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Persistent organic pollutants in food items collected in Hong Kong

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ABSTRACT

This study aims to investigate levels of POPs in meat, edible oils, nuts, milk and wine collected from Hong Kong. Naphthalene, pp-DDE, beta-, gamma-HCH and PBDE 47 were detected in most of the food items. Goose liver accumulated the highest PAHs (47.9 ng g^{-1} wet wt), DDTs (25.6), HCHs (13.0), PCBs (4.17), PBDEs (468 pg g^{-1} wet wt) among all the selected food. Meat and nut groups had significant ($p < 0.01$ or 0.05) correlations between lipid contents and concentrations of PAHs (meat: $r = 0.878$), HCHs (meat: $r = 0.753$), DDTs (meat: $r = 0.937$; nuts: $r = 0.968$) and PCBs (meat: $r = 0.832$; nut: $r = 0.946$). The concentrations of DDTs, HCHs and PCBs in vegetable oil were lower, but HCHs in fish oil were higher, when compared with other countries. The concentrations of PAHs, DDTs, PCBs and PBDEs in food tested in the present study were all below various safety guidelines.

1. Introduction

Hong Kong, located in Pearl River Delta, is one of the most prosperous regions in China, with serious environmental problems due to the rapid industrial and economic development in the past decades. Man-made chemicals, such as OCPs, PCBs, PAHs and PBDEs have been the major environmental issues in the Pearl River Delta region (Poon et al., 2005; Cheung et al., 2008). People are mainly exposed to these chemicals through inhalation, dermal contact and ingestion via diet, and more than 90% of human exposure to OCPs, PCBs and PAHs have been through dietary intake (Safe, 1998).

Due to the decline of agricultural activities in Hong Kong, the major food supply comes from mainland China, especially the Pearl River Delta (Cheung et al., 2007). However, the environmental quality around the Pearl River Delta has been deteriorating in the past decades. Our previous study showed that the levels of DDTs in fish samples collected from fish ponds around the Pearl River Delta ranged from 1.5 to 62 ng g^{-1}, with 35% of fish samples exceeding the limit of 14.4 ng g^{-1} for human consumption set by USEPA (Kong et al., 2005). Moreover, serious contamination in preserved fruits (HCB: 118 μg kg^{-1}), preserved and salted eggs (DDT and metabolites: 1010 μg kg^{-1}) has been reported in Hong Kong (Ip, 1990). The problem we now face in our densely populated city is the long-term chronic exposure to rather high levels of POPs found in food, such as fish and vegetables (Cheung et al., 2007). Our recent studies have shown that there are significant correlations ($p < 0.05$) between fish diet intake and body loadings of DDTs and PCBs (Wong et al., 2002; Qin et al., 2010), indicating that food ingestion is an important pathway for human exposure to environmental pollutants. Chronic exposure to POPs can be harmful to human health. It has been suspected that DDTs can cause breast cancer (Snedeker, 2001), and that DDTs, HCHs and PCBs are harmful to the human reproductive system (Toft et al., 2004). Contaminants, such as PAHs, PCBs, PBDEs and PCDDs have been detected in fish, eggs, meat and rice in a number of countries including USA, Germany, Belgium and Spain (Furst et al., 1990; Bocio et al., 2003; Schecter et al., 2006; Zuccato et al., 2008). Our previous study also reported rather high concentrations of PAHs, DDTs, PCBs, PBDEs and PCDD/Fs in fish, meat, egg, vegetable and rice sold in Hong Kong markets (Kong et al., 2005; Cheung et al., 2007, 2008; Tsang et al., 2009). However, there is a lack of information about these pollutant levels in edible oil, animal livers, nuts and milk, which contain relatively higher lipid contents locally. These are also popular food items sold in Hong Kong markets besides the staple food (fish, meat, egg, vegetable and rice) in which POPs concentrations have been analyzed in our previous studies (Cheung et al., 2007; Tsang et al., 2009). Therefore, in order to

Abbreviations: GC–MS, Gas Chromatography–Mass Spectrometer; OCPs, Organic chlorinated pesticides; PAHs, Polycyclic aromatic hydrocarbons; PBDEs, Polybrominated diphenyl ethers; PCBs, Polychlorinated biphenyls; POPs, Persistent organic pollutants; DDTs, Dichlorodiphenyltrichloroethane; HCHs, Hexachlorocyclohexane; HCB, Hexachlorobenzene; PCDDs, Polychlorinated dobenzo-p-dioxins.

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obtain a fuller picture on POPs contaminations in different food items in Hong Kong, the major objectives of the present work were: (1) to investigate the concentrations of PAHs, OCPs, PCBs and PBDEs in edible oil, nuts, meat, wine and milk collected in Hong Kong; and (2) to conduct the health risk assessments of edible oil, nuts, meat, wine and milk consumption.

2. Materials and methods

2.1. Sample collection

Five groups of selected products (milk, meat, wine, edible oil, nut) were purchased from supermarkets (Wellcome, Park'n Shop and Taste) and nine wet markets in Hong Kong Island, Kowloon and New Territories, Hong Kong during August–October 2007. Food samples were randomly purchased in three different local markets from each of the above regions. Since meat, wine and nuts are usually produced and sold by various markets, their origins are not always readily known. Therefore, three composite samples of each food item were used for chemical analyses whereby each composite consisted of ten individual samples. However, in the case of edible oil and milk (coconut milk and 'Carnation' evaporated milk), they are usually sold under similar brand names within the sampling regions, and, therefore, only three composite samples of each food item were used for chemical analyses in which each composite was made up of one individual sample only. The samples were homogenized immediately after collection and stored at −20 °C until further treatment.

2.2. Chemical analyses

Extraction of OCPs, PCBs, PAHs and PBDEs from different food samples was performed according to the EPA Standard Method 3540C (USEPA, 1996a). Food samples were separated and freeze-dried. The moisture contents of different food samples were determined according to weight loss before and after freeze-drying. Freeze-dried food samples were homogenized into powder and stored in desiccators prior to the extraction.

About 2 g (7 g for analysis of PBDEs) of sample was transferred into a Soxhlet apparatus and extracted by 80 mL acetone:dichloromethane mixture (1:1) in a 70 °C water bath for 18 h. As for oil, approximately 0.2 g oil was dissolved in 10 mL hexane in a seperatory funnel and then liquid–liquid extraction was performed twice with 50 mL DMSO (Takada et al., 2001). One fifth of the concentrated solution was quantified for fat content determination (Hara and Radin, 1978), and the remaining 4 mL was diluted to 5 mL. Gel-permeation column (EPA Standard Method 3640A) (USEPA, 1994) and florisil column (EPA Standard Method 3620B) (USEPA, 1996b) were applied for the cleanup of samples. The solution was concentrated to 0.2 mL before the analyses of PAHs, PCBs, OCPs and PBDEs using Gas Chromatography–Mass Spectrometer (GC–MS) (GC: 6890 N, MS: 5973, Agilent Technologies, USA). Limit of detection (LOD) of PCBs was 0.6 ng g^{-1} wet wt, 0.9 for OCPs, 0.01–0.18 for PBDEs, and 1–5 for PAHs. The recoveries of PAHs ranged from 83% to 90%, PCBs ranged from 94% to 99%, HCHs and DDTs ranged from 90% to 96%, and PBDEs ranged from 75% to 109% for solvent spike samples.

2.3. Statistical analyses

All the data were presented as median and analyzed using SPSS 16.0 software. Values below the LOD were set to zero when determining the concentration. Pearson's correlation method was used to assess correlation between POPs and lipid content.

3. Results and discussion

3.1. PAHs in food items

Table 1 shows the concentrations of 16 priority PAHs detected in all the food samples. The highest PAHs were found in the meat group, such as goose liver (47.9 ng g^{-1} wet wt), chicken skin (44.7) and chicken breast (41.2). Vegetable oils and nuts contained similar PAHs concentrations, but were lower than the meat group, except for fish oil (41.4). Milk, such as coconut milk and Carnation evaporated milk contained comparatively lower PAHs than meat, edible oil and nuts groups, but were higher than the wine group, (white wine: 7.01, red wine: 7.36) which contained the lowest lipid content. Naphthalene was the most dominant PAHs detected in all the selected food items in the present study. This result was in line with our previous study that naphthalene was the most dominant congener detected in fish collected in Hong Kong (Cheung et al., 2007). Fortunately, congeners, such as benzo[*b* + *k*]fluoranthene, benzo[*a*]pyrene, dibenz[*a*,*h*]anthracene, indeno-[1,2,3-*c*,*d*]pyrene and benzo[*g*,*h*,*i*]perylene, were below the limits of detection, a common phenomena for most food items that are not contaminated (Bordajandi et al., 2004).

As shown in Fig. S1 (Supporting information), Low Molecular Weight (LMW) PAHs (such as, naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene and pyrene) were the dominant congeners detected in milk, edible oil, meat, nut and wine groups, while High Molecular Weight (HMW) PAHs (such as, benzo(*b* + *k*)fluoranthene, benzo(*a*)pyrene, indeno[1,2,3-*c*,*d*]pyrene, dibenz[*a*,*h*]anthracene, and benzo[*g*,*h*,*i*]-perylene) were not detected. Naphthalene, fluorine, phenanthrene and fluoranthene were the dominant congeners detected in these samples. Naphthalene was the most dominant compound detected in milk (31.4%), meat (27.2%), wine (49.3%), oil (34.3%) and nuts (41.6%), while fluorine was mostly detected in milk (19.6%), meat (26.3%), wine (14.6%), oil (18.1%) and nuts (11.0%). With regards to phenanthrene, it was also one of the most dominant congeners in the meat group (22.2%), followed by oil (15.6%), milk (8.51%), and nuts (5.92%). A previous study also revealed that phenanthrene was the most predominant PAH compound in meat and fish collected in Spain (Falco et al., 2003). Higher percentages of fluoranthene were detected in the wine (27.8%), oil (13.26%) and nut (12.55%) groups. In general, LMW PAHs were detected in all the food items collected in Hong Kong. PAHs from a petrogenic source (hydrocarbon compounds associated with petroleum) were characterized by the abundance of LMW PAHs with depletion of HMW PAHs, whereas pyrogenic sources (hydrocarbon compounds associated with the combustion of petroleum, wood, coal including creosote, coal tar) were enriched with HMW PAHs (Zakaria et al., 2002). The prevalence of LMW PAHs in the profile indicated that the compounds were mainly derived from petrogenic pollution (Sauer et al., 1993).

3.2. OCPs in food items

Table 2 shows the concentrations of OCPs in different selected food items. pp-DDE was the most dominant congener detected in all food items. Meat contained higher DDTs in general, with goose liver having the highest concentration of DDTs (25.6 ng g^{-1} wet wt) among all the selected food items, followed by the nuts group whereby walnut kernel accumulated the highest DDTs (19.5). The concentration of DDTs in edible oil was lower than the nuts group, except for fish oil (12.0), which was higher than in vegetable oils. The ratio of pp-DDE/DDTs is a good indicator of recent input of DDT with a ratio lower than 0.6 indicating a recent input (Aguilar, 1984). Our result shows that the ratios of pp-DDE/DDTs of chicken