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Organochlorine pesticide concentrations in pooled serum of people in different age groups from five Chinese cities

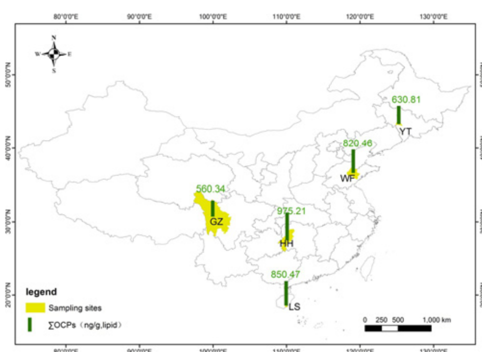
Qinghua Wang [a,c], Haodong Yuan [a], Jun Jin [a,b,*], Peng Li [a], Yulong Ma [a], Ying Wang [a]

[a] College of Life and Environmental Sciences, Minzu University of China, Beijing 100081, China
[b] Engineering Research Center of Food Environment and Public Health, Beijing 100081, China
[c] State Information Center, Beijing 100045, China

HIGHLIGHTS

- OCPs in serum differed among people of different age groups.
- The concentrations of HCB and endosulfan I were slightly higher in younger donors.
- Different OCP distributions were observed among five Chinese cities.

GRAPHICAL ABSTRACT



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ABSTRACT

Organochlorine pesticides (OCPs) were determined in human serum samples pooled by donor age (≥60, 50–59, 40–49, 30–39, and 20–29 y). The pooled samples were supplied by hospitals in five Chinese cities, Yitong (Jilin Province), Weifang (Shandong Province), Ganzi (Sichuan Province), Huaihua (Hunan Province), and Lingshui (Hainan Province). *p,p′*–Dichlorodiphenyldichloroethylene (DDE), hexachlorobenzene (HCB), β–hexachlorocyclohexane (HCH), and endosulfan I were the dominant OCPs in the serum samples. The total OCP concentrations and individual concentrations of *p,p′*–DDE, HCB, and β–HCH were relatively high compared with concentrations that have been found in other parts of the world. The total OCP, *p,p′*–DDE, and β–HCH concentrations decreased as age decreased but the HCB and endosulfan I concentrations were slightly higher in the samples from younger donors than in samples from older donors. HCB must be currently used and have new sources to people to cause this. Chinese government policies and the characteristics of the OCPs have led to OCP concentrations in serum decreasing with decreasing age. Different OCP usage patterns in different Chinese cities have led to the inhabitants of different cities having different OCP distributions in their serum.

1. Introduction

Organochlorine pesticides (OCPs) have been widely used to kill and control pests around the world. OCPs started to be used in the 1950s, and were used extensively to protect crops and prevent diseases between 1960 and 1970. Hexachlorobenzene (HCB), hexachlorocyclohexane

* Corresponding author at: College of Life and Environmental Sciences, Minzu University of China, Beijing 100081, China.
E-mail address: junjin3799@126.com (J. Jin).

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(HCH), and dichlorodiphenyltrichloroethane (DDT) were the most extensively used pesticides around the world between 1970 and 1980 (Shahawi et al., 2010). OCPs are persistent and lipophilic and can undergo long–range transport, so they have been found in various environmental media, including biota (Anderson et al., 2008; He et al., 2014; Kunisue et al., 2004; Lin et al., 2016; Pi et al., 2016). Evidence for a positive relationship between OCP concentrations in human serum and adverse effects has been gathered in recent years (Burns et al., 2012; Jain, 2014; Kezios et al., 2013). Since the 1970s, many countries have introduced controls on the production and use of OCPs. Japan implemented a law controlling the use of DDT in 1971 (NIP Japan, 2006). Sweden banned the production and use of DDT in 1975 and of HCB in 1980 (NIP Sweden, 2006). The Chinese government introduced controls on the production and use of DDT, HCH, HCB, and endosulfan in the 1980s, 1983, 2004, and 2014, respectively (NIP China, 2007). International concern led to nine OCPs (aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, HCB, mirex, and toxaphene) being included in the "dirty dozen" persistent organic pollutants required to be controlled under the terms of the Stockholm Convention in 2004. Six OCPs (chlordecone, α–HCH, β–HCH, lindane, pentachlorophenol and its salts and esters, and technical endosulfan and related isomers) were added to an annex of the Stockholm Convention in 2011.

Large amounts of OCPs have been used throughout China. China was traditionally an agricultural country, and large amounts of OCPs have been used to control malaria, termite and harmful pests. OCPs were produced and used extensively in China from the 1950s to the 1980s (Li et al., 1998). The total amounts of DDT and HCH produced in China were 0.4×10^6 and 4.9×10^6 t, respectively, accounting for 33% and 20%, respectively, of total global production (Zhang et al., 2002). More than a third of the total amounts of OCPs used in China were used in the southeastern provinces Zhejiang, Shanghai, Fujian, and Guangdong (Shahawi et al., 2010). Previous studies of OCPs in human serum in China were focused on developed areas in southeastern China (Bi et al., 2007; Lee et al., 2007), and OCP concentrations in human serum in under developed parts of China have rarely been determined. Most studies of OCPs in human serum in China have involved analyses of samples from small geographical areas (particularly developed cities, as mentioned above) so the overall trends in OCP concentrations in human serum in China are not clear. In the study presented here, we analyzed serum samples from people living in different parts of China with different climates and economies with the aim of providing datum for further research which can identify the overall trends in OCP concentrations in human serum in China. The OCP concentrations in serum samples from people living in five cities (shown in Fig. 1) were analyzed. The cities were, from north to south, Yitong (YT) in Jilin Province, Weifang (WF) in Shandong Province, Ganzi (GZ) in Sichuan Province, Huaihua (HH) in Hunan Province, and Lingshui (LS) in Hainan Province. All five cities are in under developed parts of China. Detailed information on the cities is available in Table S1 in the Supplementary Material (SM).

The objectives of this study were (1) to determine the OCP concentrations in serum from people living in five Chinese cities and to identify the differences in OCP exposure by age, (2) to assess the possible reasons for the OCP differences, and (3) to identify the dominant OCP compound the inhabitants of each city are exposed to.

2. Materials and methods

2.1. Chemicals and reagents

Aldrin, *cis*–chlordane, *trans*–chlordane, *p,p′*–dichlorodiphenyl dichloroethane (DDD), *o,p′*–DDD, *p,p′*–dichlorodiphenyldichloroethylene (DDE), *o,p′*–DDE, *p,p′*–DDT, *o,p′*–DDT, dieldrin, endosulfan I, endosulfan II, endrin, heptachlor, heptachlor epoxide A, heptachlor epoxide B, HCB, α–HCH, β–HCH, γ–HCH, δ–HCH, methoxychlor, isodrin, oxychlordane, and mirex standards were purchased from AccuStandard (New Haven, CT, USA). The internal standards $^{13}C_{12}$–labeled *p,p′*–DDE, $^{13}C_{10}$–labeled dieldrin, $^{13}C_6$–labeled HCB, $^{13}C_6$–labeled β–HCH, $^{13}C_6$–labeled γ–HCH, $^{13}C_{10}$–labeled mirex, and $^{13}C_{10}$–labeled *trans*–nonachlor were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). All organic solvents used were of pesticide analysis grade.

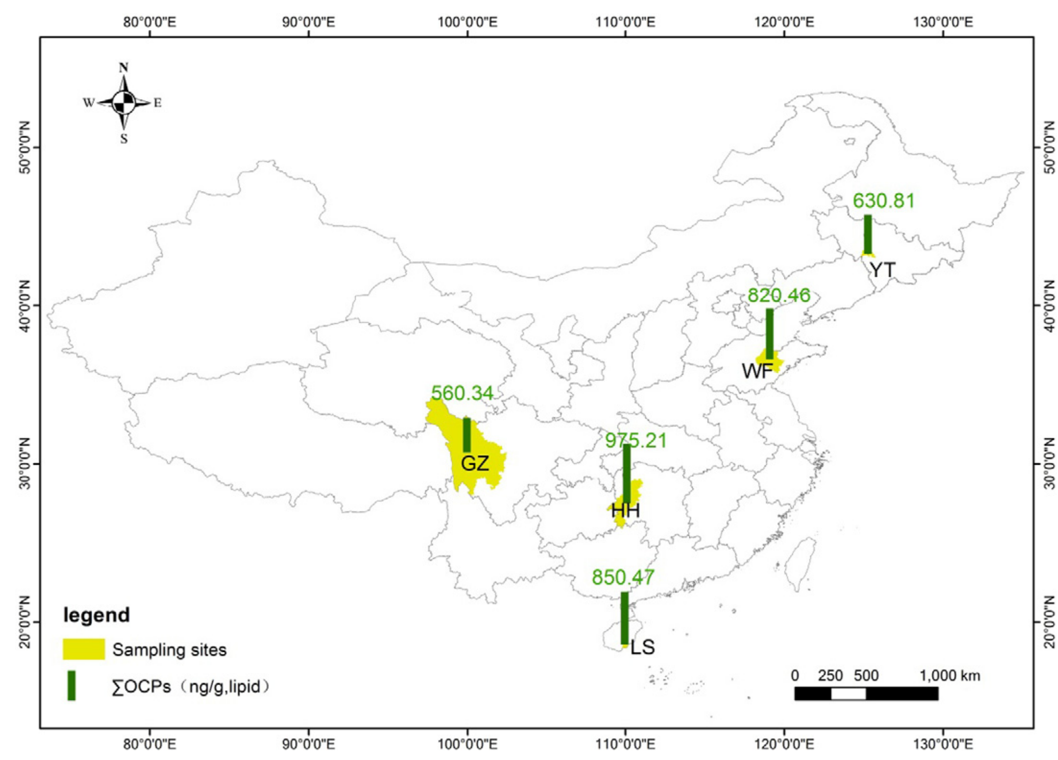

Fig. 1. The five cities the human serum samples were collected from and the total organochlorine pesticide concentrations (∑ OCPs, the sum of the HCB, α–, β–, and γ–HCH, *cis*–chlordane, *trans*–chlordane, endosulfan I and II, methoxy–DDT, *p,p′*–DDE, *p,p′*–DDD, and *o,p′*– and *p,p′*–DDT concentrations) found in the samples.