

HEALTH RISK FOR HUMANS DUE TO HEXACHLOROCYCLOHEXANE (HCH) AND DICHLORODIPHENYLTRICHLOROETHANE (DDT) IN URBAN SOILS FROM VARIOUS CITIES IN INDIA

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INTRODUCTION

Hexachlorocyclohexane (HCH) and Dichlorodiphenyltrichloroethane (DDT) have been listed by the Stockholm Convention as persistent organic pollutants (POPs). These are much of concern due to their persistence, bioaccumulation, toxicity, and potential to cause health impacts on wide variety of plant and animal species, including humans¹⁻². Their health impacts on human beings included endocrine-disrupting and carcinogenic to cause cancer of various organs³. Due to their characteristics properties of hydrophobicity, strong sorption to soil organic matter, resistance to degradation and biodegradation, DDT and HCH may persist for longer period in soils⁴. Humans may be exposed to toxic contaminants in soils through ingestion, inhalation or dermal contact due to soils have close proximity to humans. Soil ingestion could be a major pathway of exposure in health risk toxic/persistent organic pollutants⁵.

This study presents the distribution and health risk of DDT and HCH compounds in urban soils from various cities in India. Lifetime average daily dose (LADD) of HCH and DDT for human adults and children through soil ingestion and their potential incremental lifetime cancer risk (ILCR) have been discussed.

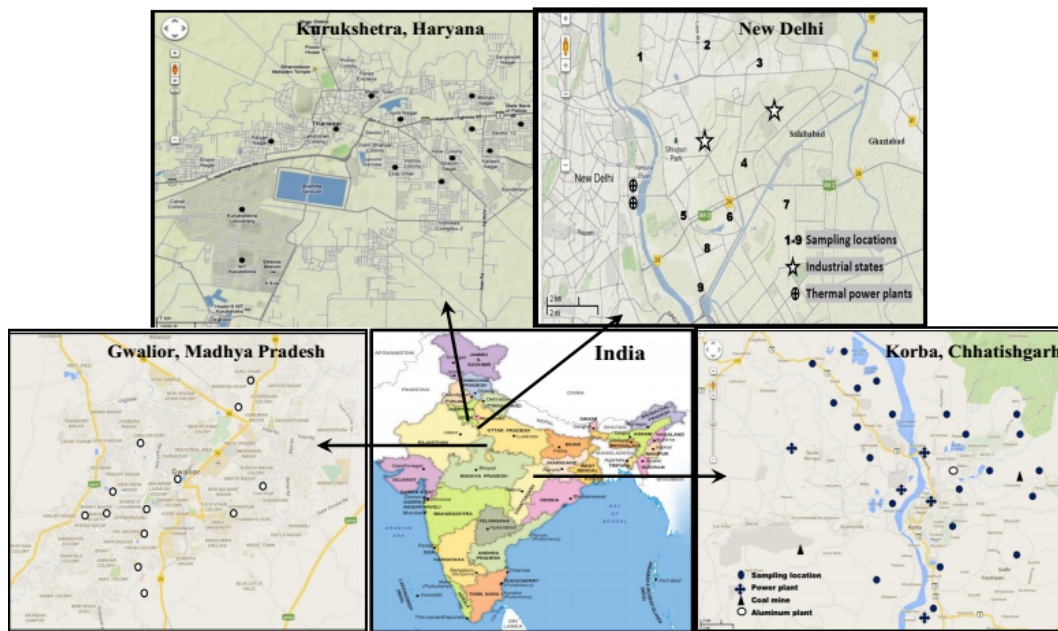


Fig.1. Map showing sampling locations in different cities in India

MATERIALS AND METHODS

Study Area and Sampling

Study areas with sampling locations were presented in Fig. 1. Study areas located in North India (Kurukshetra & New Delhi), Central India (Gwalior) and Eastern India (Chhatishgarh). A total number of 167 soil samples collected from Kurukshetra, Haryana (KUK, 39 Nos), New Delhi (ND, 36 Nos), Gwalior, Madhya Pradesh (GL, 48 Nos) and Korba, Chhatishgarh (KB, 44 Nos). For each sampling location, three sub-samples of approximately

500 grams of soil in the radius of 5-10m were taken from the same location excluding vegetation area (at the depth of 0–10 cm). After collection, pebbles, plant leaves and wood sticks were removed manually and the samples collected at the 3 points of each location are combined and mixed thoroughly to ensure a representative sample of that location. Then about 500 grams of soil was taken into cleaned wide mouth amber glass Teflon lined screw cap bottles. All collected samples were transported to the laboratory. Samples were air-dried in dark clean space at room temperature. Air dried samples were sieved through a 1-mm mesh screen and stored in glass bottles in refrigerator at $\sim -4^{\circ}\text{C}$ until extraction and analysis.

Extraction and cleanup

Extraction and cleanup of samples was carried out following validated methods from USEPA. Soil samples (~ 20 g) were extracted with mixture of acetone-hexane (1:1 v/v) in ultrasonic water bath. Extracted samples were filtered and process was repeated twice. The pooled extracts were concentrated under reduced pressure using a rotary evaporator (Eyela, Tokyo, Japan). The multilayered silica gel column chromatography was performed for cleanup and to remove interfering compounds. Hexane was used for elution of the analytes and concentrated under gentle stream of pure nitrogen using Rotary evaporator and Turbo Vap (Caliper, USA) to 1.0 ml for analysis.

Instrumental Quantification and Analytical quality control

Analysis of organochlorine pesticides (OCPs) was carried out using gas chromatograph (Perkin Elmer, Clarus 500) equipped with an electron capture detector (ECD, ^{63}Ni). Pesticide compounds were separated on Elite-1 analytical column (25 m x 0.20 mm, 0.33 μm particle of 5% diphenylpolysiloxane and 95% dimethylpolysiloxane). The column temperature was initially maintained at 170°C and increased to 220°C ($@ 7^{\circ}\text{C min}^{-1}$), temperature was ramped to 250°C ($@ 5^{\circ}\text{C min}^{-1}$) and held for 6.86 min. The injector and detector temperature was 250°C and 350°C , respectively. Purified laboratory grade nitrogen gas was used as carrier ($@ 1.0$ ml min^{-1}).

Instrument calibration and other quality control studies were undertaken using certified reference standard solutions from Supelco (Sigma-Aldrich, USA). Individual peaks in sample extracts were identified with the accurate retention time of corresponding standard. Concentration was determined with external standard method comparing peak area in samples with the standards using five level calibration curves (r^2 value, 0.999). Quality control analyses includes procedural blanks to check for cross contamination, random duplicate samples ($\text{sd} < 5$), calibration verification ($\text{sd} < 5$), and matrix spiked recovery (85-105% ± 5 -9%). Average of duplicate analysis was used in calculations. Concentrations below reporting limits ($0.01 \mu\text{g kg}^{-1}$) were considered as zero in calculations. Moisture content was determined and results were reported as $\mu\text{g kg}^{-1}$ dry weight.

Health Risk Assessment

Health risk assessment was based on assumption that humans may be exposed to pesticides contaminated soil through ingestion, inhalation and dermal contact. In this study, human exposure to HCHs and DDTs through soil ingestion pathway was considered for health risk. In this study, incremental lifetime cancer risk (ILCR) to human adult and children was assessed by calculating the lifetime average daily dose (LADD) of HCHs and DDTs through soils ingestion⁶. The following equations were used for estimating the LADD and ILCR.

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

$$\text{Incremental Life time Cancer Risk (ILCR)} = \text{LADD} \times \text{CSF} \quad [2]$$

Where, Cs is the pollutant concentration in soil (mg kg^{-1}), IR is the soil ingestion rate (100 mg d^{-1} for adult and 200 mg d^{-1} for children), F is the unit conversion factor, EF is exposure frequency (365 days/year), ED is the life time exposure duration (adults, 70 years; children, 12 years), BW is the body weight (adults, 70 kg; children, 27 kg), and AT is the averaging time for carcinogens ($\text{EF} \times \text{ED}$). CSF is oral cancer slope factor for a particular compound⁷.

RESULTS AND DISCUSSIONS

Concentrations of HCHs and DDTs in Soil

Concentrations of total and individual HCH and DDT isomers in soils from different cities in India are presented in Fig. 1 & Fig.2. The average concentrations ΣOCPs ($\Sigma\text{HCH} + \Sigma\text{DDT}$) was $1.79 \pm 1.14 \mu\text{g kg}^{-1}$, $4.03 \pm 3.49 \mu\text{g kg}^{-1}$, $9.86 \pm 6.51 \mu\text{g kg}^{-1}$ and $26.29 \pm 37.01 \mu\text{g kg}^{-1}$, respectively for KUK, ND, GL and KB soil (Fig.3). Compositional profiles of HCH and DDT isomers in the environment are used to identify the contamination sources. The changes in composition may be due to metabolic degradation of original components of pesticide. The ratio of α -HCH to

γ -HCH has been widely used to identify the possible sources of HCH in the environment. It has been reported that ratio between 3 and 7 (α/γ -HCH) indicates fresh input of technical HCH⁸, while lower ratio of ≤ 1 , indicates lindane applications⁹. The estimated pooled ratio of α/γ -HCH in this study was 1.04 ± 0.56 , 0.97 ± 0.42 , 1.34 ± 0.55 and 1.99 ± 1.07 , respectively for KUK, ND, GL and KB. These pooled mean values reflect the dominant usage of lindane. The ratio of o,p' -DDT/ p,p' -DDT are used to distinguished, contamination from technical DDT (~ 7.5) or dicofol ($0.2\text{--}0.26$)¹⁰. The observed ratio of o,p' -DDT/ p,p' -DDT in KUK, ND, GL and KB soils was 1.92 ± 0.71 , 1.72 ± 0.92 , 0.36 ± 0.31 and 0.45 ± 0.17 , respectively. These ratios of o,p' -DDT/ p,p' -DDT suggest combined contamination from past and ongoing usage of DDT coupled with the long-range atmospheric transport (LART) tendency of DDT under tropical climatic conditions but not from dicofol.

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 for vector control under WHO guidelines¹¹⁻¹².

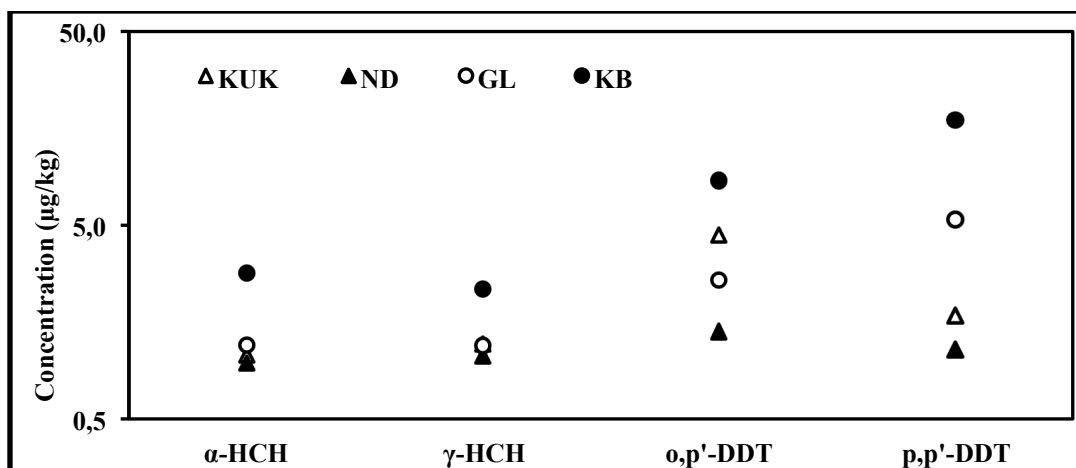


Fig.2. Comparative average concentration of OCPs in soil from different cities in India

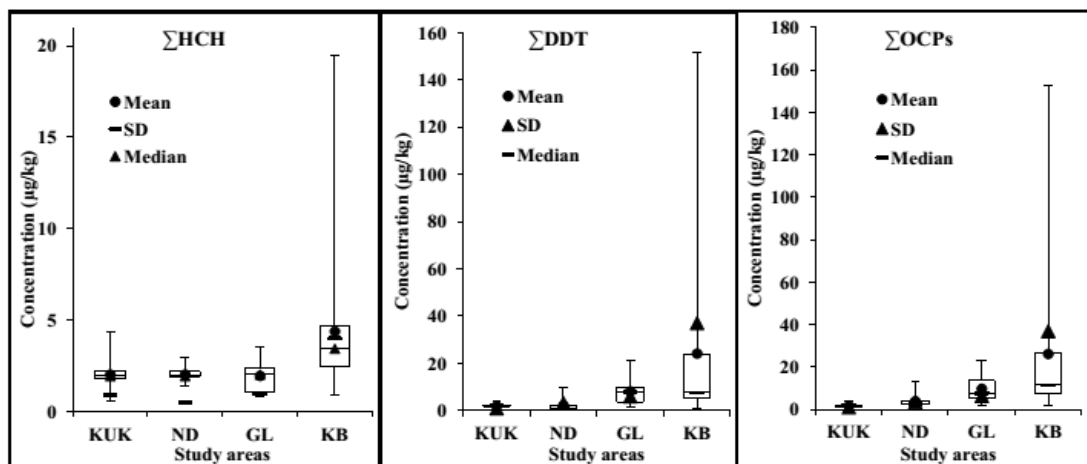


Fig. 2.Box and Whisker Plot of Σ HCH, Σ DDT and Σ OCPs in soil from different cities in India

Human Health Risk of HCHs and DDTs

Estimated average LADD and ILCR of Σ HCH and Σ DDT for human adults and children is summarized in Table 1. The estimated average LADD of Σ OCPs (Σ HCH+ Σ DDT) through ingestion of soil for the humans residing in Eastern India was comparatively more than Central India and North India. The average ILCR due to Σ OCPs (Σ HCH+ Σ DDT) in soil was 1.2×10^{-8} , 1.1×10^{-8} , 1.6×10^{-8} , and 3.9×10^{-8} for human adults residing in KUK, ND, GL

and KB, respectively. However, for children, it was 6.2×10^{-8} , 1.1×10^{-7} , 5.6×10^{-8} and 2.0×10^{-7} , respectively. These ILCR for adults and children through ingestion was within acceptable limit of 10^{-6} to 10^{-4} as acknowledged by regulatory agencies⁶. The acceptable risk distribution is constraints based percentiles and must be equal or lower than 10^{-6} to 10^{-4} using upper bound factors⁷.

For lack of data about HCHs and DDTs (organochlorine pesticides) diet exposure, we cannot give precise total daily pesticides intake. However, according to our results from ingestion of soil, we can conclude that the daily dose of HCHs and DDTs, cancer risk and non-cancer risk to human adults and children residing in Korba city of Chhattisgarh, India is lower based on the proposed guidelines⁷.

Table1. LADD ($\text{mg kg}^{-1} \text{ day}^{-1}$) & ILCR for humans due to OCPs in soil from different cities in India

Study area	Humans	Σ HCH		Σ DDT		Σ OCPs	
		LADD	ILCR	LADD	ILCR	LADD	ILCR
KUK	Adults	2.9×10^{-9}	1.1×10^{-8}	5.5×10^{-9}	1.9×10^{-9}	7.2×10^{-9}	1.2×10^{-8}
	Children	1.5×10^{-8}	5.5×10^{-8}	2.9×10^{-8}	9.8×10^{-9}	3.7×10^{-8}	6.2×10^{-8}
ND	Adults	2.8×10^{-9}	9.6×10^{-9}	3.2×10^{-9}	1.1×10^{-9}	5.6×10^{-9}	1.1×10^{-8}
	Children	1.5×10^{-8}	5.4×10^{-8}	1.7×10^{-8}	5.7×10^{-9}	3.0×10^{-8}	1.1×10^{-7}
GL	Adults	3.3×10^{-9}	1.4×10^{-8}	1.4×10^{-8}	4.7×10^{-9}	1.6×10^{-8}	1.6×10^{-8}
	Children	1.1×10^{-8}	4.7×10^{-8}	4.7×10^{-8}	1.6×10^{-8}	5.6×10^{-8}	5.6×10^{-8}
KB	Adults	6.3×10^{-9}	2.8×10^{-9}	3.5×10^{-8}	1.2×10^{-8}	4.4×10^{-8}	3.9×10^{-8}
	Children	3.2×10^{-8}	1.4×10^{-7}	1.8×10^{-7}	6.1×10^{-8}	2.0×10^{-7}	2.0×10^{-7}

CONCLUSION

Cancer risk (ILCR) due to HCH and DDT in soil from various cities was lower than acceptable guideline values, suggested low risk for human adults and children in this area of study.

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REFERENCES

- ATSDR. 2005. Toxicological profile for hexachlorocyclohexanes, ATSDR, US Public Health Service, Atlanta, GA.
- ATSDR (Agency for Toxic Substances and Disease Registry). 2008. Toxicological profile for DDT, DDE, and DDD, Agency for Toxic Substances and Disease Registry, US Public Health Service, Atlanta, GA.
- Ejaz S, Akram W, Lim CW, *et al.* 2004. *Experimental Oncology*, 26(2): 98-105.
- Ferencz L and Balog A. 2010. *Carpathian J Earth Environ Sci*, 5(1):111-118.
- Abrahams PW. 2002. *Science of the Total Environment*, 291:1-32.
- USEPA. 1989. Risk Assessment Guidance for Superfund. Human Health Evaluation Manual (Part A). EPA 540-1-89-002. Washington, DC.
- USEPA. Accessed January, 2015 at <http://www.epa.gov/reg3hwmd/risk/human>.
- Yang Y, Li D and Mu D. 2008. *Atm Environ*, 42: 677-687.
- Willett L, Ulrich EM and Hites HA. 1998. *Environ Sci Technol*, 32(15): 2197-207.
- Talekar NS, Sun LT, Lee EM, Chen JS. 1977. *J Agric Food Chem*, 25: 348-352.
- Jit S, Dadhwal M, Kumari H, *et al.* 2011. *Environ Sci Pollut Res*, 18: 586-597.
- UNEP. 2011. <http://chm.pops.int/>.