT	o,p'- DDT	p,p'- DDE	p,p'- DDD		α/γ- HCH	o,p'/p,p'- DDT	DDT/ (DDE+DDE)
Carrot (12)	0.31-0.64 (0.48) ^o	0.59-0.84 (0.72)	0.13-0.28 (0.19)	0.13-3.14 (1.23)	0.10-0.19 (0.15)	0.04-1.19 (0.39)	0.18-4.46 (2.18)	0.53-0.76 (0.64)	0.55-2.38 (1.49)	1.57-1.88 (1.23)
Cauliflower (13)	0.09-1.24 (0.56)	0.18-5.34 (1.73)	0.43-1.68 (0.97)	0.13-0.43 (0.24)	0.10-6.29 (2.01)	0.05-0.16 (0.11)	0.10-8.99 (5.22)	0.09-3.37 (0.73)	0.08-0.57 (0.27)	0.21-3.31 (1.52)
Eggplant (9)	0.05-0.62 (0.25)	0.06-2.24 (0.77)	0.12-0.23 (0.17)	0.09-0.31 (0.18)	0.04-0.63 (0.20)	0.04-0.08 (0.07)	0.65-2.77 (1.63)	0.09-2.12 (0.84)	0.69-2.50 (1.18)	0.52-3.98 (3.11)
Radish (7)	0.10-1.53 (0.94)	0.20-6.61 (1.88)	0.15-1.82 (0.85)	0.86-11.08 (3.76)	0.07-3.28 (1.04)	0.08-2.92 (0.95)	1.50-18.81 (7.00)	0.23-2.46 (1.15)	0.68-2.83 (1.88)	1.20-2.57 (1.75)
Spinach (15)	0.05-0.24 (0.18)	0.69-1.15 (0.99)	0.09-0.19 (0.13)	0.11-1.60 (0.51)	0.45-1.06 (0.69)	0.03-0.06 (0.05)	0.14-4.12 (2.12)	0.05-0.33 (0.19)	1.22-2.95 (2.08)	0.26-1.55 (1.04)
Tomato (8)	0.06-1.55 (0.67)	0.09-3.64 (1.04)	0.12-1.71 (0.46)	0.09-1.69 (0.87)	0.04-0.22 (0.11)	0.03-0.93 (0.27)	0.84-5.91 (3.25)	0.04-5.69 (1.70)	0.65-0.99 (0.79)	1.45-3.45 (2.52)
All samples (64)	0.05-1.55 (0.56)	0.06-6.61 (1.37)	0.09-1.82 (0.60)	0.09-11.08 (1.02)	0.04-6.29 (1.09)	0.03-2.92 (0.30)	0.10-18.81 (4.18)	0.04-5.69 (0.86)	0.08-2.95 (0.82)	0.21-3.98 (1.77)

^aNumber of samples, ^oaverage in parenthesis. HCH, hexachlorocyclohexane; DDT, dichlorodiphenyltrichloroethane; OCP, organochlorine pesticides; DDE, dichlorodiphenyldichloroethylene.

Table 2. Estimated average daily dose ($\text{mg kg}^{-1} \text{ d}^{-1}$) of hexachlorocyclohexane and dichlorodiphenyltrichloroethane through vegetable to humans.

Vegetables (no.) ^a	ADD of pesticides compounds						Total ADD
	α -HCH	γ -HCH	p,p' -DDT	o,p' -DDT	p,p' -DDE	p,p' -DDD	
Carrot (12)	1.5×10^{-7} – 3.1×10^{-7} (2.3×10^{-7}) ^o	2.8×10^{-7} – 4.1×10^{-7} (3.4×10^{-7})	6.3×10^{-8} – 1.3×10^{-7} (9.3×10^{-8})	6.2×10^{-8} – 1.5×10^{-6} (5.9×10^{-7})	5.0×10^{-8} – 9.2×10^{-8} (7.1×10^{-8})	2.1×10^{-8} – 5.7×10^{-7} (1.9×10^{-7})	8.5×10^{-8} – 2.1×10^{-6} (1.1×10^{-6})
Cauliflower (13)	4.4×10^{-8} – 6.0×10^{-7} (2.7×10^{-7})	8.4×10^{-8} – 2.6×10^{-6} (8.3×10^{-7})	2.0×10^{-7} – 8.1×10^{-7} (4.7×10^{-7})	6.3×10^{-8} – 2.1×10^{-7} (1.2×10^{-7})	5.0×10^{-8} – 3.0×10^{-6} (9.7×10^{-7})	2.5×10^{-8} – 7.6×10^{-8} (5.4×10^{-8})	5.0×10^{-8} – 4.3×10^{-6} (2.5×10^{-6})
Eggplant (9)	2.6×10^{-8} – 3.0×10^{-7} (1.2×10^{-7})	2.9×10^{-8} – 1.1×10^{-6} (3.7×10^{-7})	5.5×10^{-8} – 1.1×10^{-7} (8.0×10^{-8})	4.2×10^{-8} – 1.5×10^{-7} (8.5×10^{-8})	1.9×10^{-8} – 3.0×10^{-7} (9.7×10^{-8})	1.8×10^{-8} – 4.0×10^{-8} (3.2×10^{-8})	3.1×10^{-7} – 1.3×10^{-6} (7.8×10^{-7})
Radish (7)	4.8×10^{-8} – 7.4×10^{-7} (4.5×10^{-7})	9.8×10^{-8} – 3.2×10^{-6} (9.1×10^{-7})	7.1×10^{-8} – 8.8×10^{-7} (4.1×10^{-7})	4.2×10^{-7} – 5.3×10^{-6} (1.8×10^{-6})	3.2×10^{-8} – 1.6×10^{-6} (5.0×10^{-7})	3.8×10^{-8} – 1.4×10^{-6} (4.6×10^{-7})	7.2×10^{-7} – 9.1×10^{-6} (3.4×10^{-6})
Spinach (15)	2.6×10^{-8} – 1.1×10^{-7} (8.6×10^{-8})	3.3×10^{-7} – 5.5×10^{-7} (4.8×10^{-7})	4.4×10^{-8} – 9.2×10^{-8} (6.5×10^{-8})	5.2×10^{-8} – 7.7×10^{-7} (2.5×10^{-7})	2.1×10^{-7} – 5.1×10^{-7} (3.3×10^{-7})	1.6×10^{-8} – 2.9×10^{-8} (2.2×10^{-8})	6.8×10^{-8} – 2.0×10^{-6} (1.0×10^{-6})
Tomato (8)	2.8×10^{-8} – 7.5×10^{-7} (3.2×10^{-7})	4.3×10^{-8} – 1.8×10^{-6} (5.0×10^{-7})	5.9×10^{-8} – 8.2×10^{-7} (2.2×10^{-7})	4.5×10^{-8} – 8.1×10^{-7} (4.2×10^{-7})	2.1×10^{-8} – 1.1×10^{-7} (5.5×10^{-8})	1.4×10^{-8} – 4.5×10^{-7} (1.3×10^{-7})	4.1×10^{-7} – 2.9×10^{-6} (1.6×10^{-6})
All samples (64)	2.6×10^{-8} – 7.5×10^{-7} (2.7×10^{-7})	2.9×10^{-8} – 3.2×10^{-6} (6.6×10^{-7})	4.4×10^{-8} – 8.8×10^{-7} (2.9×10^{-7})	4.2×10^{-8} – 5.3×10^{-6} (4.9×10^{-7})	1.9×10^{-8} – 3.0×10^{-6} (5.2×10^{-7})	1.4×10^{-8} – 1.4×10^{-6} (1.4×10^{-7})	5.0×10^{-8} – 9.1×10^{-6} (2.0×10^{-6})

^aNumber of samples; ^oaverage in parenthesis. ADD, average daily dose; HCH, hexachlorocyclohexane; DDT, dichlorodiphenyltrichloroethane; DDE, dichlorodiphenyldichloroethylene; DDD, dichlorodiphenyldichloroethane.

Discussion

Possible sources identification

HCH formulations are available as technical HCH and lindane. Technical HCH contains 55-80% and 8-15% α -HCH and γ -HCH respectively, while lindane contains >90% of γ -HCH. Therefore, ratio of α -HCH to γ -HCH has been used for identification of possible HCH sources. The ratio of α -HCH to γ -HCH between 3 and 7 infer the input of technical HCH.⁴⁴ However, lindane source are characterized by reduced ratio (≤ 1). A higher ratio (>7) of α -HCH to γ -HCH was explained by long-range transport or recycling of technical HCH, because α -HCH has a longer atmospheric lifetime than γ -HCH by about 25%.⁴⁵ γ -HCH may be transformed by sunlight and through biological degradation into α -HCH.^{46,47} The α -HCH to γ -HCH isomers ratio (α/γ ratio) in this study was ranged between 0.04 to 5.69 with a mean of 0.82 which reflect the local input of technical HCH as well as lindane formulation. The usage of technical HCH and lindane has been banned in agriculture but, used in public health programmes to control vector borne diseases.⁶

The pattern of individual DDT isomers may presume the hypothesis of DDT sources.⁴⁸ Three major isomers (DDT, DDD and DDE) have been used for assessing the chronology of DDT input. Changes in the ratio between DDE and DDD to DDTs have been regarded as an indication of either no, or decreasing, inputs of DDT. Ratio of DDT/(DDD+DDE) greater than 1 indicates past DDT inputs in last five years, and lower ratios suggests aged DDT sources (possibly microbial degradation).^{49,50} The average ratio of DDT/(DDD+DDE) for this study was 1.77, indicating past usage of DDT. Generally, technical

grade DDT constitute 77.1 % (p,p' -DDT), 14.9 % (o,p' -DDT), 4.0 % (p,p' -DDE), 0.3 % (p,p' -DDD), and 0.1 % (o,p' -DDD).⁵¹ However, in this study we observed that p,p' -DDE and o,p' -DDT were the dominant isomers. The vapour pressure of o,p' -DDT is 7.5 times higher than p,p' -DDT leading to greater volatilization of o,p' -DDT to the atmosphere and p,p' -DDT metabolizes much faster in a tropical environment.⁵⁰ DDT is known to biodegrade to DDE under aerobic and to DDD under anaerobic conditions. DDE, the most prevalent breakdown product of DDT is persistent in the environment and bioaccumulates in the food chain thereby gets stored in fatty tissues of animals, fish, and humans.⁵² In the environment, much of the DDT might have been converted to p,p' -DDE. Elevated levels of p,p' -DDE have been interpreted as a result of its con-

version to p,p' -DDE by UV radiation.⁵³ The o,p' -DDT/ p,p' -DDT ratio reported to be 0.2–0.26 in DDT and ~7.5 in dicofol products.⁵⁴ The ratio of o,p' -DDT/ p,p' -DDT in the studied vegetables ranged from 0.08 to 2.95 with the mean of 0.82. This average ratio of o,p' -DDT/ p,p' -DDT lower than that of dicofol but higher than that of DDT. These observed ratios of DDT/(DDD+DDE) and o,p' -DDT/ p,p' -DDT suggest combined contamination effect from past and ongoing usage of DDT coupled with the long-range atmospheric transport tendency of DDT under tropical climate conditions but not from dicofol. DDT usage has been banned in agriculture,⁵⁵ however, India has permission for limited DDT production and use of residual spray in public health practices for vector control.⁵⁶ Concentration of total pesticides (HCHs+DDTs) in all vegetable samples

Figure 1. Comparative lifetime average daily dose (LADD), incremental lifetime cancer risk (ILCR) and hazard index (HI) for selected humans due to hexachlorocyclohexane and dichlorodiphenyltrichloroethane exposure through vegetables.

Table 3. Probable incremental life time cancer risk to humans from hexachlorocyclohexane and dichlorodiphenyltrichloroethane through vegetables.

Vegetables (no.) [*]	Risk of pesticides compounds						Total risk
	α -HCH	γ -HCH	p,p' -DDT	o,p' -DDT	p,p' -DDE	p,p' -DDD	
Carrot (12)	9.5×10^{-7} – 1.9×10^{-6} (1.4×10^{-6}) ^o	3.1×10^{-7} – 4.5×10^{-7} (3.8×10^{-7})	2.1×10^{-8} – 4.6×10^{-8} (3.2×10^{-8})	2.1×10^{-8} – 5.1×10^{-7} (2.0×10^{-7})	1.7×10^{-8} – 3.1×10^{-8} (2.4×10^{-8})	5.1×10^{-9} – 1.4×10^{-7} (4.6×10^{-8})	2.7×10^{-8} – 2.6×10^{-6} (1.2×10^{-6})
Cauliflower (13)	2.8×10^{-7} – 3.8×10^{-6} (1.7×10^{-6})	9.3×10^{-8} – 2.8×10^{-6} (9.2×10^{-7})	7.0×10^{-8} – 2.8×10^{-7} (1.6×10^{-7})	2.1×10^{-8} – 7.0×10^{-8} (4.0×10^{-8})	1.7×10^{-8} – 1.0×10^{-6} (3.3×10^{-7})	6.0×10^{-9} – 1.8×10^{-8} (1.3×10^{-8})	1.7×10^{-8} – 5.3×10^{-6} (2.9×10^{-6})
Eggplant (9)	1.6×10^{-7} – 1.9×10^{-6} (7.6×10^{-7})	3.2×10^{-8} – 1.2×10^{-6} (4.1×10^{-7})	1.9×10^{-8} – 3.8×10^{-8} (2.7×10^{-8})	1.4×10^{-8} – 5.0×10^{-8} (2.9×10^{-8})	6.6×10^{-9} – 1.0×10^{-7} (3.3×10^{-8})	4.3×10^{-9} – 9.6×10^{-9} (7.6×10^{-9})	2.8×10^{-7} – 2.2×10^{-6} (1.3×10^{-6})
Radish (7)	3.0×10^{-7} – 4.6×10^{-6} (2.9×10^{-6})	1.1×10^{-7} – 3.5×10^{-6} (1.0×10^{-6})	2.4×10^{-8} – 3.0×10^{-7} (1.4×10^{-7})	1.4×10^{-7} – 1.8×10^{-6} (6.2×10^{-7})	1.1×10^{-8} – 5.4×10^{-7} (1.7×10^{-7})	9.1×10^{-9} – 3.4×10^{-7} (1.1×10^{-7})	2.3×10^{-7} – 8.8×10^{-6} (3.9×10^{-6})
Spinach (15)	1.6×10^{-7} – 7.2×10^{-7} (5.4×10^{-7})	3.7×10^{-7} – 6.1×10^{-7} (5.2×10^{-7})	1.5×10^{-8} – 3.1×10^{-8} (2.2×10^{-8})	1.8×10^{-8} – 2.6×10^{-7} (8.3×10^{-8})	7.3×10^{-8} – 1.7×10^{-7} (1.1×10^{-7})	3.8×10^{-9} – 6.9×10^{-9} (5.2×10^{-9})	2.1×10^{-8} – 1.6×10^{-6} (1.0×10^{-6})
Tomato (8)	1.8×10^{-7} – 4.7×10^{-6} (2.0×10^{-6})	4.8×10^{-8} – 1.9×10^{-6} (5.5×10^{-7})	2.0×10^{-8} – 2.8×10^{-7} (7.5×10^{-8})	1.5×10^{-8} – 2.8×10^{-7} (1.4×10^{-7})	7.0×10^{-9} – 3.6×10^{-8} (1.9×10^{-8})	3.5×10^{-9} – 1.1×10^{-7} (3.1×10^{-8})	6.5×10^{-7} – 5.2×10^{-6} (2.8×10^{-6})
All samples (64)	1.6×10^{-7} – 4.7×10^{-6} (1.7×10^{-6})	3.2×10^{-8} – 3.5×10^{-6} (7.3×10^{-7})	1.5×10^{-8} – 3.0×10^{-7} (9.8×10^{-8})	1.4×10^{-8} – 1.8×10^{-6} (1.7×10^{-7})	6.6×10^{-9} – 1.0×10^{-6} (1.8×10^{-7})	3.5×10^{-9} – 3.4×10^{-7} (3.5×10^{-8})	1.7×10^{-8} – 8.8×10^{-6} (2.5×10^{-6})

*Number of samples; ^oaverage in parenthesis. HCH, hexachlorocyclohexane; DDT, dichlorodiphenyltrichloroethane; DDE, dichlorodiphenyldichloroethylene; DDD, dichlorodiphenyldichloroethane.

Table 4. Non-carcinogenic health hazard and hazard index for humans from hexachlorocyclohexane and dichlorodiphenyl-trichloroethane through vegetables.

Vegetables (no.)*	α -HCH	γ -HCH	Non-cancer HQs of pesticide compounds p,p' -DDT	o,p' -DDT	p,p' -DDE	p,p' -DDD	HI
Carrot (12)	1.9×10^{-5} - 3.8×10^{-5} (2.9×10^{-5})	9.5×10^{-4} - 1.4×10^{-3} (1.1×10^{-3})	6.5×10^{-4} - 1.4×10^{-3} (9.6×10^{-4})	1.8×10^{-7} - 4.5×10^{-6} (1.7×10^{-6})	- [#]	- [#]	6.6×10^{-4} - 2.8×10^{-3} (1.5×10^{-3})
Cauliflower (13)	5.5×10^{-6} - 7.4×10^{-5} (3.4×10^{-5})	2.8×10^{-4} - 8.6×10^{-3} (2.8×10^{-3})	2.1×10^{-3} - 8.4×10^{-3} (4.8×10^{-3})	1.8×10^{-7} - 6.1×10^{-7} (3.5×10^{-7})	-	-	1.7×10^{-3} - 1.7×10^{-2} (7.2×10^{-3})
Eggplant (9)	3.3×10^{-6} - 3.7×10^{-5} (1.5×10^{-5})	9.8×10^{-5} - 3.6×10^{-3} (1.2×10^{-3})	5.7×10^{-4} - 1.1×10^{-3} (8.3×10^{-4})	1.2×10^{-7} - 4.4×10^{-7} (2.5×10^{-7})	-	-	7.1×10^{-4} - 4.2×10^{-3} (2.1×10^{-3})
Radish (7)	6.0×10^{-6} - 9.2×10^{-5} (5.7×10^{-5})	3.3×10^{-4} - 1.1×10^{-2} (3.0×10^{-3})	7.3×10^{-4} - 9.0×10^{-3} (4.2×10^{-3})	1.2×10^{-6} - 1.6×10^{-5} (5.3×10^{-6})	-	-	8.9×10^{-5} - 1.1×10^{-2} (5.2×10^{-3})
Spinach (15)	3.2×10^{-6} - 1.4×10^{-5} (1.1×10^{-5})	1.1×10^{-3} - 1.8×10^{-3} (1.6×10^{-3})	4.6×10^{-4} - 9.4×10^{-4} (4.0×10^{-4})	1.5×10^{-7} - 2.3×10^{-6} (7.2×10^{-7})	-	-	1.5×10^{-7} - 2.4×10^{-3} (1.7×10^{-3})
Tomato (8)	3.6×10^{-6} - 9.4×10^{-5} (4.0×10^{-5})	1.4×10^{-4} - 5.9×10^{-3} (1.7×10^{-3})	6.1×10^{-4} - 8.5×10^{-3} (2.3×10^{-3})	1.3×10^{-7} - 2.4×10^{-6} (1.2×10^{-6})	-	-	1.1×10^{-3} - 8.7×10^{-3} (4.0×10^{-3})
All samples (64)	3.2×10^{-6} - 9.4×10^{-5} (3.4×10^{-5}) ^o	9.8×10^{-5} - 1.1×10^{-2} (2.2×10^{-3})	4.6×10^{-4} - 9.0×10^{-3} (3.0×10^{-3})	1.2×10^{-7} - 1.6×10^{-5} (1.4×10^{-6})	-	-	1.5×10^{-7} - 1.7×10^{-2} (4.5×10^{-3})

*Number of samples; ^oaverage in parenthesis; [#]not calculated-no reference dose assigned. HQ, health hazard; HI, hazard index; HCH, hexachlorocyclohexane; DDT, dichlorodiphenyltrichloroethane; DDE, dichlorodiphenyldichloroethylene; DDD, dichlorodiphenyldichloroethane.

was ranged from 0.04 $\mu\text{g kg}^{-1}$ to 5.69 $\mu\text{g kg}^{-1}$ with the mean value of 4.18 $\mu\text{g kg}^{-1}$. The observed levels of HCH and DDT in vegetables were compared with maximum residual levels (MRLs) and found lower than the recommended values from European countries and Indian government.^{57,58}

Human health risk

The overall LADD for all studied vegetables was ranges between 5.0×10^{-8} – 9.1×10^{-6} $\text{mg kg}^{-1} \text{d}^{-1}$ with an average value of 2.0×10^{-6} $\text{mg kg}^{-1} \text{d}^{-1}$. These estimated average daily intakes were within recommended acceptable limits of reference dose (RfD) for HCHs (α -HCH, 8×10^{-3} $\text{mg kg}^{-1} \text{d}^{-1}$; γ -HCH, 3×10^{-3} $\text{mg kg}^{-1} \text{d}^{-1}$) and DDTs (o,p' -DDT, 5×10^{-4} $\text{mg kg}^{-1} \text{d}^{-1}$; p,p' -DDT, 9.7×10^{-5} $\text{mg kg}^{-1} \text{d}^{-1}$).^{43,59} ILCR for all the samples was ranges between 1.7×10^{-8} – 8.8×10^{-6} with an average of 2.5×10^{-6} . This estimated ILCR was within the acceptable excess risk distribution range (10^{-6} – 10^{-4}).⁴³ Acceptable limits of excess cancer risks recommended by United States Environmental Protection Agency are ranges between 1 chance in 1,000,000 (1×10^{-6}) to 100 chance in 1,000,000 (1×10^{-4}). Non-carcinogenic health effects of HCH and DDT was estimated as HQ. HQ is the exact measure of the magnitude of exposure potential or a quantifiable potential for developing non-carcinogenic health effects after averaged exposure period. If the HQ for a chemical is equal to or less than one (1), it is believed that there is no appreciable risk that non-cancer health effects will occur. The potential for non-cancer effects was evaluated by comparing the estimated average daily dose ($\text{mg kg}^{-1} \text{d}^{-1}$) of the pesticide compounds with the RfD ($\text{mg kg}^{-1} \text{d}^{-1}$) for that compound. Total non-cancer risk

was derived simply by summing the HQ values for individual vegetables. This total HQ is referred to as HI. If the HI value is less than 1 (one), non-cancer risks are not expected from any chemical, alone or in combination with others. The quantified health HQ for total pesticides (HCH, DDT isomers) through vegetables for human adults has been given in Table 4. The average HI was 1.5×10^{-3} , 7.3×10^{-3} , 2.1×10^{-3} , 5.2×10^{-3} , 1.7×10^{-3} and 4.0×10^{-3} , for carrot, cauliflower, eggplant, radish, spinach and tomato, respectively. These estimated HI values were much lower than the acceptable safe risk level ($\text{HI} \leq 1$), indicating negligible risk.

Conclusions

Concentrations of analysed pesticide compounds in studied vegetables were found within the recommended MRLs. The estimated average daily intakes of HCH and DDT compounds through vegetables were lower than RfD. Subsequently, health risk to humans in terms of HQs and HI was much less than one, suggesting negligible risk. There were numbers of limitations involved in method of risk estimation which may lead to uncertainties in total risk, such as consideration of different age groups. Because, infants and childhood age group may have a greater probability of producing risk than exposure in adulthood. Body weight differences and possible synergistic effects of other toxic chemicals could also lead to uncertainties. Therefore, more in-depth studies are recommended, which may also including other persistent organic pollutants and toxic metals.

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