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Organophosphate esters in tree bark and human hair in Weifang and Yantai, Shandong Province, China: concentrations, profiles and sources

Shijie Wang[1], Anming Liu[2], Meihong Li[1], Jingxi Jin[1], Haodong Yuan[1], Jicheng Hu[1,3], Ying Wang[1,3], Jun Jin[1,3]

[1]College of Life and Environmental Sciences, Minzu University of China, Beijing 100081, China.
[2]Binhai Branch of the Weifang People's Hospital, Weifang 262737, Shandong, China.
[3]Engineering Research Center of Food Environment and Public Health, Beijing 100081, China.

Correspondence to: Prof. Jun Jin, College of Life and Environmental Sciences, Minzu University of China, Haidian District, Beijing 100081, China. E-mail: junjin3799@126.com

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Abstract

Organophosphate ester (OPE) concentrations and distributions in 15 tree bark and 59 human hair samples from Weifang and Yantai (Shandong Province, China) were determined. The total OPE concentrations in tree bark samples from Weifang and Yantai were 16.5 ng/g-78.5 ng/g and 9.34 ng/g-98.6 ng/g dry weight (dw), respectively. The total OPE concentrations in hair samples from Weifang and Yantai were 54.2 ng/g dw-8450 ng/g dw and 7.26 ng/g dw -13,900 ng/g dw, respectively. Chlorinated OPEs were dominant in the tree bark samples from both Weifang and Yantai. Tris(2-chloroethyl)phosphate (TCEP) was the dominant OPE in the hair samples from residents of Weifang and Yantai. Attention should be paid to human exposure to TCEP, which is carcinogenic. The OPE concentrations in human hair were highest for the < 20 years age group. The OPE patterns between the two cities were similar for both tree bark and hair, but the OPE patterns in the tree bark and hair samples from the same place were significantly different. This indicates that the outdoor atmosphere may not be the main source of OPEs in human hair.

Keywords: Organophosphate esters (OPEs), tree bark, human hair, concentrations, sources

INTRODUCTION

Organophosphate esters (OPEs) are used as flame retardants and plasticizers to protect or enhance the properties of electronic equipment, furniture, plastics, textiles, and other materials[1]. Global restrictions on the production and use of polybrominated diphenyl ethers (e.g., the banning of penta- and octa-bromodiphenyl ethers and the phase-out of decabromodiphenyl ether) have led to sharp increases in OPE intake[2]. OPEs are usually added to (but not chemically bonded to) polymers. Similar to other organic pollutants, OPEs can enter the body through airways, dermal absorption, or ingestion. It has been suggested that some OPEs may be carcinogens, endocrine disruptors, neurotoxicants, and thyroid disruptors[2-8]. OPEs in polymers may therefore be released into the environment through volatilization, abrasion, or leaching while the treated products are produced, used, and disposed of Refs.[1,9,10]. OPEs have been found widely in the atmosphere[11,12], sediment[13], drinking water[14], and biota[15-18].

Tree bark has a relatively high lipid content, is widespread, and is convenient to sample, so it is often analyzed to allow persistent organic pollutant concentrations in the atmosphere to be monitored[19-21]. Atmospheric deposition is the primary pathway for compounds to enter tree bark. The bark is in direct contact with the atmosphere, and the particulate state of the atmosphere is easily trapped by the bark pores, while the gaseous state can enter the bark via gas diffusion transcutaneous pores. On the other hand, the scleroderma is filled with suberin, which is a lipid that strongly adsorbs lipophilic POPs[22]. Bark surfaces are very porous. Metabolic processes do not occur in bark, so inorganic and organic chemicals in bark are little affected by degradation. It is reported that the outer bark will shed and grow from inside after approximately two years[23]. Bark is therefore a useful passive bioindicator of airborne pollutants[24]. However, data on tree bark OPE concentrations are extremely limited[25,26]. OPEs have ubiquitous detection in indoor environments[27-32]. In general, OPEs are more abundant in indoor than in outdoor environments[10,33]. Concerns about human exposure and possible environmental effects are still rising[10], and it is still worth exploring the correlation between OPE in the outdoor environment and OPE in the human body. Hair is a keratinous matrix consisting of 88% protein and 3%-4% lipid. Persistent organic pollutants concentrations in hair can be determined easily[34]. Hair is cheap to obtain, accessible, stable, and easily transported and stored[9], and thus is a good example of a matrix that can be acquired noninvasively to allow human biomonitoring to be performed. Hair can accumulate pollutants through external exposure (deposition from air and dust) and internal exposure (through contact with blood at the hair follicle/root)[35,36]. Hair can provide retrospective and integral exposure of pollutants as a biomarker, since it is difficult to distinguish external and internal exposure[10]. Hair is often used to identify the main pathway through which humans are exposed to persistent organic pollutants[37-39]. To date, many studies have used hair to assess human exposure to various organic pollutants, such as polychlorinated biphenyls[40], polybrominated diphenyl ethers[41], polycyclic aromatic hydrocarbons[42], and Dechlorane Plus[43]. Few studies of OPE concentrations in hair have been published[9,10,36,44,45], and most have focused on economically developed cities (e.g., Chongqing and Guangzhou) or specific groups of people (e.g., children, mothers, and students). Few studies of OPE concentrations in the general population of China have been performed. Furthermore, associations between OPE concentrations in tree bark and human hair from the same areas have not previously been assessed. Tree bark can reflect local air pollution. Hair has been identified as a potential noninvasive matrix to assess the level of human exposure to POPs[41]. The combination of the two matrices can help to identify whether the source of OPEs comes from the outdoor atmospheric environment.

The aims of this study were: (1) to determine the concentrations of nine OPEs in bark samples from Weifang and Yantai (in Shandong Province, China) and in hair samples from people of different ages living in Weifang and Yantai; and (2) to identify trends in the OPE patterns in hair from people of different ages

and the sources of OPEs to human hair.

EXPERIMENTAL

Sampling

The samples were collected in May 2016 in Weifang and Yantai, in Shandong Province, China. Bark was removed from around the trunks of three pine trees at each site, at ~1.5 m above the ground (the average height at which humans inhale), using a chisel. The mean trunk diameter at the sampling height was 35 cm. Trees of this size were selected to ensure that the bark had been exposed for enough time to effectively sorb pollutants from the air. The tree bark samples were freeze-dried, and then we used stainless steel scissors to cut the bark into small, uniform pieces. The stainless-steel scissors were sonicated with hexane three times for 10 min before use. In total, 15 bark samples were collected (eight in Weifang and seven in Yantai, Supplementary Table 1). The hair samples collected and analyzed in this study are the same as those reported in our previous study[46,47]. Hair samples were collected from people living within 10 km of where the tree bark samples were collected. It is reported that the hair growth rate is 1 cm/month[48]. In this study, a hair sample of the posterior occipital area was collected at 3 cm from the scalp with stainless scissors to represent about three months of exposure. Each person provided 2 g -3 g of occipital posterior area hair. In total, 59 non-dyed hair samples were collected. Samples were collected from 26 people (mean age 29 years) in Weifang and 33 people (mean age 33 years) in Yantai. The samples were stored at -20 °C until they were analyzed.

Sample preparation

Chemicals: Tris(2-chloroethyl)phosphate (TCEP)-d_{12} and triphenyl phosphate (TPHP)-d_{15} were purchased from Cambridge Isotope Laboratories. All organic solvents were obtained from J.T. Baker (Phillipsburg, NJ, USA) and were liquid chromatography or pesticide analysis grade. Ultra-pure water was produced using a Milli-Q system (EMD Millipore, Billerica, MA, USA). Silica gel was purchased from Merck. Neutral alumina and anhydrous sodium were purchased from Sinopharm Chemical Reagent.

Each bark sample was freeze-dried and crushed, and then a 3.0 g aliquot was spiked with internal standards [50.0 ng tris(2-chloroethyl)phosphate (TCEP)-d_{12} and 50.0 ng triphenyl phosphate (TPHP)-d_{15}] and extracted with 30 mL of a 1:1 (v/v) hexane/acetone mixture with ultrasonication for 15 min. The solvent was removed, and then the extraction was repeated two times. The extracts were combined and then evaporated to 1.0 mL. The extract was then passed through a column filled from bottom to top with quartz wool, 5.0 g neutral alumina, 8.0 g neutral silica gel, and 2.0 g anhydrous sodium. Before use, the column was cleaned with 30 mL of a 1:1 (v/v) hexane/dichloromethane mixture, and then the sample extract was added and the column eluted with 100 mL of ethyl acetate. The extract was then evaporated to 1.0 mL and passed through a gel permeation chromatography column. The extract was next evaporated to a small volume, and a glass tube with a capacity of 500 μL was used to determine the volume of the concentrated extract.

Each hair sample (n = 59) was cut into small pieces and cleaned of external contaminants by placing it in a vessel containing Milli-Q water, placing the vessel in an ultrasonic bath for 10 min, and removing the water and repeating the cleaning procedure two times. The sample was then allowed to dry naturally in the air. A 25.0 mg aliquot of a clean hair sample was added to 2.5 mL of a 1:1 (v/v) hexane/acetone mixture containing internal standards (50.0 ng TCEP-d_{12} and 50.0 ng TPHP-d_{15}). The mixture was allowed to stand at room temperature for 20 min, and then was placed in a refrigerator at 4 °C overnight. The mixture was then centrifuged at 2500 rpm for 5 min, and the supernatant was removed. The sample was extracted two more times with 1 mL of a 1:1 (v/v) hexane/acetone mixture, and then the supernatants were combined. The extract was finally evaporated to 500 μL under a gentle stream of nitrogen.

Instrumental analysis

The extracts were analyzed using a TRACE 1310 gas chromatograph with a TSQ 8000 Evo triple quadrupole mass spectrometer (Thermo Fisher Scientific, Waltham, MA, USA). The mass spectrometer was operated in electron impact ionization and selected reaction monitoring modes. The carrier gas was helium, and the flow rate was 1.0 mL/min. The source temperature was 280 °C. The OPEs were separated using a TG-5HT column (30 m long, 0.25 mm i.d., 0.1 μm film thickness; Thermo Fisher Scientific). The oven temperature program started at 100 °C, which was held for 2 min, and then it was increased at 20 °C /min to 300 °C, which was held for 3 min.

Quality control and assurance

The target compounds were quantified using an internal standards method. The standard curve for each analyte had a correlation coefficient > 0.9995. The TCEP-d_{12} and TPHP-d_{15} recoveries in the bark samples were 79% ± 13% and 83% ± 6%, respectively. The TCEP-d_{12} and TPHP-d_{15} recoveries in the hair samples were 107% ± 15% and 96% ± 13%, respectively. The detection limits were defined as the concentrations giving signal-to-noise ratios of 3. Every sample batch included a method blank. The analyte concentrations in the blank samples were < 10% of the concentrations in the samples; thus, the concentrations in the samples were not blank-corrected. Ma *et al.*[49] and Li *et al.*[16] previously documented the method. The limits of detection for the analytes in bark and hair were 2.01 pg/g-18.4 pg/g and 70.0 pg/g-1960 pg/g dw, respectively. An analyte concentration below the limit of detection was assigned a value of zero when data analysis was performed.

Statistical analysis

All statistical analyses were performed using Origin 9.6 and SPSS 22.0 for Windows software. Paired-samples *t*-tests ($P < 0.05$) were used to compare groups of results. Spearman correlation analyses and regression analyses were used to identify relationships between the concentrations of different OPEs in the hair and tree bark samples.

RESULTS AND DISCUSSION

OPEs in tree bark

OPE concentrations in tree bark

The detection frequencies and concentrations of the nine OPEs that were analyzed [Tris(2-chloroethyl)phosphate (TCEP), tris(2-chloroisopropyl) phosphate (TCPP), tris(2,3-dichloropropyl) phosphate (TDCPP), tris(2-ethylhexyl) phosphate (TEP), tris(*n*-butyl)phosphate (TNBP), triphenyl phosphate (TPHP), tris(*o*-cresyl) phosphate (TOCP), tris(*m*-cresyl) phosphate (TMCP), and tris(*p*-cresyl) phosphate (TPCP)] in bark from Weifang and Yantai are shown in Table 1. The total ($\sum_9$) OPE concentrations in the bark samples from Weifang were 16.5 ng/g dw-78.5 ng/g dw (median 23.0 ng/g dw), and the $\sum_9$OPE concentrations in the bark samples from Yantai were 9.34 ng/g dw-98.6 ng/g dw (median 44.9 ng/g dw) [Figure 1]. The TEP detection frequencies for the bark samples from Weifang and Yantai were 100% and 86%, respectively. The other eight OPEs were detected in all of the bark samples (i.e., the detection frequencies were 100%). The high detection frequencies of the OPEs for bark from both Weifang and Yantai strongly imply that OPEs have been widely used in Shandong Province in recent years.

OPE profiles in tree bark

As shown in Figure 2, the chlorinated OPEs (TCEP, TCPP, and TDCPP) and non-chlorinated OPEs (TEP, TNBP, TPHP, TOCP, TMCP, and TPCP) contributed 81.5% and 18.6%, respectively, of the $\sum_9$OPE concentrations in the bark samples from Weifang. For bark samples from Yantai, the contributions of chlorinated and non-chlorinated OPEs were 88.9% and 11.2%, respectively. There were no significant differences between the OPE concentrations in the bark samples from Weifang and Yantai (paired-samples

Table 1. Concentrations (ng/g dry weight) of nine organophosphate esters in tree bark samples from Weifang and Yantai, Shandong Province, China

	Weifang bark *n* = 8				Yantai bark *n* = 7			
	Range	Mean ± SD	Median	DF(%)	Range	Mean ± SD	Median	DF(%)
TEP	0.692-6.17	2.19 ± 1.77	1.51	100	ND-5.83	2.04 ± 1.83	1.59	86
TNBP	0.212-4.08	1.16 ± 1.29	0.580	100	0.364-4.67	2.06 ± 1.43	1.67	100
TCEP	3.39-33.4	12.2 ± 12.0	6.11	100	2.87-25.6	17.2 ± 7.80	18.3	100
TCPP	1.72-17.6	6.20 ± 6.02	3.80	100	1.54-59.7	21.1 ± 18.8	13.6	100
TPHP	0.261-2.53	1.10 ± 0.890	0.725	100	0.273-1.52	0.762 ± 0.445	0.833	100
TOCP	0.0334-0.650	0.320 ± 0.207	0.294	100	0.00558-0.301	0.129 ± 0.100	0.0921	100
TMCP	0.172-1.56	0.499 ± 0.456	0.332	100	0.101-0.566	0.341 ± 0.153	0.314	100
TPCP	0.216-6.83	1.18 ± 2.29	0.369	100	0.129-0.493	0.292 ± 0.119	0.262	100
TDCPP	4.33-13.2	9.95 ± 3.27	10.7	100	0.603-12.0	6.48 ± 4.43	4.20	100
$\sum_9$OPEs	16.5-78.5	34.8 ± 23.1	23.0		9.34-98.6	50.4 ± 26.8	44.9	

TCEP: Tris(2-chloroethyl)phosphate; TCPP: tris(2-chloroisopropyl)phosphate; TEP: tris(2-ethylhexyl)phosphate; TNBP: tris(*n*-butyl)phosphate; TPHP: triphenyl phosphate; TOCP: tris(*o*-cresyl)phosphate; TMCP: tris(*m*-cresyl)phosphate; TPCP: tris(*p*-cresyl)phosphate; TCP: tricresyl phosphate, TOCP + TMCP + TPCP; TDCPP: tris(2,3-dichloropropyl)phosphate; OPEs: organophosphate esters; SD: standard deviation; ND: not detected; DF: detection frequency (%).

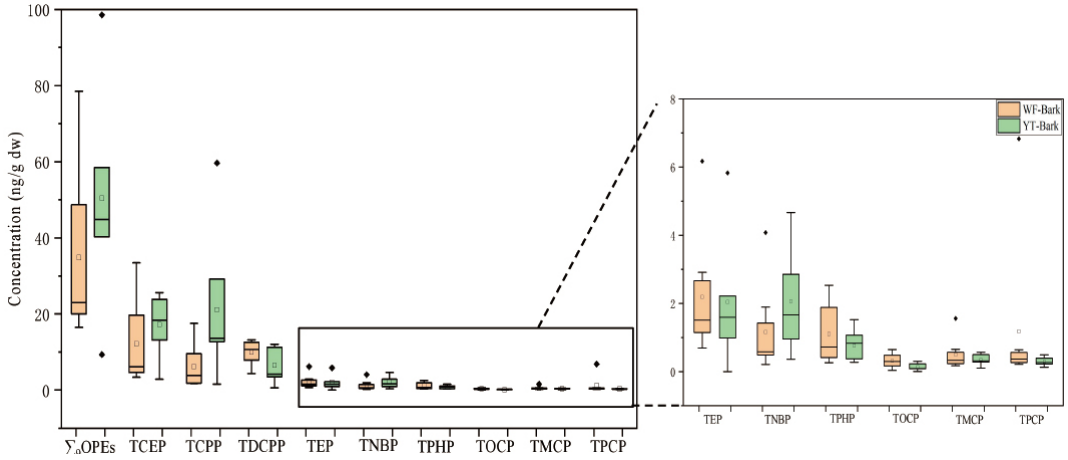

Figure 1. Concentrations of organophosphate esters in tree bark from Weifang (WF) and Yantai (YT), Shandong Province, China. OPEs: Organophosphate esters; TCEP: Tris(2-chloroethyl)phosphate; TCPP: tris(2-chloroisopropyl)phosphate; TDCPP: tris(2,3-dichloropropyl)phosphate; TEP: tris(2-ethylhexyl)phosphate; TNBP: tris(*n*-butyl)phosphate; TPHP: triphenyl phosphate; TOCP: tris(*o*-cresyl)phosphate; TMCP: tris(*m*-cresyl)phosphate; TPCP: tris(*p*-cresyl)phosphate.

t-test, $P > 0.05$), indicating that OPE pollution in the outdoor atmosphere is similar in Weifang and Yantai. Chlorinated OPEs were the dominant OPEs in bark from Weifang and Yantai, which agreed with the results of our previous studies on OPEs in human and environmental media from Weifang[49,50]. Chlorinated OPEs appear to be resistant to degradation[51], which may explain the bark from Weifang and Yantai having higher chlorinated OPE concentrations than non-chlorinated OPE concentrations.

In the study by Peverly, Salamova, and Hites[26], seven OPE compounds were measured in tree bark from St. Louis, Michigan, and the proportion of TDCPP was 43%. Ren *et al.*[25] reported three OPE congener concentrations of tree bark surrounding a manufacturing plant in Hengshui, China, and the most abundant compounds were TCPP and TCEP, which was consistent with our study. The Spearman correlation analysis

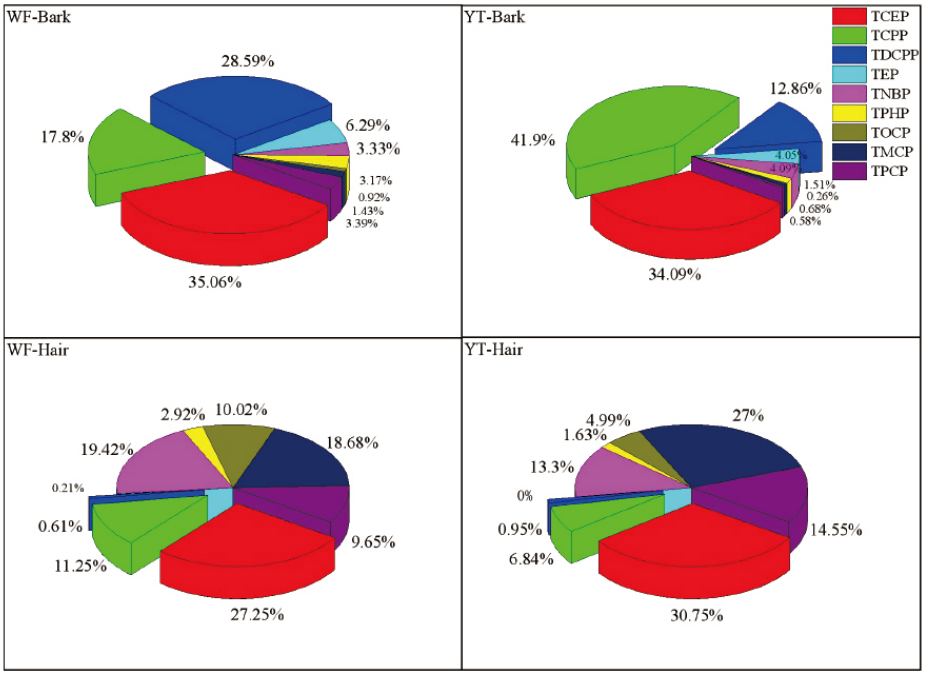

Figure 2. Contributions of nine organophosphate esters in human hair and tree bark samples from Weifang (WF) and Yantai (YT), Shandong Province, China. TCEP: Tris(2-chloroethyl)phosphate; TCPP: tris(2-chloroisopropyl)phosphate;TDCPP: tris(2,3-dichloropropyl)phosphate; TEP: tris(2-ethylhexyl)phosphate; TNBP: tris(*n*-butyl)phosphate; TPHP: triphenyl phosphate; TOCP: tris(*o*-cresyl)phosphate; TMCP: tris(*m*-cresyl)phosphate; TPCP: tris(*p*-cresyl)phosphate.

results indicate that the TCPP and TCEP concentrations in the bark samples from Weifang and Yantai significantly correlated ($r = 0.881$, $P < 0.01$ for Weifang and $r = 0.857$, $P = 0.014$ for Yantai). The TCPP and TCEP concentrations did not correlate with the TDCPP concentrations. The results show that the source of TDCPP to bark in Weifang and Yantai was different from the source of TCPP and TCEP. TCPP is often used as an alternative to TCEP[52,53], which could explain the relationship between the TCEP and TCPP concentrations in the bark samples in both sampling areas. Yantai City has the largest polyurethane industry in Shandong Province, and TCPP is often added to polyurethane. This is probably why the mean TCPP concentration in the bark samples from Yantai was about three times the mean TCPP concentration in the bark samples from Weifang. TDCPP is used as an additive flame retardant in foam, latex products, and resins. Most foams treated with TDCPP are used by the automotive industry, and some are used in furniture[51]. The TDCPP concentrations in the bark samples from Weifang and Yantai may have been rather high because of the automotive industry plants in Weifang and Yantai. There are plants at which TEP, TNBP, and TPHP are manufactured in Weifang, but these OPEs were not the dominant OPEs in the bark samples from Weifang. The TEP, TNBP, and TPHP concentrations in the bark samples from Weifang and Yantai were similar [Figure 1]. This may be due to the small effect of TEP, TNBP, and TPHP production on their concentration in the atmosphere in Weifang.

OPEs in hair

OPE concentrations in hair

As shown in Table 2, the median $\sum_9$OPE concentration in the hair samples from Weifang residents (340 ng/g dw) was higher than the median $\sum_9$OPE concentration in the hair samples from Yantai residents (122 ng/g dw). OPEs including TEP, TCEP, TPHP, TCP, and TDCPP are manufactured in Weifang, but no OPEs are manufactured in Yantai. OPEs are used as additives in treated materials and can therefore easily be released into the environment through abrasion or volatilization. Therefore, Weifang residents are

Table 2. Concentrations (ng/g dw) of organophosphate esters in hair from residents in Weifang and Yantai, Shandong Province, China

	Weifang hair (*n* = 26)				Yantai hair (*n* = 33)			
	Range	Mean ± SD	Median	DF (%)	Range	Mean ± SD	Median	DF (%)
TEP	ND-19.1	1.80 ± 4.16	ND	35	ND-0.462	0.0140 ± 0.0804	ND	3
TNBP	11.8-1560	168 ± 371	52.7	100	1.70-1450	158 ± 309	15.3	100
TCEP	8.45-3120	236 ± 616	54.7	100	ND-5320	366 ± 991	17.5	97
TCPP	ND-1960	97.6 ± 380	19.9	73	ND-1170	81.4 ± 212	12.9	97
TPHP	ND-136	25.3 ± 29.1	15.6	96	ND-144	19.4 ± 33.5	7.32	88
TOCP	ND-467	86.9 ± 91.9	68.1	100	ND-941	59.4 ± 171	0.798	64
TMCP	4.64-1890	162 ± 381	71.6	100	ND-3630	321 ± 751	2.54	94
TPCP	ND-1290	83.7 ± 259	18.9	85	ND-2410	173 ± 458	ND	45
TDCPP	ND-46.5	5.28 ± 10.9	ND	42	ND-116	11.3 ± 25.8	1.92	85
$\sum_9$OPEs	54.2-8450	867 ± 1690	340		7.26-13,900	1190 ± 2720	122	

TCEP: Tris(2-chloroethyl)phosphate; TCPP: tris(2-chloroisopropyl)phosphate; TEP: tris(2-ethylhexyl)phosphate; TNBP: tris(*n*-butyl)phosphate; TPHP: triphenyl phosphate; TOCP: tris(*o*-cresyl)phosphate; TMCP: tris(*m*-cresyl)phosphate; TPCP: tris(*p*-cresyl)phosphate; TCP: tricresyl phosphate, TOCP + TMCP + TPCP; TDCPP: tris(2,3-dichloropropyl)phosphate; OPEs: organophosphate esters; SD: standard deviation; ND: not detected; DF: detection frequency (%).

exposed to higher OPE concentrations than Yantai residents.

The TCEP concentrations in the hair samples from Weifang and Yantai (n = 59) ranged from not detected to 5320 ng/g dw (median 45.7 ng/g dw), and the TNBP concentrations were 1.70 ng/g dw-1570 ng/g dw (median 41.4 ng/g dw). The TCEP and TNBP concentrations were slightly higher than that previously found in human hair from Belgium, Chongqing, Guangzhou, and Norway[1,9,10,36]. TCEP is added to many cellulose products, coatings, polyester resins, polyurethane foams, polyvinyl chloride products, and textiles[51,54,55]. TCEP has a rather high saturated vapor pressure, so it readily volatilizes from products and enters the environment. High TCEP concentrations have been found in indoor environments[1,56,57]. TCEP was found in almost all of the hair samples we analyzed (detection frequency > 97%), indicating that TCEP is regularly present in the environment in Weifang and Yantai. TCEP is classed as "likely to be carcinogenic to humans" (IARC Group 3)[58]. TCEP is therefore not a good substitute for brominated flame retardants and should be used less than currently. TNBP has been used for a long time, is added to anti-foaming agents, glues, lacquers, paints, and waxes, and is used in industrial processes[55]. As shown in Table 2, the high TNBP concentrations in hair samples from various regions indicate that TNBP is currently widely used[9,10,36,44,45]. The TPHP concentrations in the hair samples from Weifang and Yantai ranged from not detected to 144 ng/g dw (median 11.9 ng/g dw). These concentrations were in the same order of magnitude as previously found in hair samples from Belgium, China (various regions), Chongqing, Guangzhou, and Norway[1,9,10,36]. TPHP is widely added to the foam used in furniture, general electronic equipment (e.g., lithium-ion batteries and video display units cables), and plastic products[55]. Electronic items may supply a large proportion of the TPHP found in the indoor environment[59]. TCPP and TDCPP are widely used as flame retardants in conveyor belts, polyurethane foam, and rubber products[55]. The TCPP and TDCPP concentrations in the hair samples from Weifang and Yantai were significantly correlated (r = 0.576, P < 0.01). We concluded that TCPP and TDCPP may be used as flame retardants in the same types of products. The TDCPP concentrations in hair from residents of Weifang and Yantai ranged from not detected to 116 ng/g dw (median 1.44 ng/g dw). The TCPP concentrations were higher (not detected to 1960 ng/g dw, median 14.1 ng/g dw). This may be due to the higher price of TDCPP, so it is only used when its flame retardant properties are particularly appropriate. The minimum and maximum OPE concentrations were somewhat different for each set of samples. The types of building materials, electric appliances, and

furniture used in the study areas might have caused the wide concentration ranges[45].

OPE profiles in hair

The contributions of the individual OPEs to the $\sum_9$OPE concentrations in the hair samples are shown in Figure 2. The most abundant OPEs in hair from Weifang residents were TCEP, TNBP, and TMCP, which contributed 27.3%, 19.4%, and 18.7%, respectively, of the $\sum_9$OPE concentrations. The most abundant OPEs in hair from Yantai residents were TCEP, TMCP, and TPCP, which contributed 30.7%, 27.0%, and 14.5%, respectively, of the $\sum_9$OPE concentrations. There were no significant differences between the OPE patterns in the hair samples from Weifang and Yantai (paired-samples *t*-test, $P > 0.05$). These results indicate that there were no significant differences between the exposure pathways and accumulation patterns for hair for Weifang and Yantai residents.

The OPE patterns in the hair samples were different from patterns found in previous studies. TNBP and TPHP were the dominant OPEs in hair samples from Norway[10], while TDCPP and TCEP were the dominant OPEs in hair samples from Belgium[36]. Liu, He, Hites and Salamova [31]Liu analyzed four OPEs in hair samples from the USA and found that TCPP was the dominant OPE[44]. The dominant OPEs in hair samples from college students in Guangzhou (China) were TDCPP, TEHP, and TPHP[45]. TCPP and TPHP were the dominant OPEs in hair from residents of rural areas in Chongqing, but TMPP was the dominant OPE in hair from residents of urban areas in Chongqing[9]. The OPE profiles in the hair samples were probably different because of different types of OPEs being used as flame retardants in different products[9,45].

The non-chlorinated OPEs contributed 60.9% and 61.5% of the $\sum_9$OPE concentrations in the hair samples from residents of Weifang and Yantai, respectively. This contrasted with the contributions in the bark samples from Weifang and Yantai. Studies have shown that the concentration of lipophilic pollutants in bark is correlated with the atmospheric concentration measured nearby[26]. The OPE patterns in the human hair and tree bark samples from the same place were significantly different (paired-samples *t*-test, $P < 0.05$). The results show that the outdoor atmosphere may not be the main source of OPEs in the hair of residents of Weifang and Yantai. TCP is a mixture of mainly three isomers: TOCP, TMCP, and TPCP. The total TOCP, TMCP, and TPCP contributions to the $\sum_9$OPE concentrations in hair from the Weifang and Yantai residents were 38.4% and 46.5%, respectively. TOCP, TMCP, and TPCP are used as flame retardants in building materials, electronics, furniture, machinery, plastics, and textiles[49,51,60]. They are used widely and have higher octanol-water distribution coefficient (log *Kow* = 5.11, Supplementary Table 2), so they may be easily sorbed onto human hair. van der Veen and de Boer[51] reported that the isomers of TCP have different degradation rates, with TOCP degrading more quickly than TMCP and TPCP. This may be the reason TMCP and TPCP accounted for a relatively high proportion of OPEs in Weifang and Yantai human hair. The indoor environment (classrooms, dormitories, houses, and offices) and other sources of OPEs may contribute significantly to OPE concentrations in human hair[33,61,62], and these sources may have caused the non-chlorinated OPE concentrations to be higher than the chlorinated OPE concentrations in hair from the residents of Weifang and Yantai.

OPE in hair from residents in different age groups

The hair samples were divided into groups based on the donors' ages (< 20, 20 years-39 years, and 40 years-60 years). The concentrations of the OPEs were higher in hair from the < 20 years group than in hair from the other age groups for residents of both Weifang and Yantai [Figure 3], but there were no statistical differences (Mann-Whitney *U* tests, $P > 0.05$). The highest TCPP concentrations were found in hair from residents of Weifang and Yantai in the 20 years-39 years group. It is interesting that the highest OPE

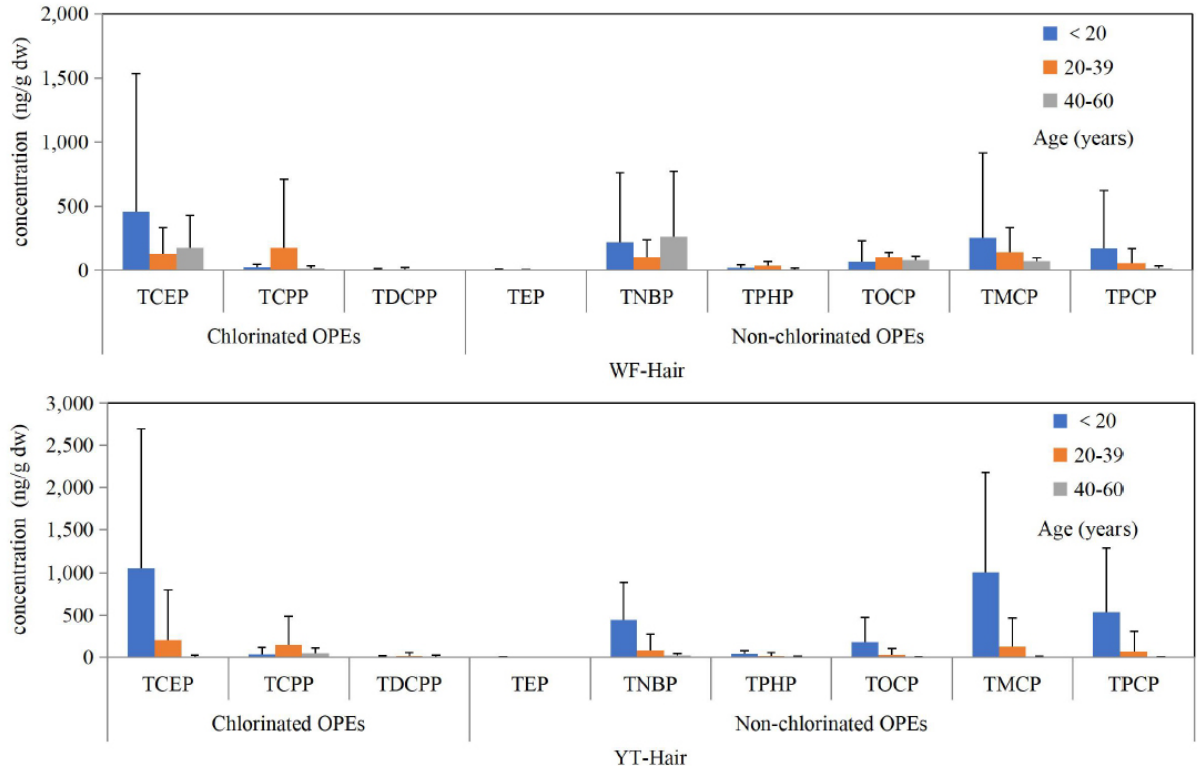

Figure 3. Chlorinated and non-chlorinated organophosphate ester (OPE) concentrations in hair samples from residents of Weifang (WF) and Yantai (YT) grouped by the residents' ages. (Weifang, < 20 years (n = 8), 20 years-39 years (n = 13), and 40 years-60 years (n = 5); Yantai, < 20 years (n = 9), 20 years-39 years (n = 12), and 40 years-60 years (n = 12)). TCEP: Tris(2-chloroethyl)phosphate; TCPP: tris(2-chloroisopropyl)phosphate; TDCPP: tris(2,3-dichloropropyl)phosphate; TEP: tris(2-ethylhexyl)phosphate; TNBP: tris(*n*-butyl)phosphate; TPHP: triphenyl phosphate; TOCP: tris(*o*-cresyl)phosphate; TMCP: tris(*m*-cresyl)phosphate; TPCP: tris(*p*-cresyl)phosphate.

concentrations were found in hair from young people. OPEs have been used as flame retardants for a relatively short time, but they have been widely used as flame retardants in various products (electronic equipment, household items, building materials, plastic products, tires, toys, and other products)[5]. Most people spend most of their time indoors, so OPE concentrations in the indoor environment can strongly affect human exposure to OPEs[57,63,64]. Compared with other age groups, young people have different lifestyles, such as the types and quantities of consumer goods they own and their frequency of cleaning[33,62]. Moreover, the reason for the highest OPEs level of OPEs in people aged < 20 years could be related to the living and learning environment, including classrooms and dormitories, which have strict fire safety regulations as public buildings[62]. This might explain why the OPE concentrations were higher in the hair samples from people aged < 20 years than in the hair samples from people > 20 years.

The TCEP and non-chlorinated OPE concentrations in hair decreased as age increased for the samples from the Yantai residents [Figure 3]. However, the TCEP and TNBP concentrations were higher for the 40 years-60 years group than for the younger groups for the hair samples from the Weifang residents, but there were no statistical differences (Mann-Whitney U tests, $P > 0.05$). The OPE patterns in the hair from Weifang and Yantai residents were similar (paired sample t-test, $P > 0.05$), but the age-related trends showed some differences, indicating that Yantai inhabitants may have slightly different exposures to OPEs over time.

Potential sources of OPEs

Principal component analysis (PCA) was used to investigate the source of OPEs in the tree bark and hair samples of Weifang. Components 1 and 2 accounted for 41.2% and 15.5% of the total variance, respectively [Figure 4A]. Tree bark samples (WB1-WB8) were plotted toward the negative direction of Component 1, whereas human hair samples were mainly located to the right of Figure 4A. The positive direction of Component 2 was mainly the hair samples from people aged < 20 years (WH1-WH8), and the negative direction was mainly the bark samples and the hair samples from people aged > 20 years. The PCA score plots indicate that all tree bark samples (WB1-WB8) were grouped in Group I. Group II mainly included the hair samples from people aged < 20 years (WH1-WH8).

According to the PCA score plots of the tree bark and hair samples of Yantai [Figure 4B], Components 1 and 2 accounted for 34.3% and 20.9% of the total variance, respectively. All bark samples of Yantai (YB1-YB7) were grouped in Group I. Group II mainly included the hair samples from people aged < 20 years (YH1-YH9). The result of PCA indicates that < 20-year-old local residents may have different exposure characteristics of OPEs compared to the residents aged > 20 years.

We compared the results with a previous study that reported the OPEs concentration of the indoor dust and students' hair in Beijing[65] and found that there were no significant differences between the OPE patterns in hair from residents of Beijing and Shandong (Weifang and Yantai). Moreover, there were no significant differences between the OPE patterns in hair from Weifang and Yantai and indoor dust from Beijing (paired samples *t*-test, $P > 0.05$), and significant differences were found between the OPE patterns in indoor dust in Beijing and in bark from Weifang and Yantai (paired samples *t*-test, $P < 0.05$). The source of OPEs in the hair of residents in Weifang and Yantai may not be the outdoor atmosphere. The indoor environment may be an important source.

There are also several limitations of the present study. First, the sample size of this study is relatively small, which may not be fully reflected in the exposure. Second, for human hair samples, only age and gender information were available, which limited the accurate assessments of the possibilities of analyzing the source of exposure. More information needs to be collected to understand this, such as lifestyle information.

CONCLUSIONS

The high OPE detection frequencies (> 90%) for the Weifang and Yantai tree bark samples strongly indicate that OPEs are ubiquitous in the environments of Weifang and Yantai. The OPE detection frequencies for the hair samples from Weifang and Yantai were also high, indicating that residents of Weifang and Yantai are exposed to OPEs. TCEP and TCPP made marked contributions to the $\sum_9$OPE concentrations in both bark and hair samples, and particular attention should be paid to human exposure to chlorinated OPEs as some have proven carcinogenic properties. The OPE concentrations were higher in the hair samples from people aged < 20 years than people > 20 years. Residents aged < 20 years may have been more exposed to OPEs than people aged > 20 years. The OPE patterns in the bark samples from Weifang and Yantai were not significantly different, indicating that OPE pollution in the outdoor atmosphere of the two cities follows similar patterns. No significant differences were found between the OPE profiles in the human hair samples from Weifang and Yantai, but the OPE profiles in the tree bark and human hair samples from the same area were significantly different. This indicates that the outdoor atmosphere was not the main source of OPEs in human hair samples from the two cities.

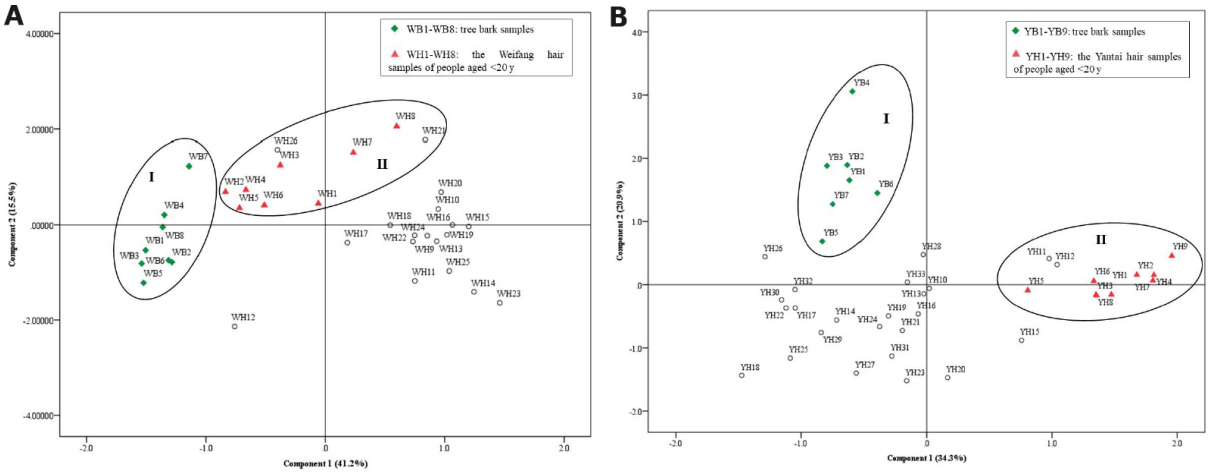

Figure 4. Principal component analysis score plot for OPEs concentrations in the tree bark and human hair samples collected in Weifang (A) and Yantai (B), Shandong Province, China. (A) Eight tree bark samples (WB1-WB8) and twenty-six hair samples (WH1-WH26) are included. WH1-WH8: The Weifang hair samples of people aged < 20 years. (B) Nine tree bark samples (YB1-YB9) and thirty-three hair samples (YH1-YH33) are included. YH1-YH9: The Yantai hair samples of people aged < 20 years. WB: the tree bark samples collected in Weifang; WH: the human hair samples collected in Weifang; YB: the tree bark samples collected in Yantai; YH: the human hair samples collected in Yantai.

DECLARATIONS

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Authors' contributions

Conceptualization, formal analysis, validation, writing - original draft, writing - review & editing, visualization, data curation: Wang SJ; Jin JX
Investigation, Validation: Liu AM; Li MH
Formal analysis: Yuan HD
Conceptualization, Validation: Hu JC; Wang Y
Writing - review & editing: Wang Y
Conceptualization, validation,writing - review& editing, visualization, supervision, project administration, funding acquisition: Jin J

Availability of data and materials

The data is available in the report. Additional data and information can be made available at request from individuals interested.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Consent was taken from participants.

Consent for publication

Not applicable

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