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Occurrence of DDT in foodstuffs and skin wipes from a rural area, South China: insight into human exposure pathway

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Abstract

Dichlorodiphenyltrichloroethanes (DDTs) are ubiquitous in dust and air, which may be responsible for human dermal exposure to DDT. However, existing DDT exposure studies mainly concentrate on dietary exposure, and studies on skin exposure pathway are lacking. To gain further insight into the human exposure pathway of DDT, skin wipe and food samples were collected in a rural area in southern China, where new input of DDTs was found in both indoor and outdoor environments. The total DDT concentrations in skin wipes and foodstuffs varied from < method quantification limit (MQL) to 1470 ng/m^2 and from < MQL to 12.8 ng/g wet weight, respectively. In foodstuffs, egg showed the highest DDT concentration, while forehead showed the highest DDT concentration in the four skin locations. *p,p'*-DDT was predominant in the hand and forearm wipes, while *p,p'*-DDE was dominant in the forehead, upper arm, and food samples. The total DDT daily absorption median levels via dermal contact, hand-mouth contact, and food consumption were 0.187, 0.0237, and 0.994 ng/kg/d, respectively. Organic eggs and wild fish contaminated by DDTs released locally are the main sources of human dietary exposure to DDTs, and reducing the intake of contaminated fish and eggs would help to significantly reduce human DDT absorption; in this

case, the contribution of dermal exposure cannot be not negligible.

Keywords: Dichlorodiphenyltrichloroethanes (DDTs), dermal exposure, dietary ingestion, foodstuffs, skin wipes

INTRODUCTION

As one of the first group of chemicals controlled by the Stockholm Convention, dichlorodiphenyltrichloroethane (DDT) is known for its harmful effects on the environment and human. The adverse health implications that have been reported associated with short and long-term exposure to DDTs include skin rash, vomiting, convulsions, respiratory problems, infertility, cancers, hepatotoxicity, birth defects, neurotoxicity, Parkinson's disease, and so forth[1-6]. DDT was widely used as a pesticide in agricultural production in China since the 1950s and was banned in the 1980s, with a historical production of 400,000 tons[7]. After its ban, it was still added to the antifouling paint on the surfaces of fishing ships until 2009 and used in the production of dicofol until 2019[7]. Although the production and application of DDT in agricultural and non-agricultural fields have been banned successively, its residues are still prevalent in air, dust, soil, food, human body, *etc.*[7-11]. As such, humans are exposed to DDT and its transformation products, dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD), and it is crucial to quantify human exposure to these residues and figure out how they enter the human body.

Current knowledge about human exposure to DDTs has largely focused on dietary ingestion, and food products contaminated with DDTs are widely believed to be the main source of human exposure to these compounds[10,12-14]. In a previous study, we found that wooden furniture paint was a new source of DDTs in indoor dust in residences from a rural area in South China[15]. This raised question of whether dermal absorption plays an important DDT exposure pathway for residents. Recently, dermal absorption has been recognized by a growing number of researchers as a significant pathway for human exposure to semi-volatile organic compounds (SVOCs)[16-19]. Several studies have evaluated the skin absorption of DDT through dust[10,20,21]. However, in addition to dust, all kinds of product surfaces, gaseous and particulate phases have also been reported as potential sources of skin contact with SVOCs[22-24]. Thus, just considering skin absorption from dust may greatly influence the accuracy of the skin exposure assessment. Skin wipe method has been recommended as a more suitable alternative for assessing skin exposure, which can be accomplished by direct determination of contaminant levels on the skin surface[25-27]. This method has now been applied in the assessment of human skin exposure to a variety of compounds, including polychlorinated biphenyls, polycyclic aromatic hydrocarbons, brominated and organophosphate flame retardants, *etc.*[16,17,28,29], but there seems to be no report of DDTs using this method. Hence, it is necessary to assess skin exposure to DDTs using wipe method to gain a deeper understanding of the significance of DDTs' skin absorption pathway.

In this study, food samples and human skin wipe samples from four body sites (forehead, hand, forearm, and upper arm) were collected from rural areas in southern China, and the DDT residues in these samples were determined. Then the measured DDT concentrations were used to estimate human skin absorption, hand-to-mouth contact ingestion, and dietary exposure. Finally, to gain a better understanding of the human exposure pathway and source of DDTs, comparisons of uptake estimates through dermal contact and diet ingestion with those through the other pathways (inhalation and dust ingestion) were conducted. The current study aims to provide a full-scale assessment of human exposure to DDTs.

MATERIALS AND METHODS

Study area and sample collection

The rural areas in Qingyuan, China were selected as the sampling areas in this study. One study on the halogenated organic compounds in insects and plants indicated that new inputs of DDTs still exist in the study area[30], and our previous study[15] found that wooden furniture paint was an important indoor emission source of DDT in this region. Therefore, the area was selected to study the comprehensive exposure of DDT to the local villagers. A total of 178 food samples and 120 skin wipe samples were collected from January to August 2020 in rural areas of Qingyuan, China. Specifically, 98 hen eggs, 46 fish, and 8 vegetable categories were obtained from the local farmhouses/market, local river, and local farmland, respectively. Raw chicken (n = 6) and pork (n = 10) samples were purchased from the produce market. Roast goose/duck (n = 9) was bought from the deli. Considering subsequent labor intensity, 98 eggs were combined into 11 mixed samples and 46 fish were combined into 17 mixed samples. Thirty villagers were recruited from the study area, each giving verbal informed consent before providing personal information (body weight, height, age, gender, *etc.*) and skin wipe samples. The four skin areas (hand, forehead, forearm, and upper arm) of each person were wiped with a sterile gauze pad (7.5 cm × 7.5 cm) pre-soaked with isopropanol. Every participant was asked not to clean these skin areas for at least 2 h before sample collection. This study was ethically approved by the Research Ethics Committee of Guangzhou Institute of Geochemistry prior to proceeding (GIG-2019-001).

Sample analysis

Skin wipe sample

Each skin wipe sample was spiked with 100 ng of internal standard (PCB 82) and mixed with 20 mL of hexane/acetone (3:1, *v/v*) and then ultrasonicated for 10 min. This ultrasound extraction process was repeated in triplicate. The extracts from the three extractions were collected together and evaporated to approximately dry with a rotary evaporator and/or a nitrogen concentration system, and then solvent-exchanged into approximately 1 mL of hexane. Afterward, Silica solid phase cartridges (CNWBOND Si SPE, 6 mL, 2-g bed weight, ANPEL Laboratory Technologies) were preconditioned with 8 mL of ethyl acetate and 6 mL of hexane and then used to purify the resulting extract. The target fraction was eluted using 20 mL of hexane/dichloromethane (1:1, *v/v*), evaporated to near drying, and finally redissolved in 100 μL of iso-octane. PCB 65 was added as a recovery standard prior to injection.

Food sample

Food samples (2 g dry weight each) were weighted, spiked with PCB 82 standard solution, and then extracted with 7 mL of acetonitrile (containing 10% toluene) in an ultrasonic bath three times. Subsequent clean-up of the combined extracts was performed through Silica solid-phase cartridges following the steps described above. The resulting eluate was further purified with 200 mg of dispersive SPE (graphitized non-porous carbon sorbent powders) to remove the pigments in vegetable and egg matrix, and/or treated with concentrated sulfuric acid (H_2SO_4) to remove the lipids in food of animal origin. The final sample extract was dissolved in iso-octane and PCB 65 standard solution for GC-MS (gas chromatography-mass spectrometry) analysis. Details of the DDT instrument analysis are described in our previous literature[15] and in the Supporting Information [Supplementary Text 1].

Quality assurance and quality control (QA/QC)

Field blank and procedural blank were incorporated into our QA/QC system to monitor potential contamination and make appropriate corrections. The DDT amounts measured in the blank samples were subtracted from the obtained values of all the samples. Spiking 10 ng DDT standards into soaked sterile gauze pads (n = 3) and solvent (n = 3), and the resulting spiked recoveries of DDTs were 90%-112% for the spiked matrices and 86%-105% for spiked blank. The recovery of internal standard (PCB 82) was

97.5% ± 19%, and the reported DDT concentrations were not corrected according to this value. The inlet breakdown of DDT was less than 15% in the spiking studies. The method quantification limits (MQLs) were calculated as the average concentration of procedural blanks plus triple standard deviations. For the congeners that were not detected in blanks, MQLs were assigned as a value that is 10 times signal-to-noise ratios, using the lowest standard level. The MQLs of DDTs varied from 0.01 to 0.08 ng/wipe for skin wipes and from 0.0036 to 0.1064 ng/g dry weight for food samples.

Data statistics and analysis

To facilitate subsequent exposure assessment, all of the food concentration values are expressed as the wet weight (ww) concentrations in this study. The concentration values below the MQLs were replaced with half of the MQLs in the statistical analyses and with the value zero in the exposure assessment. And only compounds with detection frequencies higher than 50% were used for analysis for statistical purposes in this study. The relationships between the DDT concentrations in the four skin locations were analyzed using Spearman's rank correlation test. The statistical significance level was set at $P = 0.05$.

The daily absorption dose (DAD) of DDTs through dermal contact, hand-to-mouth contact and dietary ingestion was assessed using the concentration data of skin wipes and food items. The permeability coefficient model[28] was used to calculate the dermal absorption dose (equation 1) in this study. Hand-to-mouth contact exposure and food exposure were estimated from equation 2[28] and equation 3[10], respectively. Details of the equation and parameter are as follows:

$$DAD_{dermal} = \frac{(C_{head} \times A_{head} + C_{hand} \times A_{hand} + C_{ob} \times A_{ob} + C_{cc} \times A_{cc}) \times K_{p-l} \times ED}{l_m \times BW} \quad (1)$$

where C_{head}, C_{hand}, C_{ob}, and C_{cc} (ng/m^2) are the area-based DDT concentrations on the head, the hand, the other bare skin, and the clothing-covered skin surfaces, respectively, we assumed that the dermal absorption levels of DDT in the other bare skin and the clothing-covered skin areas are same as those of the forearm and upper arm, respectively; A_{head}, A_{hand}, A_{ob}, and A_{cc} are the skin surface area of the head, hand, other bare skin surface (forearm + shank + foot), and clothing-covered skin surface (whole body areas-others), respectively[31]; K_{p-l} is the permeability coefficient of chemicals from skin lipids into dermal capillaries (cm/h) [Supplementary Table 1]; l_m is the thickness of skin lipid, which was assumed to be 1.3 μm[32]; ED is the exposure duration (24 h/day); BW is the body weight, and the average weight of all the participants (57 kg) was used for exposure assessment in this study.

$$DAD_{hand-mouth\ contact} = \frac{C_{hand} \times TE \times F \times SAC \times A_{hand}}{BW} \quad (2)$$

where TE is the transfer efficiency (50%, mass fraction of the chemical transferred per contact)[33]; F is the frequency of hand-mouth contact (24 /day) ; SAC is the fraction of hand area per contact (10%)[16].

$$DAD_{inge-food} = \frac{C_{food} \times DFC \times E_{ing}}{BW} \quad (3)$$

where C_{food} (ng/g wet weight) is the DDT concentrations in food samples; DFC (g/day) is daily food consumption rates (20.5, 56.8, 51.2, 85.1, 83.5, 284 g/day for egg, fish, chicken, pork, goose/duck, and vegetable, respectively), the values were obtained from the Statistical Yearbook of Guangdong Province, China; E_{ing} is the uptake efficiency of DDTs in food via human intestine, the value (~90%) was obtained from previous literature[34].

RESULTS AND DISCUSSION

Concentrations and profiles of DDT in skin wipes

Descriptive statistics for the concentrations of DDTs (including *o,p'*-DDT, *p,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDE, and *p,p'*-DDE) in skin wipes are summarized in Table 1. Among the six target DDTs, the five isomers, including *o,p'*-DDT, *p,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, and *p,p'*-DDE, were detected in skin wipes, and *o,p'*-DDE was not detected in any of the skin samples. The total DDT concentrations (sum of all detected chemicals) varied from < MQL to 1,470 ng/m^2 in skin samples, with a median of 19.8 ng/m^2. There seems to be no other study reporting DDT occurrence in human skin wipes, so it is hard to make a comparison of the DDT levels between our study and other literature.

The total DDT concentrations from forehead (median 102 ng/m^2) were remarkably higher than those from hand (20.4 ng/m^2), forearm (13.8 ng/m^2), and upper arm (12.0 ng/m^2) ($P < 0.05$), while no statistical difference was observed among levels from the latter three skin sites. This could be owing to the more sebum on forehead than the other three skin sites, the abundant oils secreted by sebum tend to favor the adsorption of DDTs[35,36]. Similar skin site distributions were also found for organophosphate flame retardants[22] and polycyclic aromatic hydrocarbons[28]. Spearman correlation analysis showed there were significant positive associations for the DDT concentrations among the four skin locations (r = 0.654~0.883, $P < 0.05$), revealing the probably similar exposure sources of DDT among these skin sites.

Regardless of detection frequency and concentration, *p,p'*-DDE and *p,p'*-DDT exhibited their predominance in skin wipe samples. Specifically, *p,p'*-DDE was detected in 96%-100% of skin wipe samples. The detection frequencies of *p,p'*-DDT were 50% in the forehead skin samples and were up to 83%-88% in the other three skin locations (hand, forearm, and upper arm). *p,p'*-DDE and *p,p'*-DDT were the major isomers in 53.3% and 43.9% of the skin samples with levels of 34.7 ± 67.2 and 57.1 ± 158 ng/m^2, respectively.

Dust and air are two important sources of DDT accumulated on the surface of human skin[37]. DDTs in air and dust samples inside and outside the residences of the same population as in the current study were reported in our previous study[15]. The total concentrations of *p,p'*-DDE and *p,p'*-DDT accounted for 50.3-89.3% of Σ_5DDTs in air and 63.2%-100% in dust samples. This result is in good agreement with the predominance of these two isomers in the skin wipes found in this study.

To further elucidate the possible sources and routes of dermal exposure, a detailed comparison of DDT composition between the skin wipes and the environment matrices was performed [Figure 1]. The proportion of *p,p'*-DDE on the forehead and upper arm was higher than that of *p,p'*-DDT, consistent with the profile in PUF samples[15], while the fraction of *p,p'*-DDE on the hand and forearm was lower than that of *p,p'*-DDT, in line with the profile in dust samples, which suggested that air-derived DDTs are very likely a major contributor to the DDTs attached with the skin surface of forehead and upper arm, and dust-derived DDTs could contribute substantially to the amounts of DDT enriched on the hand and forearm surface. Air-to-skin transport is generally the primary pathway of dermal exposure on forehead, so it can be speculated that the same compound should have similar proportions between the forehead and air samples. However, the percentage of *p,p'*-DDE on forehead (71%) far exceeded that in PUF samples (~45%), and *p,p'*-DDT's percentage on forehead (13%) was about half that of the PUF samples (~25%). This observation implies that there may be more *p,p'*-DDE being transmitted from air to the surface of forehead than *p,p'*-DDT, or/and *p,p'*-DDT accumulated on forehead could degrade into *p,p'*-DDE. Both the percentages of *p,p'*-DDE and *p,p'*-DDT were slightly higher on upper arm (51% and 34%, respectively) than in air (about 45% and 25%, respectively), indicating that besides the air-(to-clothing)-to skin transport route, dust adhesion also contribute to the DDT amounts on upper arm to some extent. DDTs on hand and forearm were

Table 1. Concentrations of DDT residue in skin wipe samples (ng/m^2)

Compound	Forehead		Hand		Forearm		Upper Arm	
	Df (%)[a]	Median (range)	Df (%)	Median (range)	Df (%)	Median (range)	Df (%)	Median (range)
p,p'-DDE	96	40.0 (< MQL-392)	97	6.33 (< MQL[b]-226)	100	4.54 (0.95-180)	100	5.05 (0.64-181)
p,p'-DDT	50	7.58 (< MQL-733)	83	7.45 (< MQL-828)	87	5.01 (< MQL-871)	88	3.33 (< MQL-823)
o,p'-DDT	13	< MQL (< MQL-58.6)	30	< MQL (< MQL-51.8)	37	< MQL (< MQL-57.3)	33	< MQL (< MQL-52.8)
p,p'-DDD	58	8.63 (< MQL-94.7)	70	2.15 (< MQL-333)	70	1.63 (< MQL-109)	58	0.83 (< MQL-133)
o,p'-DDD	25	< MQL (< MQL-13.6)	70	1.79 (< MQL-31.2)	70	0.82 (< MQL-11.5)	67	0.72 (< MQL-13.2)
$\sum_5$DDTs		102 (8.36-1062)		20.4 (< MQL-1470)		13.8 (1.46-1229)		12.0 (1.65-1203)

[a]Df: detection frequency; [b]MQL: method quantification limit.

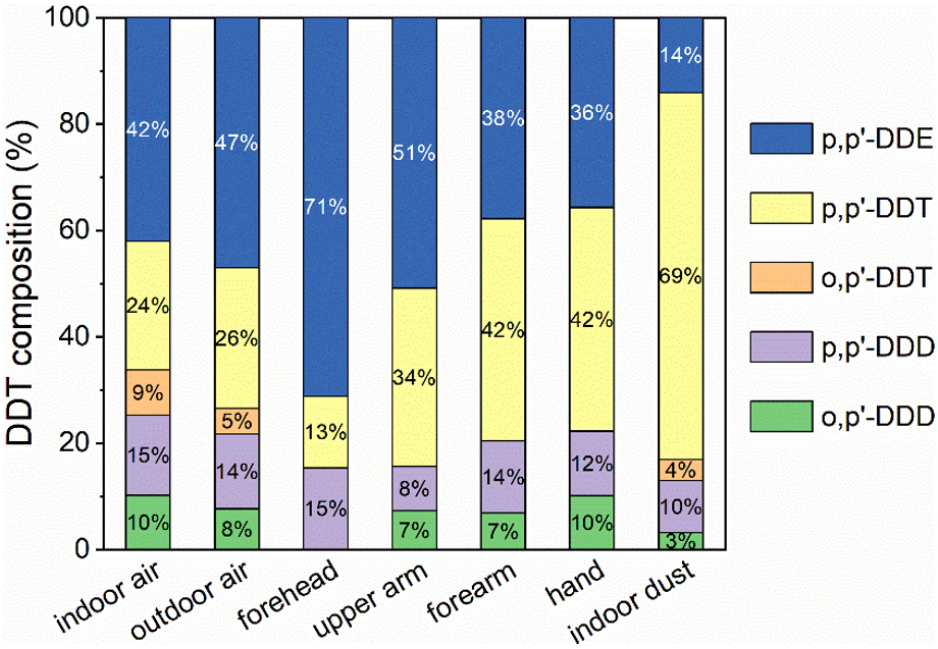

Figure 1. Compositions of individual DDTs in skin wipes, dust and gaseous air samples. Dust and air data are from Lv *et al.* (2023)[15]. The percentages were calculated with the median concentrations of DDT.

accumulated mainly through dust adhesion, but since both skin locations were exposed to air during the summer when the samples were collected, air-to-skin transport also more or less contributed to the levels of DDT on the surfaces of these sites. And the contribution of air-to-skin transport route could reduce the proportion of *p,p'*-DDT and increase the proportion of *p,p'*-DDE in these two skin sites relative to the dust, which is consistent with the results shown in Figure 1.

Occurrence of DDTs in the foodstuffs

Concentrations and detection frequencies of DDT isomers in foods are provided in Table 2. The two isomers, *o,p'*-DDT and *o,p'*-DDD, were not detected in any of the dietary samples. Moreover, *o,p'*-DDE was detected in all fish samples, with a median concentration of 0.01 ng/g ww, but was not found in any other samples. Therefore, only the three DDT isomers (*p,p'*-DDT, *p,p'*-DDD, and *p,p'*-DDE) in dietary samples are further discussed herein. *p,p'*-DDE was the most prevalent target chemical in food samples, being detected in half of the raw chicken and in all of the fish, hen egg, vegetable, and roast goose/duck samples. The average residual levels of *p,p'*-DDE in six food categories were in the order: egg (2.04 ± 3.13 ng/g ww) >

Table 2. Concentrations of DDT residue in foodstuffs (ng/g wet weight)

Food category	*p,p'*-DDE		*o,p'*-DDE		*p,p'*-DDT		*p,p'*-DDD		$\sum_4$DDTs
	Df (%)[a]	Median (range)	Df (%)	Median (range)	Df (%)	Median (range)	Df (%)	Median (range)	Median (range)
Chicken egg	100	0.57 (0.01-9.46)	0		73	0.46 (< MQL-3.19)	64	0.03 (< MQL-0.19)	1.10 (0.01-12.8)
Fish	100	0.22 (0.10-1.27)	100	0.01 (0.01-0.02)	29	< MQL (< MQL–0.13)	100	0.08 (0.02–0.63)	0.39 (0.13-1.98)
Vegetable	100	0.02 (0.02-0.88)	0		0		11	<MQL (< MQL-0.01)	
Roast goose/duck	100	0.07 (0.02-0.12)	0		0		78	0.05 (< MQL-0.09)	0.12 (0.06-0.20)
Raw chicken	50	0.03 (< MQL-0.20)	0		0		0		0.03 (< MQL-0.20)
Raw pork	0		0		0		0		

[a]Df: detection frequency; [b]MQL: method quantification limit.

fish (0.36 ± 0.36 ng/g ww) > vegetable (0.13 ± 0.28 ng/g ww) > roast goose/duck (0.08 ± 0.03 ng/g ww), raw chicken (0.05 ± 0.08 ng/g ww) > raw pork (not detected). *p,p'*-DDD was not detected in any of the pork and chicken samples, but was detected in one of the nine kinds of vegetables. For the other food species, the detection frequencies of *p,p'*-DDD varied from 64% (egg) to 100% (fish). The highest *p,p'*-DDD concentration was found in fish samples (0.63 ng/g ww). *p,p'*-DDT was detected in 73% of egg samples with average concentrations of 0.78 ± 0.97 ng/g ww and detected in 29% of fish samples with average concentrations of 0.03 ± 0.05 ng/g ww. No *p,p'*-DDT was detected in the other food samples. The variability in DDT concentrations among different food types may be attributed to the potential contamination in a variety of processes, including food production, transportation, processing (or cooking), storage, and so forth[10,38]. Our previous studies reported new input of DDTs in the indoor and outdoor environment in the study area[15,30], from which it can be speculated that the accumulated DDTs in hen eggs and fish may be mainly derived from local contamination.

The Chinese total diet study conducted in 2007 measured DDT concentrations in different food items[39]. And DDT concentrations in fish, meat, and vegetables in the present study were observed to decrease by approximately one to two orders of magnitude, as compared to 2007, reflecting the effectiveness of the DDT control policy. However, the DDT levels in hen eggs in this study (0.01-12.8 ng/g ww, average 2.87 ± 4.14 ng/g ww) were slightly higher than those reported in 2007 (<MQL-7.32 ng/g ww, 1.19 ± 2.01 ng/g ww), indicating that the new source of DDT found in the study area cannot be ignored. The current DDT concentrations detected in this study were similar to those in North China surveyed in almost the same period[40], but were significantly lower than the values reported in Pakistan[12] and some African countries[14,41].

Human dermal, hand-mouth contact and dietary absorption

The whole-body dermal exposure levels of $\sum_5$DDTs (*o,p'*-DDT, *p,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, and *p,p'*-DDE) ranged from 0.0226 to 7.23 ng/kg/d with a median value of 0.187 ng/kg/d [Table 3]. Of the five DDT isomers, *o,p'*-DDT exhibited about two orders of magnitude lower dermal exposure levels than the other four isomers. This is not only due to the lower concentration of *o,p'*-DDT in skin wipes, but also due to its lower skin permeability coefficient, which partly limits its daily levels of dermal absorption[42]. Therefore, the magnitude of skin exposure depends on both the concentration of compound and its physicochemical properties.

Table 3. The estimated median daily absorption of DDTs via dermal contact, hand-mouth contact, and food ingestion (ng/kg/d)

	p,p'-DDE	*o,p'*-DDE	*p,p'*-DDT	*o,p'*-DDT	*p,p'*-DDD	*o,p'*-DDD	$\sum_5$DDTs[a]	$\sum_4$DDTs[b]
Dermal exposure levels (ng/kg/d)								
Whole-body	8.19E-02		1.59E-02	3.37E-04	2.25E-02	1.57E-02	1.87E-01	
Hand-mouth contact exposure levels (ng/kg/d)								
	7.58E-03		8.29E-03	0	2.35E-03	2.08E-03	2.37E-02	
Dietary ingestion levels (ng/kg/d)								
egg	1.83E-01	0	1.49E-01		1.09E-02			3.57E-01
fish	2.01E-01	1.15E-02	0		6.97E-02			3.49E-01
vegetable	1.09E-01	0	0		0			1.09E-01
Roast goose/duck	9.87E-02	0	0		6.11E-02			1.54E-01
Raw chicken	2.56E-02	0	0		0			2.56E-02
Raw pork	0	0	0		0			0
Total diets	6.17E-01	1.15E-02	1.49E-01		1.42E-01			9.94E-01

[a]Sum of *p,p'*-DDE, *p,p'*-DDT, *o,p'*-DDT,*p,p'*-DDD, and *o,p'*-DDD; [b]Sum of *p,p'*-DDE, *o,p'*-DDE, *p,p'*-DDT,and *p,p'*-DDD.

Although the DDT concentrations were higher in forehead than in the other three skin locations, the absorption of DDTs from the skin of forehead was found to be lower than in the other three skin areas due to the smaller skin area [Supplementary Figure 1]. Similarly, because of the relatively large skin surface area of the clothing-covered sites, the DDT absorption levels from the skin covered by clothing were comparable to those from the uncovered skin area [Supplementary Figure 2], indicating the significance of assessing skin exposure in the covered area by clothing. The same conclusion was reported in a recent study of organophosphate flame retardants by Fan *et al.*[22]. These results also indicate that previous exposure studies considering only skin absorption from hand significantly underestimated the dermal exposure levels of contaminants.

Oral uptake levels of $\sum_5$DDTs through hand-mouth contact varied from 0 to 1.47 ng/kg/d with a median of 0.0237 ng/kg/d, which was obviously lower than the whole-body dermal exposure levels [Table 3]. This result was consistent with those of other chemicals reported by Guo *et al.*[28] and Cao *et al.*[16]. In these two studies, whole-body dermal absorption was 1-2 order of magnitude higher than hand-mouth exposure for polycyclic aromatic hydrocarbons, halogenated flame retardants, and polychlorinated biphenyls, highlighting the greater contribution of skin exposure to total human exposure than hand-mouth exposure.

The levels of the total dietary uptake of $\sum_4$DDTs (*p,p'*-DDT, *p,p'*-DDD, *o,p'*-DDE, and *p,p'*-DDE) ranged from 0.28 to 10.3 ng/kg/d with a median value of 0.994 ng/kg/d [Table 3], which were comparable to that reported in North China[40]. Egg and fish contributed to 71% of total food ingestion, with median values of 0.357 ng/kg/d and 0.349 ng/kg/d, respectively, followed by roast goose/duck (0.154 ng/kg/d), vegetable (0.109 ng/kg/d), and chicken (0.0256 ng/kg/d). Of the 11 pooled egg samples, eight were bought from villages where free-range hens were raised around the house and the other three pooled egg samples were bought from local markets. These eggs were laid by hens in intensive chicken farms. The DDT absorption by consumption of market eggs (0.002-0.046 ng/kg/d) was significantly lower than that by free-range eggs (0.153-4.157 ng/kg/d). Accordingly, less consumption of locally DDT-contaminated free-range eggs and fish may be helpful in reducing human dietary uptake of DDTs.

Comparison of the different exposure pathways of DDTs

DDT concentrations in air and dust from the daily activity area of the participants recruited in this study were reported in one of our previous studies[15]. In order to compare different human exposure pathways of

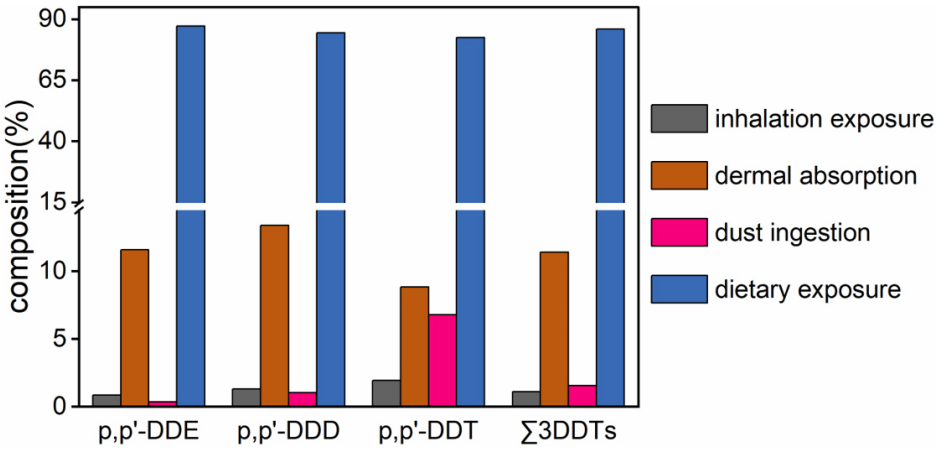

Figure 2. Comparison between different exposure pathways (dermal absorption, inhalation exposure, dust and diet ingestion) of DDTs. Dust and air data are from Lv *et al.* (2023)[15]. The percentages were calculated with the median exposure levels of DDT.

DDTs, these air and dust data were used in the present study to calculate the respiratory exposure and dust ingestion of DDTs. Detailed assessment formulas and parameters are provided in Supporting Information [Supplementary Text 2]. Comparisons among the four exposure pathways (dermal absorption, inhalation exposure, dust and diet ingestion) are shown in Figure 2. Given the different DDT isomers detected in food and non-food samples, only three isomers (*p,p'*-DDT, *p,p'*-DDD, and *p,p'*-DDE) were used for the comparison here. The percentages of the human daily absorption of DDTs through food consumption, dermal contact, dust intake, and inhalation were 82.5%-87.2%, 8.82%-13.4%, 0.35%-6.77%, and 0.86%-1.94%, respectively. This result seems to be consistent with many previous studies in which food ingestion is considered to be the most dominant route of human exposure to DDTs. It should be noted that the exposure dose by dermal absorption was in the same order of magnitude as those of food consumption, regardless of the median levels or the max levels. Therefore, the contribution of dermal exposure cannot be ignored in this case.

However, the real case might be much more complicated than this. The dietary uptake levels depend on pollution levels in foodstuffs, food consumption rate, and bioaccessibility via food ingestion[40,43]. It should be noted that cooking or not, different cooking methods could affect food pollution levels[10]. Different individuals, and even the same individuals in different seasons, may have different food type consumption preferences and intake rates[40]. In addition, the gastrointestinal bioaccessibility of a chemical in food is associated not only with the chemical's physicochemical properties[34], but also with the food category[44]. These factors are currently not fully elucidated and thus not incorporated in our dietary assessment, making it difficult to conclude that dietary exposure must be the most dominant route of exposure to DDTs.

CONCLUSION

This study provides a more comprehensive understanding of human exposure to DDTs. The comparison of DDT profiles among various sample media indicates that air-skin transport may be the major route of dermal exposure to DDTs in forehead and upper arm locations, while dust adhesion may be the main route of dermal exposure to DDTs in hand and forearm locations. The eggs laid by the free-range chickens in the villages and wild fish were the major sources of DDT in the diet of local residents, so we recommend avoiding consuming these foods and consuming farm eggs and farm fish as alternatives. These factors, such as cooking and gastrointestinal bioaccessibility of DDTs in various foods, need to be studied in the future in order to conduct a more accurate assessment of human exposure to DDTs, and thus to gain a better

understanding of the relative importance of dermal and dietary exposure to DDTs.

DECLARATIONS

Authors' contributions

Conception and design of the study and performed data analysis and interpretation: Luo XJ, Lv YZ
Performed data acquisition: Lv YZ, Feng QJ, Zhu CY
Administrative, technical, and material support: Zeng YH, Mai BX

Availability of data and materials

Data published as supplementary information in the journal.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

This work has received approval for research ethics from Guangzhou Institute of Geochemistry (GIG-2019-001). Consent was obtained from all the participants after they were informed of the purpose of the study.

Consent for publication

Not applicable.

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