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7 authors, including:

Zhiqiang Nie
Chinese Research Academy of Environmental Sciences

Qingqi Die
Beijing Academy of Agriculture and Forestry Sciences

Jie He

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RESEARCH ARTICLE

Human health risk assessment of pesticide residues in market-sold vegetables and fish in a northern metropolis of China

Yanyan Fang · Zhiqiang Nie · Yanmei Yang · Qingqi Die · Feng Liu · Jie He · Qifei Huang

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Abstract With growing concerns about food safety and stricter national standards in China, attention has focused on vegetables and fish as they are an important part of the Chinese daily diet, and pesticide residues can accumulate in these foodstuffs. The local consumption habits of vegetables and fish were determined using questionnaires distributed in the major regions of the northern metropolis. Then, the samples of fruit-like vegetables, leafy and root vegetables, and five species of fish (freshwater and marine) were collected from supermarkets and traditional farmers' markets in the city. The concentrations and profiles of pesticide residues (hexachlorocyclohexane (HCH), dichlorodiphenyl trichloroethane (DDT), and endosulfan) in the samples were determined and compared. For the vegetables, the concentration ranges of ΣDDT, ΣHCH, and Σendosulfan were not detectable (ND) to 10.4 ng/g fresh weight (f.w.), ND to 58.8 ng/g f.w., and ND to 63.9 ng/g f.w., respectively. For the fish samples, the corresponding values were 0.77–25.0 ng/g f.w., 0.02–1.42 ng/g f.w., and 1.22–22.1 ng/g f.w., respectively. Only one celery sample exceeded the maximum residue limits (MRLs) of HCH residues set by Chinese regulations (GB2763-2014). The estimated daily intakes (EDIs) and hazard ratios (HRs) were calculated using data from the recently published Exposure Factors Handbook for the Chinese Population. The EDIs and HRs showed that the levels of organochlorine pesticide (OCP) residues in vegetables and fish in this area are safe.

Keywords Market-sold vegetables and fish · Questionnaires · Pesticide residues · Risk assessment

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Y. Fang · Y. Yang
School of River and Ocean Engineering, Chongqing Jiaotong University, Chongqing 400074, China

Y. Fang · Z. Nie (✉) · Q. Die · F. Liu · J. He · Q. Huang (✉)
State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
e-mail: niezq@craes.org.cn
e-mail: huangqf@vip.sina.com

Introduction

In recent years, because of increasing concern about food safety and public awareness of pesticide residues in food, research in China has focused on investigating pesticide residues in foodstuffs. Organochlorine pesticides (OCPs) are persistent and highly stable in the environment (Salem et al. 2009). Hexachlorocyclohexane (HCH), dichlorodiphenyl trichloroethane (DDT), and endosulfan are three OCPs that are widely used in agriculture (Tao et al. 2009). China began to produce and use HCHs and DDTs in 1950, and their production was not banned until 1983. Over the 30 years of production of these pesticides in China, 0.4 million tons of HCHs and 4.9 million tons of DDTs were produced, accounting for 20 and 33 % of the world production, respectively (Hua and Shan 1996; Zhang et al. 2009). DDTs and HCHs are frequently detected as predominant OCP pollutants in the environment (Feng et al. 2011; Zhang et al. 2013). Endosulfan is still in production and used in China, and in 2008, 5177 t were produced in China (China 2010; Wang et al. 2007).

Residues of HCH and DDT are frequently detected in vegetables, fruits, fishes, trees, wheat, and milk (Darko and Acquaah 2008; Gebremichael et al. 2013; Guler et al. 2010; Liu et al. 2010; Marco and Kishimba 2006; Mishra and Sharma 2011). Because endosulfan is extremely toxic to fish and aquatic invertebrates, its accumulation in vegetables and fish has caused public concern (Li et al. 2009). Many studies have investigated OCPs like HCH and DDT in vegetables and fish from different regions and cities. However, these studies have not looked at local dietary habits, and linkage analysis has been neglected (Wei et al. 2006; Xiao et al. 2006; Zhu et al. 2007). Studies have focused more on the concentrations and profiles of OCPs in vegetables and fish, rather than assessing the effect of these pollutants on human health (Barriada et al. 2010; Bempah et al. 2012). The limited research assessing the human health risk has been based on parameters from foreign countries. The recently published Exposure Factors Handbook of Chinese Population provides data that can be used to evaluate the human health risk more precisely. As Chinese national standards become stricter and control measures for pesticide residues in vegetables and fish increase, food safety research is important (MEP 2013).

As the capital of China and with a large population (about 20.7 million according to Beijing Municipal Statistics Bureau in 2013), food safety issues in Beijing are problematic. The aim of the present study was to provide data to improve food safety protocols in Beijing. Local dietary habits for vegetable and fish consumption were determined using a questionnaire. The profiles of HCH, DDT, and endosulfan residues in commonly consumed vegetables and fish were then determined in samples obtained from supermarkets and traditional farmers' markets. Then, the potential risk to human health because of the consumption of these vegetables and fish in Beijing, China was evaluated.

Materials and methods

Questionnaires on consumption of vegetables and fish

In May 2013, 350 diet questionnaires were delivered in the major communities in Beijing and via the Internet. Of the 350 questionnaires, 208 were completed correctly. The questionnaire asked what vegetables and fish people consumed, how frequently they consumed them, in what quantities they were consumed, where they obtained them from, how they cleaned them before cooking, and the age and gender of the respondent and how many family members they had.

Sampling, preparation, and chemical analysis

In August 2013, samples of 10 different vegetables and 5 species of fish were randomly acquired in supermarkets and traditional farmers' markets in Beijing, China (Fang et al. 2014). Vegetable species included fruit-like vegetables (tomato, cucumber, green beans, chilies, and *Benincasa hispida*), leafy vegetables (celery cabbage, *Spinacia oleracea*, and celery), and root vegetables (potato, radish). The fish collected included freshwater fish (cyprinoid, crucian, grass carp) and marine fish (hairtail, yellow croaker). The collected samples were put in clean plastic bags and brought to the laboratory for sample treatment as soon as possible.

Fish scales, innards, and inedible parts of the vegetables were removed first. To remove soil, the vegetable and fish samples were washed three times with tap water and then three times with deionized water. The samples were then dried with tissue paper. The edible parts of each sample were removed with a knife, pureed, and placed in sealed polyethylene bags and kept frozen at −20 °C until analysis.

Each sample was treated according to a modification of GB/T 5009.19-2008 in the Chinese standards. Vegetable and fish samples (5.00 g) were treated with 15 mL of n-hexane and 15 mL of dichloromethane in a centrifuge tube. After ultrasonic extraction for 30 min, the supernatant was separated by centrifugation. The extraction with 15 mL of n-hexane and 15 mL of dichloromethane was then repeated twice. The three supernatant fractions were combined and transferred to a 250-mL separatory funnel. Concentrated sulfuric acid (10:1) was added to the separatory funnel to sulfonate the samples. Then, sodium sulfate (2 %) was added with shaking for 2 min. After shaking, the separatory funnel was left for 30 min to allow the solution to separate into distinct layers. The n-hexane layer was transferred into a round bottom flask and concentrated to 1–2 mL on a rotary evaporator. Each sample was then passed through a florisil column (35 cm×10 mm) packed with 10 g of florisil between two layers of anhydrous sodium sulfate (2 cm). The extract was eluted with 60 mL of a mixture of dichloromethane:n-hexane (1:4, *v*/*v*). The eluent was evaporated to dryness, and the residue was dissolved in 2 mL of n-hexane and concentrated to 0.5 mL under a gentle nitrogen flow for further analysis.

Instrument analysis

Target compounds were firstly confirmed with gas chromatography-mass spectrometer (7890A, Agilent, Santa Clara, CA), and then, the concentrations of the target compounds were measured with HP 6890 gas chromatograph equipped with a ^{63}Ni electron capture detector and HP-5 capillary column (30 m×320 mm×0.25 μm, Agilent, Santa Clara, CA). Nitrogen was used as the carrier gas. The injector and detector temperatures were 250 and 315 °C, respectively. The gas chromatography column temperature was initially set at 60 °C for 2 min, ramped at 6 °C/min to 200 °C, 1 °C/min to 210 °C, 10 °C/min to 290 °C, and held at 290 °C for 10 min. An external standard curve was used for quantitative analysis.

Quality control and quality assurance

Laboratory quality assurance samples were used to evaluate the quality of the analytical data. Laboratory quality control included laboratory blanks. The results for the laboratory blanks indicated that samples were not contaminated during processing in the laboratory. The recoveries ranged from 79.3

to 119.3 %, and the detection limits ranged from 0.01 to 0.07 ng/g.

Risk assessment

Various international organizations have successively established a series of standards and protocols for evaluating the potential health risks from environmental pollutants in fish (USEPA 2013). A straightforward risk assessment involves comparing results with the levels set by laws and guidelines. However, this comparison is made without considering different eating habits and consumption rates. Thus, in this study, for a comprehensive health risk assessment, we first investigated the consumption habits for vegetables and fishes using a questionnaire in Beijing, China. From the results of this questionnaire, the most frequently consumed vegetables and fishes were chosen to calculate the estimated daily intakes (EDI) of HCH, DDT, and endosulfan. The EDI was calculated using Eq. (1). The hazard ratio (HR) for evaluating cancer risk was assessed by comparing the EDI with the benchmark concentration (BMC) (Solomon et al. 2000; Yohannes et al. 2014) using Eq. (2). The BMC for carcinogenic effects represents the exposure concentration at which the lifetime cancer risk is one in a million for lifetime exposure. A HR that is greater than 1 indicates that there is potential risk to human health (Dougherty et al. 2000).

$$\mathrm{EDI} = \frac{C \times \mathrm{DR}}{\mathrm{BW}}, \tag{1}$$

$$\mathrm{HR} = \frac{\mathrm{EDI}}{\mathrm{BMC}}. \tag{2}$$

In the above equations, C is the measured concentration of OCPs in vegetable or fish samples (ng/g fresh weight (f.w.)), DR is the average daily consumption rate of vegetable or fish (g/day) derived from the Exposure Factors Handbook, and BW is the average adult body weight, which was set at 60 kg (WHO 2010). The BMC values were derived from US EPA data (USEPA 2012).

Results and discussion

Questionnaires

The results of questionnaires concerning food origins showed nearly an equal preference for supermarkets (46.7 %) and farmers' markets (39.1 %), while farmers' stalls accounted for 14.2 %. The four most frequently consumed vegetables in Beijing, China were tomato, potato, celery, and Chinese cabbage (Fang et al. 2014). Additionally, the questionnaire results showed that 21.4 % did not soak vegetables to clean them before cooking, while soaking for 10 min was the most common method of cleaning (71.4 %) followed by 15 min or more (7.20 %). Soaking with fresh water or baking soda water (2 %) for 10 min before cooking is an effective method to remove pesticide residues in vegetables (Liang et al. 2013). In the present study population, 78.6 % of the respondents choose to soak vegetables for 10 min or longer before cooking. In addition to soaking, it is also recommended that vegetables are peeled before cooking (Clostrea et al. 2014).

The questionnaire results indicated that while residents are concerned about the pesticide residues in food, not all understand how to prepare vegetables to reduce the presence of pesticides. The questionnaire results indicate that the general public needs to be provided with more details about OCP residues and how to prepare various foodstuffs such as vegetables and fish. Earlier studies with farmers who use pesticides have suggested that training programs covering current government policies on vegetable safety and on the risks associated with highly toxic pesticides should be provided, especially to older and less educated vegetable farmers (Zhou and Jin 2009).

Concentrations and profiles of OCPs in vegetables

The concentration ranges of ΣDDT, ΣHCH, and Σendosulfan were not detectable (ND) to 10.4 ng/g f.w., ND to 58.8 ng/g f.w., and ND to 63.9 ng/g f.w., respectively. According to the latest maximum residue limits (MRLs) for food in China (GB2763-2014), the DDT and HCH concentrations in the vegetable samples were all below the MRLs, except for one celery sample. Compared with an earlier study, the DDT and HCH residues in the vegetable samples in the present study were much lower, and the concentrations of DDTs and HCHs were lower than those found in Jilin Province, China, and Indonesia (Gu 2011; Shoiful et al. 2013). For endosulfan, only provisional limits for four vegetables have been established in China (GB2763-2014). Compared with these limits, the detected endosulfan residues in the cucumber and potato samples in the present study were all within the MRLs.

HCHs and DDTs

Figures 1 and 2a, b give the concentrations and profiles for HCHs and DDTs in the fruit-like vegetables, leafy vegetables, and root vegetables. HCH and DDT residues were generally detected at levels lower than the national standards. One celery sample exceeded the HCHs limit by 1.18 times and also contained high levels of DDTs. This is probably a result of increased pesticide application because of the frequent insect pests encountered in celery production (Wei et al. 2006). According to the questionnaire data, 85.71 % people choose to consume celery in large quantities (5–10 kg) every

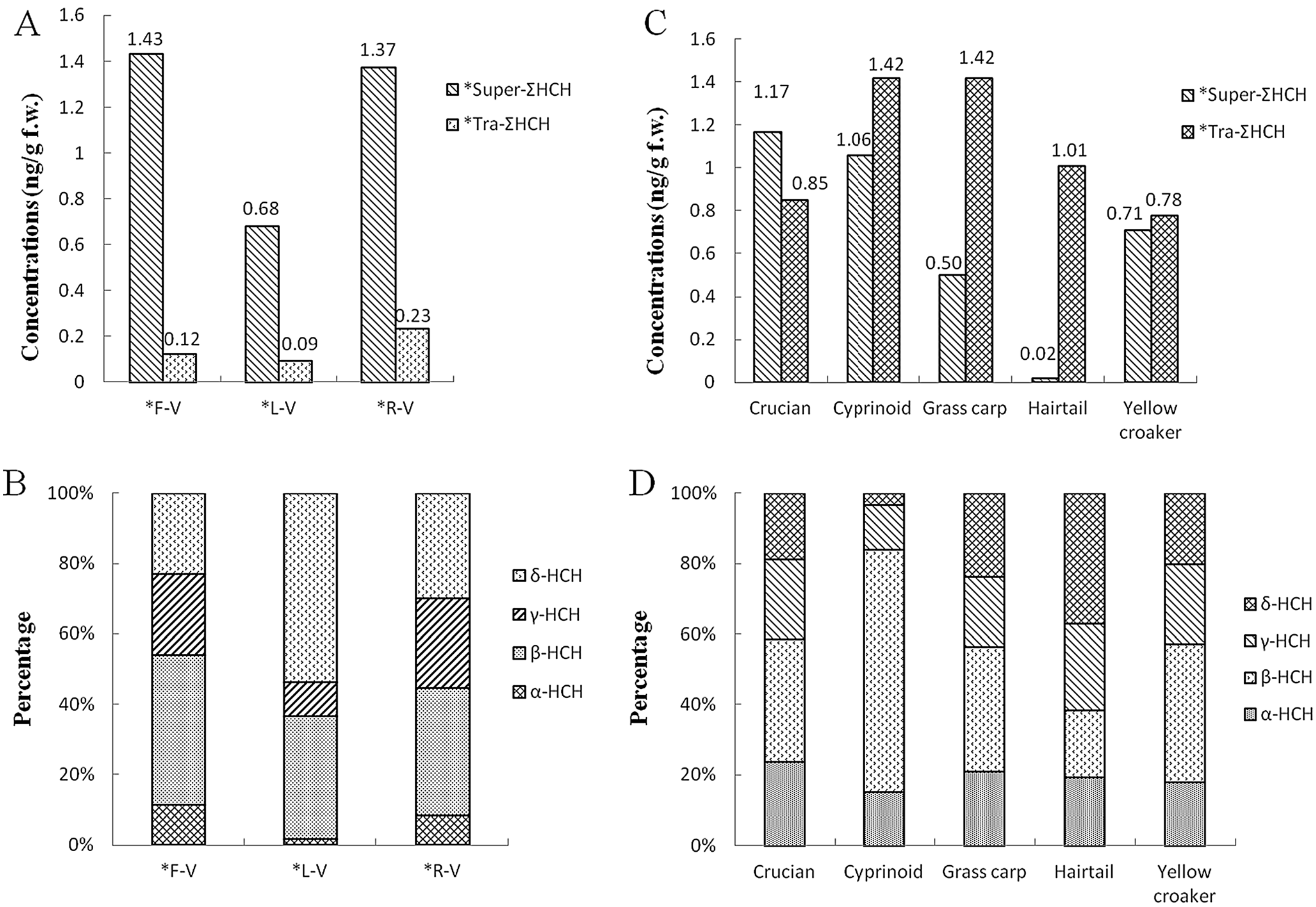

Fig. 1 HCH concentrations in different vegetables and fish (**a** and **c**) and the proportions of its four isomers (**b** and **d**). *F-V* fruit-like vegetables, *L-V* leafy vegetables, *R-V* root vegetables, *Super-ΣHCH* concentrations of ΣHCH in vegetable and fish samples from supermarkets, *Tra-ΣHCH* concentrations of ΣHCH in vegetable and fish sample from traditional farmers' markets

week. Because of the high pesticide residue levels, the consumption of large quantities of celery could be harmful to human health. As for the other three most frequently consumed vegetables, HCH and DDT residues showed quite low levels, ranging respectively from ND to 2.7 ng/g f.w. and ND to 4.02 ng/g f.w. The ΣHCH concentrations detected in the leafy vegetables (celery samples excluded) were obviously lower than those in the fruit-like vegetables and root vegetables, while celery samples showed higher concentrations of HCHs comparing with root and fruit vegetables. For ΣDDT, the concentrations detected in the leafy vegetables and fruit-like vegetables were higher than those in the root vegetables.

Among the isomers of HCH, the concentrations of β-HCH were slightly higher than those of the other isomers, which is consistent with the stability of β-HCH (Chen 2009; Sun et al. 2009). The values of α-HCH/γ-HCH were lower than 1, except for a Chinese cabbage sample (α-HCH/γ-HCH=1), which indicates transformation between homologues occurs. As for the isomers *p,p′*-DDE and *p,p′*-DDD, the leafy vegetables showed better absorption than that of the fruit-like vegetables and root vegetables. For the isomers *o,p′*-DDT and *p,p′*-DDT, the absorption capacity was in the order fruit-like vegetables > leafy vegetables > root vegetables.

When comparing the vegetables from supermarkets and farmers' markets, the supermarket products had higher HCH and DDT residue levels. The DDT and HCH residues detected in vegetables from Beijing were lower than those of residues detected in vegetables from Guiyang, Nanjing, Tai'an, and Jiangxi (China), similar to those detected in vegetables from Cambodia, and slightly higher than those in vegetables from North-Eastern Poland (celery excluded) (Hao and Jiang 2005; Hu et al. 2010; Luo et al. 2009; Qozowicka et al. 2012; Wang et al. 2011; Wang et al. 2004). The OCP residues showed a wide distribution trend in leafy vegetables, and compared with the 2008 results, the proportion of samples that exceeded MRLs was obviously reduced. This indicates that the banning of HCH and DDT has effectively reduced their contamination of vegetables. However, because of the stability and persistence of these pollutants, they need to be monitored and controlled for a long time (Chen et al. 2010).

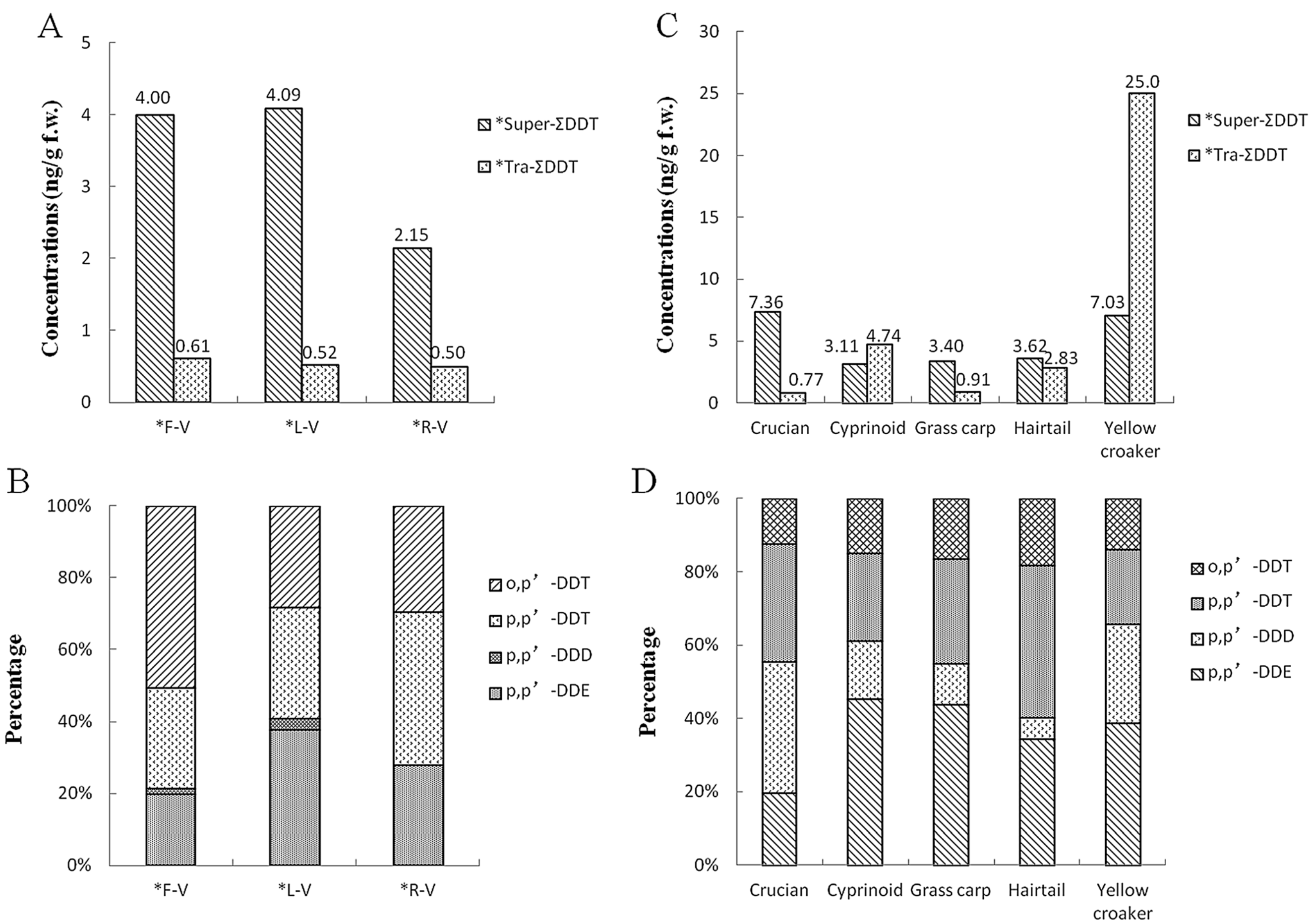

Fig. 2 DDT concentrations in different vegetables and fish (**a** and **c**) and the proportions of its four isomers (**b** and **d**). *F-V* fruit-like vegetables, *L-V* leafy vegetables, *R-V* root vegetables, *Super-ΣDDT* concentrations of ΣDDT in vegetable and fish samples from supermarkets, *Tra-ΣDDT* concentrations of ΣDDT in vegetable and fish samples from traditional farmers' markets

Endosulfan

Figure 3 shows the concentrations of endosulfan in vegetables and fish. The concentrations of Σendosulfan in vegetables ranged from ND to 63.9 ng/g f.w. The highest level of endosulfan was found in a green bean sample (63.9 ng/g f.w.). Compared with the concentrations of endosulfan in Hyderabad, Pakistan, and North-Eastern Poland, the vegetables in the present study had higher endosulfan concentrations. This indicates that more specific control and management of endosulfan residues in the environment are required in China (Latif et al. 2011; Qozowicka et al. 2012). Using EU pesticide residue MRLs, green bean (63.9 ng/g f.w.) from a supermarket, and radish (58 ng/g f.w.) from a traditional farmers' market exceeded the MRLs (50 ng/g f.w.). This is because endosulfans are still used in agriculture. Industrial-grade endosulfan consists of two isomers (α and β), with a composition of 70 % α- and 30 % β-endosulfan (Wan et al. 2005). Where endosulfan residues were detected in vegetables, the α-endosulfan isomer was present in higher concentrations than that of the β-endosulfan isomer. The absorption capacities of the different types of vegetables for α-endosulfan were in the order fruit-like vegetables > root vegetables > leafy vegetables. For β-endosulfan, the fruit-like vegetables and leafy vegetables showed better absorption capacity than that of the root vegetables. Vegetables from traditional farmers' markets contained higher concentrations of Σendosulfan than those from supermarkets, and this result can be related to the relatively uncontrolled management of traditional farmers' markets.

Concentrations and profiles of DDTs, HCHs, and endosulfans in fish

The concentration ranges of ΣDDT, ΣHCH, and Σendosulfan were ND to 25 ng/g f.w., ND to 1.42 ng/g f.w., ND to 22.1 ng/g f.w. Using the latest Chinese OCP MRLs (GB2763-2014), the concentrations of DDTs and HCHs in fish were all below the limits. For endosulfan, Chinese standards are only available for poultry (provisional limits). The average lip weight of fishes are tested, and the results are respectively 5.50 % for

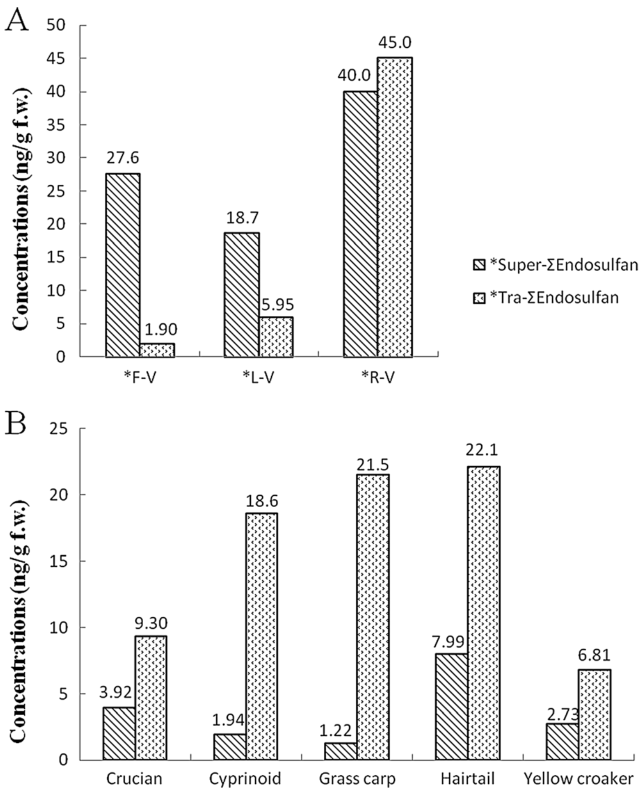

Fig. 3 The concentrations of endosulfan in different vegetables (**a**) and fish (**b**). *F-V* fruit-like vegetables, *L-V* leafy vegetables, *R-V* root vegetables; *Super-Σendosulfan* concentrations of Σendosulfan in vegetable and fish samples from supermarkets, *Tra-Σendosulfan* concentrations of Σendosulfan in vegetable and fish samples from traditional farmers' markets

crucian, 6.09 % for cyprinoid, 5.20 % for grass carp, 4.90 % for hairtail, and 6.80 % for yellow croaker.

Figures 1 and 2c, d give the concentrations and profiles of HCHs and DDTs in the fish samples. The HCH concentrations showed little variability in freshwater fishes (0.02–1.42 ng/g f.w.). The concentrations were generally lower than those from earlier studies of fish from cities in the south and east of China, the South China Sea, and the Qiantang River in China (Jiang et al. 2005; Wan et al. 2013; Zeng 2010; Zhou et al. 2008). The present results were similar to those for fish from a Rift Valley lake (Lake Ziway) in Ethiopia (Yohannes et al. 2014). Compared with the levels in fish from Asia, Oceania, and Korea and mollusks from the coast of the Bohai Sea, the HCH residues in fish from markets in Beijing were low (Kannan et al. 1995; Yang et al. 2004; Yim et al. 2005). In the present study, fishes from traditional famers' markets (crucian excluded) have higher HCH concentrations comparing with supermarkets. While yellow croaker fish from traditional farmers' markets showed higher DDT concentrations than that of supermarket fish, and for crucian, grass carp, and hairtail samples, supermarkets have higher DDT concentrations comparing with traditional famers' markets. The DDT concentrations were much lower than those detected in fish from the Republic of Bénin (Pazou et al. 2006). Compared with results from Indonesia (12–1100 ng/g lipid wt.), the DDT residues in market fishes were quite low, which suggests that the ban on DDTs has resulted in the good control of these residues (Sudaryanto et al. 2007).

The highest level of endosulfan was found in a hairtail sample (Fig. 3). The supermarket samples contained lower concentrations of endosulfan than that of the samples from traditional farmers' markets. The concentrations were lower than those detected in fish from the Republic of Bénin (Pazou et al. 2006). Compared with marine fish from Coimbatore, India (5–23 ng/g f.w.), the fish from markets in Beijing contained slightly higher concentrations of endosulfan (Muralidharan et al. 2009). The pesticide residues in fish are mainly from water pollution and bio-concentration (Wang 2013).

For stricter controls on endosulfan's production, circulation, usage, import, and export, more detailed and comprehensive MRLs for endosulfan residues in vegetables and fish are required. The presence of endosulfan residues in vegetables and fish is of concern, and measures should be taken to increase the quality of vegetables and fish sold in markets.

Table 1 The EDI values of HCH, DDT, and endosulfan (ng/kg bw/day) and the HR values of HCH and DDT

Category	EDI-HCH		EDI-DDT		EDI-endosulfan		HR-HCH ($\times10^{-4}$)		HR-DDT ($\times10^{-4}$)	
	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female
F-V	0.69–4.12	0.80–4.83	1.13–8.87	1.32–10.4	6.46–51.0	7.56–58.1	0.01–0.04	0.007–0.04	0.03–0.26	0.03–0.31
L-V	0.47–45.1	0.55–52.7	1.22–8.20	1.43–9.60	13.3–21.5	15.5–25.2	0.004–0.41	0.005–0.48	0.04–0.24	0.04–0.28
R-V	1.14–3.00	1.33–3.51	1.45–5.40	1.69–6.32	44.7–59.7	52.3–69.9	0.01–0.03	0.01–0.03	0.04–0.16	0.05–0.18
F-F	0.89–1.15	1.04–1.34	2.00–34.4	2.34–40.3	6.12–10.5	7.16–12.3	0.008–0.01	0.009–0.012	0.06–0.11	0.07–0.13
M-F	0.48–46.8	0.56–54.7	2.99–17.2	3.50–20.2	4.42–36.9	5.17–43.2	0.004–0.43	0.005–0.49	0.09–0.43	0.10–0.51

HCH γ-HCH, *F-V* fruit-like vegetables, *L-V* leafy vegetables, *R-V* root vegetables, *F-F* freshwater fish, *M-F* marine fish

Health risk assessment

The consumption of vegetables and fish are two major routes of human exposure to organic contaminants. To understand the concentration levels better, the concentrations of HCHs, DDTs, and endosulfans were evaluated against international limits. The EDI was calculated and compared with the acceptable daily intake (ADI) recommended by the Food and Agriculture Organization and the World Health Organization (FAO/WHO) Joint Meeting on Pesticide Residues (WHO 2010). The EDIs of HCH, DDT, and endosulfan are expressed in nanograms per kilogram of body weight per day (ng/kg bw/day) from the consumption of vegetables and fishes (Table 1). The HR values for HCH and DDT were calculated as well. The EDIs of HCHs, DDTs, and endosulfans were all far below the ADI (5000 ng/kg bw/day for HCH, 10,000 ng/kg bw/day for DDT, and 570 ng/kg bw/day for endosulfan), indicating that the consumption of vegetables and fish does not currently pose a human health risk.

As shown in Table 1, the HRs for HCH ranged from 0.04 to 0.41 for vegetables, and the highest value was detected in a celery sample. While for DDT, the HRs ranged from 0.03 to 0.31 for vegetables, and the average was higher than that of the HRs of HCH. The fish HRs were 0.004–0.49 for HCH and 0.06–1.19 for DDT. HRs for females are lower than those for males because of their lower caloric intake and lighter weight. Marine fish showed higher HR values than that of fresh water fish. The HRs calculated were lower than that of a target risk of $>1\times10^{-4}$, which is considered unacceptable. These results indicate the DDT and γ-HCH levels in vegetables and fish from supermarkets and traditional farmers' markets in Beijing are safe and will not cause cancer through consumption. The HRs calculated for vegetables and fishes in Beijing markets are much lower than those for lake fishes from Ethiopia (Yohannes et al. 2014).

Conclusion

Although production and use of DDTs and HCHs have been banned for more than 30 years in China, these pesticides are still widely detected in vegetables and fish. Their concentrations are typically much lower than that of the national standards. In the present study, two samples of vegetables and fish had endosulfan concentrations that exceeded the MRLs (European Union MRLs), and compared with the concentrations of DDTs and HCHs, the concentrations of endosulfan were much higher. This suggests that banning DDT and HCH has been effective in reducing the levels of these pesticides in the environment. The results show that DDT, HCH, and endosulfan residues in vegetables and fish are not present at levels that are harmful to human health. However, these chemicals are persistent and can accumulate in the human body. China has passed new restrictions (effective March 26th of 2014) on endosulfan usage, with its only allowed use in the prevention of cotton bollworm and tobacco budworm. This measure will contribute greatly to the control of endosulfan input to the environment. However, the effective management of the endosulfan residues already present in the environment needs to be implemented. More restrictions on endosulfan use and new control measures based on food research results are required. Pesticide detection programs need to be strengthened to regularly assess the human health risk from potentially contaminated foodstuffs.

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