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Levels, distributions, and source identification of organochlorine pesticides in the topsoils in Northeastern China

WANG Xu[1], REN Nanqi[1], QI Hong[1], MA Wanli[1], LI Yifan[1,2,*]

1. International Joint Research Center for Persistent Toxic Substances (IJRC-PTS), State Key Laboratory of Urban Water Resource and Environment (SKLUWRE), Harbin Institute of Technology, Harbin 150090, China. E-mail: ijrc_pts_paper@yahoo.com
2. Science and Technology Branch, Environment Canada, Toronto, Ontario M3H 5T4, Canada

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Abstract

Seventeen topsoil samples (9 urban, 4 suburban, 3 rural and 1 background) were collected in/around Harbin, a typical city in northeast of China, to measure concentration levels of organocholrine pesticides (OCPs) in topsoil of Northeastern China in 2006. Hexachlorohexanes (HCH), dichlorodiphenyltrichloroethane (DDT), and hexachlorobenzene (HCB) were detected in soil samples with mean concentrations (in pg/g dry weight (dw)) of 7120, 5425, and 1039, respectively. The mean concentrations for other OCPs were very low, 4.8 pg/g dw for chlordane and 3.3 pg/g dw for endosulfan. Source identification analysis reveals that all OCPs found in soil samples were due to historical use of these chemicals or from other source regions through long- and short-range atmospheric transport. DDT was mainly used in the rural sites, whereas the sources of HCB, chlordane and endosulfan were mainly in the urban area. HCH was found almost equally in both urban and rural area. Soil concentrations of all detected OCPs, except HCHs, in and around Harbin were much lower than those in the southeast of China, which is expected since the use of these OCPs in the former was much lower than that in the latter, however higher HCH concentrations in and around Harbin than those found in most places of the Southeast China is not expected. It is suggested that high HCH concentration in soil of Northeast China was most likely due to long-range atmospheric transport (LRAT) from Southeast China and the cold condensation process.

Key words: organocholrine pesticides (OCPs); soil; residue
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Introduction

Organocholrine pesticides (OCPs) have become ubiquitous in the environment because of their persistence, bioaccumulation and long-range atmospheric transport (LRAT). OCPs have been found in soil of China by many research groups (Chen *et al*., 2005a; Gao *et al*., 2006, 2008; Jean, 2007; Li *et al*., 2006a; Shi *et al*., 2005; Wang *et al*., 2006a, 2007a, 2007b; Zhou *et al*., 2008), most of which focused on the central and south of China, such kind of study in the northeast of China, however, is lacking. In addition, most of the reported studies focused on DDT (dichlorodiphenyltrichloroethane) and HCH (hexachlorocyclohexane), and the study for other OCPs, such as endosulfan, chlordane, and HCB (hexachlorobenzene) were scarce. HCB with higher volatility and LRAT potential was used in past, not only as pesticides but also as an intermediate of some chlorinated industrial processes, such as metal production and various combustion processes. Although HCB is believed to be extremely persistent in the environment, information of HCB in developing countries, such as China, was lacking (Bailey, 2001; Cai *et al*., 2008).

The study of OCPs in the northeast of China, consisting many industrial urban areas and also food production regions, is very important in understanding the source, pathways, and the fate of OCPs in China. Harbin City is the capital of Heilongjiang Province, China. With a population of approximate 3 million, Harbin is a major commercial, industrial, and transportation center surrounded by farmland. The climate there is typically cool monsoon weather with the annual average temperature of 3.6°C, with –13.2~24.8°C in January, and 18.1~22.8°C in July. There was little information on OCPs residue levels in topsoil in this region. The objective of this study was to investigate the current status of residues for OCPs, including HCH, DDT, HCB, chlordane and endosulfan, in topsoil of Harbin and surrounding area, a typical place in Northeastern China.

1 Materials and methods

1.1 Sampling

Topsoil (0–20 cm) samples were collected from seventeen sites within and outside of urban area of Harbin City in October 2006, among which, 9 were urban (UR), 4

* Corresponding author. E-mail: ijrc_pts_paper@yahoo.com

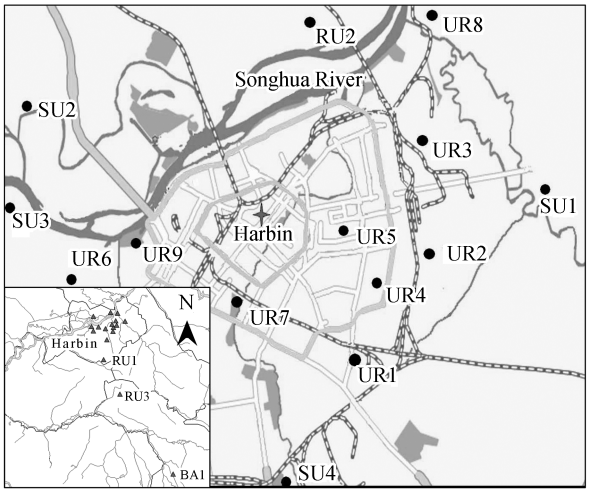

Fig. 1 Location of sampling sites. UR: urban; SU: suburban; RU: rural, BA: background. The other three sampling sites (RU1, RU3 and BA1) were located 10 km, 30 km south and 90 km southeast of the city, respectively.

were suburban (SU), 3 were rural (RU) and 1 background (BA). The locations of these sites are shown in Fig. 1, and the information of the sites is presented in Table 1. Soil sampling sites were chosen away from any point pollution sources and any roads. Topsoil was collected using a stainless steel scoop that had been pre-washed by acetone. Five sub-samples composed one sample, which was placed in a pre-washed glass flask with teflon cap and transported to the International Joint Research Center for Persistent Toxic Substances (IJRC-PTS) laboratory in Harbin Institute of Technology, Harbin, where they were stored frozen (at –20°C) until required for extraction.

About 20 g of each soil sample was Soxhlet extracted for 24 h with 100 mL mix solvent (*n*-hexane/acetone, 1:1, *V/V*). The extract was filtered through a funnel filled with anhydrous sodium sulfate then rotary-evaporated to 4 mL. The extract was passed through 10 g silica gel column, eluted with 60 mL mixture of hexane and dichloromethane (DCM, 1:1, *V/V*). The elution was rotary-evaporated to 2 mL and then reduced to 1 mL under a gentler nitrogen gas flow.

Table 1 Information of sampling sites

Code	Site type	Land use	TOC (%)	Soil dry weight (g)	Sampling date (y-m-d)
BA1	Background	Wood	3.43	20.35	2006-10-27
RU1	Rural	Vegetable	3.19	21.81	2006-10-26
RU2	Rural	Vegetable	2.89	25.59	2006-10-26
RU3	Rural	Cropland	2.08	25.39	2006-10-27
SU1	Suburban	Vegetable	1.05	26.43	2006-10-28
SU2	Suburban	Vegetable	2.8	22.93	2006-10-26
SU3	Suburban	Cropland	1.14	18.50	2006-10-14
SU4	Suburban	Uncultivated	3.31	23.72	2006-10-26
UR1	Urban	Uncultivated	7.61	22.34	2006-10-19
UR2	Urban	Wood	1.97	27.12	2006-10-28
UR3	Urban	Uncultivated	2.33	23.77	2006-10-14
UR4	Urban	Vegetable	2.74	20.94	2006-10-19
UR5	Urban	Wood	3.13	25.58	2006-10-20
UR6	Urban	Uncultivated	5.85	20.73	2006-10-21
UR7	Urban	Wood	1.11	22.94	2006-10-29
UR8	Urban	Grass	2.53	23.97	2006-10-14
UR9	Urban	Uncultivated	3.43	20.88	2006-10-14

1.2 Analysis

Details of analysis of soil samples are reported elsewhere (Ren *et al*., 2007), and a brief description is given here. Analysis of the samples by GC-ECNI-MS was performed on an Agilent 6890GC-5975B MS detector with a HP-5MS capillary column (30 m × 0.25 mm i.d., 0.25 μm film thickness, J&W Scientific, USA), operating on selective ion monitoring (SIM) mode. Helium was used as carrier gas and methane was used as reaction gas with a flow of 2.2 mL/min. GC oven program was 80°C for 1 min, 10°C/min to 160°C, 2°C/min to 250°C, 30°C/min to 300°C. Injections (2 μL) were splitless with split opened after 1.0 min, injector and transfer line temperatures were 250 and 300°C, respectively. The soil organic carbon (SOC) for each sample was measured by using a Shimadzu TOC Analyzer (Model TDC-VCPN, Kyoto, Japan). Fourteen OCPs targeted in this study were quantified against a five point calibration curve. The instrument was operated in SIM mode with target/qualifier ion 255/257 for HCHs, 284/286 for HCB, 410/408 for *cis*- and *trans*-chlordane, 444/446 for *trans*-nonachlor, 406/408/410 for α-, β-endosulfan and endosulfan sulfate, 318/316 for *p,p′*-DDE, 248 for *p,p′*-DDD, 246/248 for *o,p′*-DDT and 248/250 for *p,p′*-DDT.

1.3 Quality assurance/quality control

The detection limits (in pg/g) of OCPs described as 3:1 signal-to-noise value (S/N), were range from 5 to 50 for HCHs, from 0.2 to 1 for HCB, chlordane and endosulfan, from 10 to 100 for DDT. Replicate analyses were performed on site SU3. The relative standard deviations (RSD) were ranged 2.59%–13.90% for HCH, 0.05%–0.11% for chlordane, 6.06%–15.19% for all DDT metabolizes except for *p,p′*-DDT (23.94%). To check method recoveries, two spike samples were prepared by adding 100 μL of the standard pesticide mixure (60 ng) (Accustandard, New Haven, USA). Average recoveries ranged from 74.8% to 101.7% except for β-endosulfan (49.6% ± 27.8%) and endosulfan sulfate (61.1% ± 20.9%). Blank were run by soxhlet extracting a thimble filled with clean sodium sulfate and treating the extract as a sample. Blank values for all compound were below the quantification limits expect for γ-HCH (40 pg/mL), which was still below 5% of the mean concentrations for γ-HCH for all soil samples. All data were reported without recovery and blank correction.

2 Results and discussion

2.1 Soil concentrations of OCPs in Harbin

Concentration levels for the selected OCPs are depicted in Fig. 2, and the descriptive statistics of data on the OCPs are given in Table 2. Figure 2 indicates that much higher concentration levels for HCH, DDT, and HCB than chlordane and endosulfan were found in the region. Concentrations (in pg/g dw) ranged from 136 to 51800 with a mean of 7120 for ∑HCH, from 72 to 28222 with a mean of 5430 for ∑DDT, and from 64 to 3942 with a mean of 1040

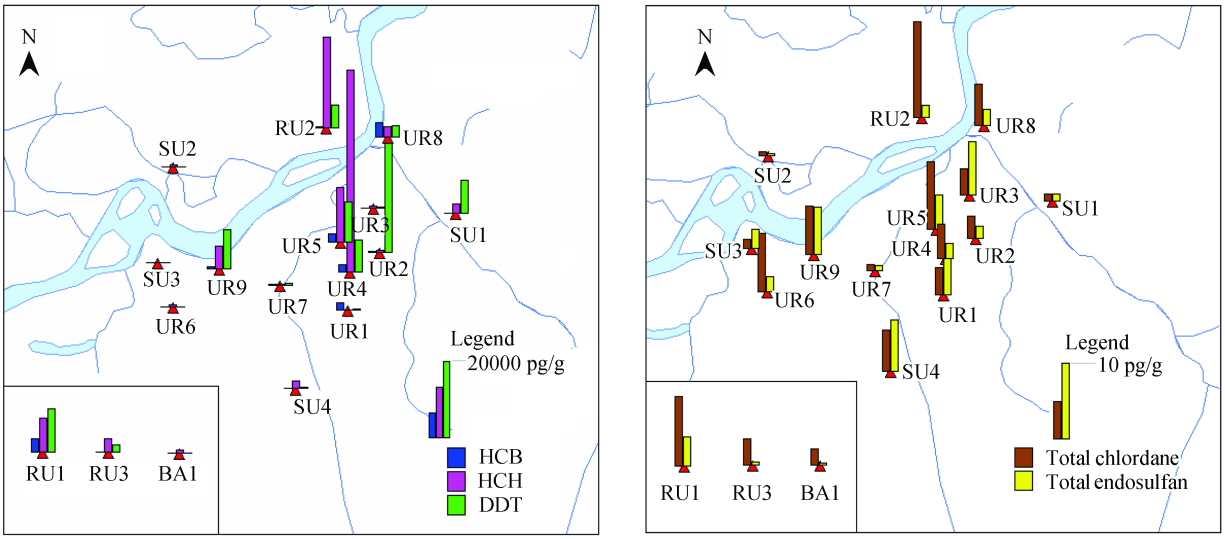

Fig. 2 Concentrations of OCPs in soils of Harbin (in pg/g dw) for HCB, HCH, and DDT (a), and chlordane and endosulfan (b).

Table 2 Descriptive statistics of data on OCPs measured in Harbin topsoils (unit: pg/g dw)

Compound	Number of detected samples	Minimum	Maximum	Mean	Median
HCB	17	64.2	3940	1040	448
α-HCH	17	24.5	2870	672	206
β-HCH	16	BDL	48200	6210	1940
γ-HCH	17	10.8	726	239	84.8
∑HCH	17	136	51800	7120	2230
α-HCH/γ-HCH	–	1.97	3.96	2.65	2.53
β-HCH/α-HCH	–	–	16.8	6.69	5.17
TC	17	0.218	1.92	0.998	1.13
CC	13	BDL	3.71	1.41	1.33
TN	17	0.454	7.77	2.35	1.92
∑Chlordane	17	0.697	12.6	4.75	3.67
TC/CC	–	–	1.14	0.561	0.721
α-Endosulfan	13	BDL	3.09	1.07	0.965
β-Endosulfan	7	BDL	2.27	0.553	BDL
Endosulfan sulfate	17	BDL	4.34	1.75	1.25
∑Endosulfan	17	BDL	8.99	3.34	2.21
o,p′-DDT	14	BDL	1670	478	247
p,p′-DDT	13	BDL	12000	3140	1850
p,p′-DDE	17	72.0	136000	1550	524
p,p′-DDD	15	BDL	1050	415	210
∑DDT	17	72.0	28200	5430	2110
p,p′-DDT/*o,p′*-DDT	–	–	12.2	5.41	5.14
p,p′-DDT/*p,p′*-DDE	–	–	7.58	2.54	2.92

BDL: below detection limit; –: no sense.

for HCB. Differences of 2–3 orders of magnitude were found among all sites for these chemicals. The highest concentrations of ∑HCH occurred at Site UR4 (vegetable land) followed by Site RU2 (vegetable land), most likely due to the higher usage of HCH on vegetable lands and gardens than other soils in Chinese urban area (Wang *et al.*, 2006b). The highest concentration of DDT was found in Site UR2 (woodland) and the lowest concentration was in Site SU3 (cropland). The highest HCB concentration in grassland (UR8) followed by vegetable land (RU1). Concentrations for chlordane and endosulfan were much lower and more uniform among all sites, ranged (in pg/g dw) from 1 to 12.6 with a mean of 4.75 for ∑chlordane, from below detection limit (BDL) to 7.1 with a mean of 3.34 for ∑endosulfan. No significant correlation was found between those OCPs and total soil organic carbon (SOC) among all sites.

2.2 Residual characteristics of OCPs

As a commercial insecticide, HCH was used in two formulations: technical HCH, which is a mixture of several isomers, among which, α-HCH comprises 60%–70%, γ-HCH comprises 10%–12% and β-HCH comprises 5%–12%, and lindane, which consists almost all of γ-isomer (Li *et al.*, 2000). In the technical HCH mixture, the α-HCH/γ-HCH ratio is around 5–7 and α-HCH/β-HCH ratio is about 5–14, while the average ratios of α-HCH/γ-HCH were 2.7 and α-HCH/β-HCH were 0.1 in the investigated soil samples from Harbin area, suggesting that there has not been fresh input of technical HCH and lindane in the Harbin area for a long time. This result is consistent with relatively low historical use of both technical HCH and

lindane in the region (Li *et al*., 2001), and therefore the major HCH source in Harbin region was from historical local technical HCH contamination, and probably also those through LRAT.

Technical DDT typically contains 77.1% *p,p′*-DDT, 14.9% *o,p′*-DDT, 4% *p,p′*-DDE (Zhu *et al*., 2005). DDT can be converted to DDE under aerobic condition while to DDD under anaerobic condition (Heberer and Dünnbier, 1999). In general, the ratio of DDT/DDE has been used to estimate the age of DDT (Bidleman, 1999), a small value of *p,p′*-DDT/*p,p′*-DDE ratio (< 1) is indicative of aged DDT, while ratio > 1 means its fresh inputs (Daly *et al*., 2007). But several factors, such as soil type, temperature, moisture and organic carbon content, can affect the conversion rate. Recent studies found that the high proportion of parent DDT relative to its derivatives does not necessarily indicate the fresh use of technical DDT in this region. Villa *et al*. (2006) suggested that the process of conversion from DDT to DDE was very slow in soil while volatilization was the main process for the DDT dissipation in tropical countries, so low temperatures in cold regions lead to a reduction in microbiological activity and also volatilization for DDT. Dimond and Owen (1996) reported that the half-life is 20–30 years with DDT and ratio of DDT/DDE ranged from 1.3 to 13 after twenty-five years, slowly metabolism of DDT in Maine, USA, due to low average annual temperature (5°C). Another similar example is the study carried out in southern Ontario, Canadian in fall 2004 and spring 2005 (Kurt-Karakus *et al*., 2006). Very high soil concentrations of ∑DDT (1.899×10^7 pg/g dw) and high ratio of *p,p′*-DDT/*p,p′*-DDE (3.5) in soil of vegetable farming area were found even though this insecticide was banned for agricultural uses in Canada about three decades ago. A volatilization half-life of 200 years was estimated for ∑DDT in the upper 5 cm of the soil column in the area (Kurt-Karakus *et al*., 2006).

As shown in Table 2, the ratio of *p,p′*-DDT/*p,p′*-DDE for the 17 soil samples ranged from BDL (Sites UR6, SU2, SU3 and BA1) to 7.6 (Site SU1) with average ratio of 2.5, which is higher than Beijing (Zhu *et al*., 2005), Tianjin (Wang *et al*., 2006b) and Guangzhou (Chen *et al*., 2005b) (Table 3). Due to lower temperature with an annual average of 3.6°C in the city of Harbin, DDT in soils of this region is expected more persistent in comparison to the south part of China, and the relatively high DDT concentrations and high ratio of *p,p′*-DDT/*p,p′*-DDE will possibly continue in the region.

The ratio of *p,p′*-DDT/*o,p′*-DDT can also provide further information on whether the residues are caused by technical DDT or by technical dicofol. Generally, *p,p′*-DDT/*o,p′*-DDT ranged from 3 to 5 in technical DDT and from 0.01 to 0.77 in technical dicofol (Qiu *et al*., 2005). Our present study gave the average value of 5.4 for *p,p′*-DDT/*o,p′*-DDT, showing the source from technical DDT but not from technical dicofol. Our result is very close to the Alabama study. The average value for *p,p′*-DDT/*o,p′*-DDT was 6 in soil of Alabama, USA (Harner *et al*., 1999), little higher than technical DDT. It was explained that *p,p′*-DDT and *o,p′*-DDT have declined at similar rates since application ceased in Alabama. Table 3 presents low ratios of *p,p′*-DDT/*o,p′*-DDT (1.0–2.9) in other places in the south of China, possibly indicating the use of dicofol in this regions (Qiu *et al*., 2005).

Technical chlordane, including *trans*-chlordane (TC), *cis*-chlordane (CC), and *trans*-nonachlor (TN), has the proportion of 1:0.85:0.42 for TC:CC:TN (Jantunen *et al*., 2000). Degradation rate of chlordane is in the order of TC > CC > TN (Harner *et al*., 1999). Average proportion of TC:CC:TN in Harbin soil samples were 1:1.78:2.54, similarly indicating that chlordane observed in Harbin topsoil originated from past technical chlordane use, or due to LRAT. Indeed, it was reported that chlordane has not been used in the northeast of China since 1988 (Zhao, 2006).

Technical endosulfan is a mixture of two isomers, α- and β-Endosulfan, in the ratio of 7:3 (Gonzalez *et al*.,

Table 3 Comparison of mean concentration for DDT and HCH in soil of Harbin with other region in China (unit: pg/g)

Location	Land use	∑DDT	*p,p′*-DDT/ *p,p′*-DDE	*p,p′*-DDT/ *o,p′*-DDT	∑HCH	α-HCH/ γ-HCH	α-HCH/ β-HCH	Survey year	Reference
Harbin	Urban and suburban	5425	2.5	5.4	7120	2.7	0.1	2006	This study
Beijing	Orchard	993600	0.5	–	64200	1.8	0.0	2003	Shi *et al*., 2005
Beijing	Suburban	140790	1.1	2.9	10350	0.5	0.2	–	Zhu *et al*., 2005
Beijing	Urban and suburban	38200	0.4	1.2	5800	0.5	0.2	2003	Li *et al*., 2006a
Beijing	Agricultural soils	77200	0.4	8.2	1500	1.4	0.7	2003	Zhang *et al*., 2007
Beijing	Park soil	162000	0.9	1.0	10500	0.7	0.1	–	Li *et al*., 2008
Beijing	Suburban	64400	0.3	1.6	15800	1.2	0.1	2001	Ma *et al*., 2003
Beijing	Guanting Reservoir	5100[a]	–	–	560[a]	Nd[b]–1.9	–	2003	Zhang *et al*., 2005
Tianjing	Industrialized area	80100	0.06	1.3	3600	500	0.2	2002	Tao *et al*., 2005
Shanghai	Agricultural soils	84000	0.3	–	2400	3.5	0.5	2001	Nakata *et al*., 2005
Taihu	Basin agricultural soils	7800	–	–	680	–	–	–	Nakata *et al*., 2005
Haining, Zhejiang	Agricultural soils	83000	–	–	710	–	–	–	Nakata *et al*., 2005
Wuxi, Jiangsu	Agricultural soils	163200	–	–	11100	–	–	–	Nakata *et al*., 2005
Guangzhou	Vegetable	81400	–	–	4390	2.5	0.6	–	Chen *et al*., 2005b
Guangzhou	Agricultural soils	64600	0.8	–	6200	1.7	–	2004	Gao *et al*., 2008
Hong Kong	Rural and urban	520	–	–	6190	6.0	–	–	Zhang *et al*., 2006
30 studies in China	–	60000	0.25	–	8700	2.7	–	–	Cai *et al*., 2008

[a] Values as meadian; [b] not detected. "–" means not available.

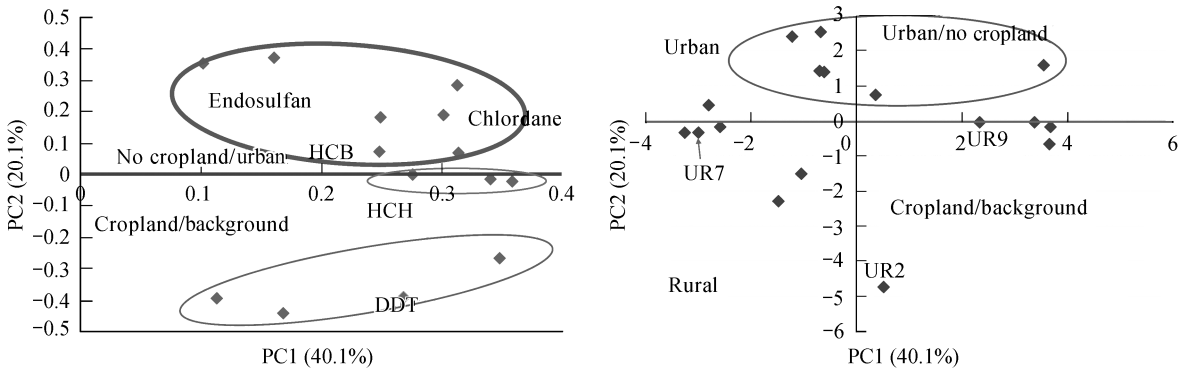

Fig. 3 The first two principal components (PC1 and PC2) for OCPs. The Score shows the top portion is for non cropland in urban sites, and the rest is for cropland sites with some exceptions (UR2, UR7, and UR9).

2003; Yao *et al*., 2006). α- and β-Endosulfan in soil are degraded to the highly toxic endosulfan sulfate. Our results showed that endosulfan sulfate level in Harbin topsoil were almost equal or higher than its parent compounds in most of soil samples, indicating degraded characteristics of the chemicals in soil. This, along with the fact that very low concentration and its fairly uniform pattern among farm land, woodland, and grassland, suggest that endosulfan in this area may be due to LRAT, and no or very few of this pesticide has been used in this region.

2.3 Principal component analysis

The principal component analysis (PCA) was performed (software provided by Bristol Chemometrics, http://www.chm.bris.ac.uk/org/chemometrics) on the selected OCPs at 17 sites and all dataset was normalized to reduce the wide range of concentration data for different pesticides (Table 2). The first two PCs account for 40.1% and 20.1% of the total variance respectively, and both loading and score are presented in Fig. 3. The top cluster in the Loading, shows that HCB, chlordane, and endosulfan are mainly found in urban non-cropland sites, indicated in the Score, while HCH and DDT are most likely linked to cropland (including vegetable land in the urban area). The result probably reflects the variety of different sources for these compounds.

HCB was used in China not only as fungicides but mainly as an intermediate of some chlorinated industrial processes, such as synthetic rubber, polyvinyl chloride plastics, dye, and metal production and various combustion processes (www.china-pops.net), thereby, it is not surprising that the major source of HCB were found in the urban area. As discussed above, no or very few chlordane and endosulfan was used in northeast of China, our PCA result still indicates possible existence of a small amount of local sources of these two chemicals in the urban soil in Harbin. Chlordane was mainly used in the urban area to protect new constructions (Zhao, 2006) and endosulfan was also used on vegetable in urban area (Xu *et al*., 2005; Jia *et al*., 2008, 2009). In fact, our data showed that, the mean concentration of endosulfan in the urban and suburban soils (3.9 pg/g dw) was twice as many as that in rural and background soils (1.6 pg/g dw). Currently-used pesticide endosulfan found in urban non-cropland probably transported from vegetable land nearby. It seems obvious that insecticides DDT and HCH were link to cropland since they were mainly used on land of rice, cotton, wheat, maize, soybean, and sorghum in the rural (agricultural) area, and also on urban vegetable soils (Li *et al*., 1998a, 1998b, 2001). In comparison to DDT, however, HCH is also close to the cluster for HCB and chlordane, indicating that the HCH in urban non-cropland was also important. In fact, HCH was found almost equally in both urban and rural area, probably due to the heavy use of this pesticide on agricultural land in rural area and on vegetable and garden sites in the urban area many years ago (Wang *et al*., 2006b), and also due to many yeas of short- and long-range transport of this insecticide, leading a more evenly distribution pattern among cropland and noncropland.

2.4 Comparison with OCPs in other regions in China

The concentration levels of HCB in Harbin were higher than that in Hong Kong (20 pg/g dw) (Zhang *et al*., 2006), in the same order as that in suburban soil of Beijing (1840 pg/g dw) (Zhu *et al*., 2005), but lower than those in the region of Taihu Lake (ranging from 1860 to 5130 pg/g dw) (Wang *et al*., 2007b), and the chemical industry zone in Beijing (3100 pg/g dw) (Zhou *et al*., 2007).

Concentrations of ∑chlordane (TC+CC+TN) were 1–3 orders of magnitude lower than those from Taihu Lake Region (40 pg/g dw) (Nakata *et al*., 2005), Pearl River Delta (1530 and 1610 pg/g dw for TC and CC in natural soil) (Li *et al*., 2006b) in China and archived soils in UK (range from < 90 to 1600 pg/g dw) (Meijer *et al*., 2001).

Concentrations of total endosulfan in Harbin soil were much lower as compared to the other studies in China. For instance, it was reported that α-endosulfan was in the range 1600–3800 pg/g dw in topsoil in Taihu Lake region (Wang *et al*., 2007b) and range from 250 to 3690 pg/g dw (mean 1680 pg/g dw) in topsoil near River Wuchuan of Southeast China (Zhang *et al*., 2002).

Table 3 gives comparison of mean concentration for HCH and DDT in soil of Harbin with those in other region in China published recently. While concentration of ∑DDT in soil samples from Harbin was lower than those measured in most places in the Southeast China, mean concentration of ∑HCH in Harbin soil was higher than those in these area, including Taihu Basin, Guangzhou and

Hong Kong.

It is expected that concentrations of chlordane, endosulfan and DDT in topsoil in Harbin were lower than those in other southern places in China. From 1970 to 2003, China has produced 4340 tons of chlordane, among which 1480 tons of chlordane was exported and 2860 tons of chlordane was used in China, more than 95% of which have been used in fighting termites to protect new constructions (Zhao, 2006). The intensive use of chlordane was concentrated in the southeast part of China, Yangtze Delta and Zhujiang Delta in particular, and no use in Harbin (Zhao, 2006). According to Jia *et al*. (2009a, 2009b), around 25700 tons of endosulfan were used in China between 1994 and 2004, mainly on cotton, wheat, tea, tobacco, and orchard. The use of this pesticide in the northeast of China is very low, leading low concentrations of endosulfan in Harbin topsoil.

Lower concentration of ∑DDT in soil samples from Harbin than those measured in most places in Southeast China is expected since the use in Heilongjiang was much lower than the Southeast China. China used 270000 tons DDT from 1952 to 1984, mainly in southeast of China (Li *et al*., 1998b), and the usage in Heilongjiang Province is < 2% of the national total (Liu *et al*., 2006). Higher concentrations of HCHs in the Northeast China than those in most places in Southeast China, however, is not expected. China used approximately 4 millions tons of technical HCH from 1952 to 1984 (Li *et al*., 1998a) and 3200 tons for lindane between 1991 and 2000 (Li *et al*., 2001), mostly in the Southeast China, Yangtze Delta and Zhujiang Delta in particular, with only < 3% in Heilongjiang Province with about 5% of total country area (Liu *et al*., 2006). It was suggested that high HCH concentration in soil of Northeast China was most likely due to LRAT from the Southeast China, and acted a sink of this compound due to this low temperature in the region. This distribution process has gone for many decades, and has been referred as "cold condensation" phenomenon (Wania and Mackay, 1993). Interestingly, it has been reported that Northeast China has been a major receiver of -HCH under the summer monsoon regime (Tian *et al*., 2009).

3 Conclusions

Seventeen topsoil samples from Harbin in Northeast China have been analyzed for selected OCPs. The mean concentrations (in pg/g dw) of HCH, DDT, HCB, chlordane and endosulfan are 7120, 5425, 1039, 4.8, 3.3, respectively. β-HCH and *p,p*'-DDT were two dominant substances with mean concentration of 6208 and 3138 pg/g dw. Levels of chlordane and endosulfan were generally low in all the soil samples as compared to the other studies in China, indicating no or very few usage of these two pesticides in the study region. The residues DDT were found to be from past usage. High HCH concentration in soil of Northeast China was most likely due to LRAT from the Southeast China, and acted a sink of this compound due to this low temperature in the region. It was also observed that concentrations of HCHs and DDTs in most of soil samples were much lower than the First Grade (50 ng/g) permitted in "Environment quality standard for soils in China (GB15618-1995)".

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