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# Temporal trends in daily dietary intakes of DDTs and HCHs in urban populations from Beijing and Shenyang, China



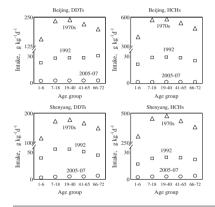
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#### HIGHLIGHTS

- ► Three representative time periods were selected.
- ► Daily dietary intakes of DDTs and HCHs by target populations decreased temporally.
- Relative contributions of foods to the intakes of DDTs and HCHs varied with time.
- ► DDT and HCH intakes in 2005–2007 would not exert adverse effect on human health

#### G R A P H I C A L A B S T R A C T

Daily dietary intakes of DDTs and HCHs by general population show temporally decreasing trend in Northern China.



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#### ABSTRACT

The temporal trends in daily dietary exposure of the populations in Beijing and Shenyang, China to DDTs and HCHs through consumption of various food categories and the associated health risks were investigated in this work. The estimated dietary intakes of DDTs and HCHs in 2005/2007 were almost one and two orders of magnitude lower than those in 1992 and in the 1970s, respectively, revealing the historical decrease of DDT and HCH in Chinese foods after their applications were banned. The relative contributions of different foodstuffs to the overall intakes of DDTs and HCHs in the 1970s and in 1992 were very different from those in 2005/2007, mainly due to the remarkable concentration variance in foodstuffs and changes in diet habit of population. According to the hazard guidelines, it is concluded that intakes of these two pesticides in 2005/2007 may not exert adverse effects on human health.

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#### 1. Introduction

Historically, technical dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) were the most important organochlorine pesticides (OCPs) world widely (Jones and de Voogt,

1999). These chemicals came into usage for agricultural practice in the early 1950s in China (Wong et al., 2005). Although these two insecticides benefited agricultural production, they also brought some side effects (Carvalho, 2006). Since DDT and HCH are environmentally persistent, long-range transportable and lipophilic (Lohmann et al., 2007), these chemicals widely distributed in air, water, soil, foodstuffs, as well as in human body in China (Gong et al., 2004). They were reported to be the causing agents for cancer

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(von Muhlendahl, 1999), dysfunction of the reproductive system (Safe, 2004), endocrine-disturbing (Tilson, 1998) and slow development of the fetus (Spyker, 1975).

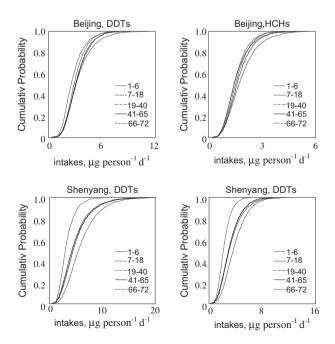
Production and use of DDT and HCH was banned in many developed countries in the 1970s (Li, 1999; Wong et al., 2005), while the amount of their usage in China reached a maximum in this time period, and their production and application were not officially prohibited until 1983 (Nakata et al., 2002). As a whole, the residue levels of DDT and HCH in various media gradually decreased in China since their application was banned (Li et al., 1998). However, their concentrations in foodstuffs were still relatively high in 1990 (Chen and Gao, 1993). Recent information on the residue levels of DDT and HCH in Chinese foodstuffs is scarce and fragmentary. As a consequence we know little about the current time trend in dietary intakes of these pesticides and the resulting health risks to the general public in urban areas in China (Hura et al., 1999).

The three time periods (i.e., of the 1970s, 1992 and 2005/2007) represented respectively, three different historical stages for the use of DDT and HCH in China, namely: the 'rush period' (in the 1970s); the period 10 years after the ban (in the 1990s) and the period 20 years after the ban (in the 2000s). In the present study, such representative time periods were selected to investigate the dynamic changes in daily dietary intakes of DDT and HCH and the resulting health risks to the residents in Beijing and Shenyang, China. Beijing and Shenyang were chosen mainly due to the fact that some work has been done to probe the exposure of the residents in southern China to OCPs (e.g., DDT and HCH), but such information for northern China is very limited. Additionally, there are considerable differences in the dietary habit of residents and the historical usage of OCPs in northern and southern China.

#### 2. Materials and methods

#### 2.1. Population

The age of the target populations ranges from 1 to 72 years old. All people were divided into five age groups, and they were 1-6, 7-18, 19-40, 41-65 and 66-72 years old, respectively. Consumption profiles of the representative foodstuffs and the body weight of



**Fig. 1.** Probability distributions of the daily dietary intakes of DDTs and HCHs by various age groups of the populations in Beijing and Shenyang in 2005–2007.

the target populations in each age group were taken from the Chinese total diet studies in 1992 and 2002 (Ge, 1992; Liu et al., 1995; Wang, 2005; Zhai and Yang, 2006). The detailed food consumption, body weight of the target populations are presented in Tables S1–S12 in the Supporting material (SM).

#### 2.2. Exposure probability analysis

Mont Carlo simulation (5000 runs) was used to estimate the daily dietary intakes of DDT and HCH by the residents from Beijing and Shenyang whose age ranged from 1 to 72 years old and to evaluate their health risk probability. Parameters used for simulation included food consumption, concentrations of DDTs and HCHs in foods and human body weight. Concentrations of DDTs and HCHs in foods in 2005/2007 have been reported in our previous studies (Tao et al., 2008; Yu et al., 2009), and their historical data (1992, 1970s) were taken from Zhao et al. (2003). T test was used to compare mean of the residue levels of DDTs and HCHs in foods, and the estimated daily dietary intakes of DDT and HCH between Beijing and Shenyang. In all cases, significance level of p < 0.05 was considered. SPSS 13.0 was used for data analysis.

#### 3. Results and discussion

#### 3.1. Daily dietary intakes of DDTs and HCHs in 2005/2007

The cumulative probability distributions of the estimated daily dietary intakes of DDTs and HCHs by each age group of populations in Beijing and Shenyang in 2005/2007 are shown in Fig. 1 (with detailed data presented in Tables S13 in the SM). Table 1 shows the measured means and standard deviations in  $\mu g \, kg^{-1} \, d^{-1}$ , which are compared with those reported for 2002 and 1970s (Zhao et al., 2003). Difference among years and between two cities were all significant (p < 0.05).

It was evident that the exposure doses of DDTs and HCHs in all age groups in Shenvang were slightly higher than those in Beijing. due to the fact that the vegetables and cereals consumed by the residents in Shenvang had much higher concentrations of DDTs and HCHs relative to those in Beijing (Fig. 1). Children (1-6 years old) were exposed to the lowest amount of DDTs and HCHs because they ingested much smaller amount of foods in contrast to other age groups (S1 and S4 in the SM). No statistical difference in ingestion doses of DDTs and HCHs was found among other age groups (p > 0.05). It was reported that Intake of HCHs and DDTs by residents in Shanghai in 2002 were 2.16 and 0.13 μg person<sup>-1</sup> d<sup>-1</sup> (Nakata et al., 2002), which were comparable to HCHs intake, but higher than DDTs intake by residents in Beijing and Shangyang. However, DDT and HCH intakes were all much higher than those reported for Sweden (0.523 and 0.081  $\mu g$  person<sup>-1</sup> d<sup>-1</sup>) (Darnerud et al., 2006) and for Japan (1.10 and 0.018  $\mu$ g person<sup>-1</sup> d<sup>-1</sup>) (Sudaryanto et al., 2007) in the same time period, suggesting that the people in Beijing and Shenyang ingested much more DDTs and HCHs than those in some developed countries in 2005/2007. Different food categories contributed differently to the DDT and HCH exposure to human beings. As for Beijing residents, the greatest contribution to the overall dietary intake of DDTs and HCHs was from milk (DDTs, HCHs: 17.1-38.8%, 25.6-50.7%), followed by cooking oil (13.9–19.5%, 22.5–35.9%). Vegetables and cereals contributed 11.7-18.4% and 6.62-9.70% of DDT intake, and 2.86-5.07% and 6.20-10.4% of HCH intake, respectively. Fish and meat contributed 6.77-12.5% and 12.1-16.3% of DDT intake, and 3.44-5.74% and 6.11-9.16% of HCH intake, respectively. Egg and fruit were the smallest contributors to DDT (3.76-4.20%) and HCH (2.19-2.44%) intake. For the case in Shenyang, cooking oil (22.8-23.5%, 29.9-31.9%), cereals (24.4-26.4%, 29.7-33.6%) and vegetables (29.6–36.4%, 10.3–12.8%) were most important contributors

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