Environ. Sci. Technol. **2009,** *43,* 1316–1321

Seasonal Patterns and Current Sources of DDTs, Chlordanes, Hexachlorobenzene, and Endosulfan in the Atmosphere of 37 Chinese Cities

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Received August 25, 2008. Revised manuscript received December 11, 2008. Accepted December 11, 2008.

China has a history of large scale production and application of organochlorine pesticides (OCPs) although, data on their nationwide distribution and seasonal variations in the atmosphere is still sparse. Passive air samplers (PAS) were therefore utilized to obtain seasonal data from 37 Chinese cities and three background sites in 2005. Concentrations and spatial and seasonal distribution of dichlorodiphenyltrichloroethanes (DDTs), chlordanes (CHLs), hexachlorobenzene (HCB), and endosulfans (Endo) are presented in this paper, and their potential sources are discussed based on the dataset. It is estimated that ca. 95% of DDTs present in the atmosphere of Chinese cities was still from technical DDT, while only ca. ∼5% was "dicofol-type of DDT". DDT application for public health control and DDT activated antifouling paint for fishing ships may be the two most important current sources of technical DDT. The DDT concentrations in several Chinese cities seem to match well with the reported DDT concentrations in human breast milk. A low TC/CC ratio was observed across China in the winter to spring, which may provide a fingerprint of Chinese chlordane emission. It was suggested that "weathered" chlordane emitted from urban construction foundations in winter may give the distinctively low TC/CC ratio. The data showed that China is an important global source for HCB. Higher HCB concentrations were observed in winter and spring, and in colder cities, highlighting an important contribution from combustion sources. Samples with higher endosulfan concentrations occurred in the cotton production areas, indicating its major use in killing cotton pests.

Introduction

The Stockholm Convention on persistent organic pollutants (POPs) has focused attention on the need to identify sources of these chemicals and to monitor for their presence in the environment (*1*). From a global perspective, recent attention on potential source areas for the "old generation" organochlorine pesticides (OCPs) has focused on developing countries in tropical and subtropical regions. China is of particular interest, because of (i) the size of its land mass and population; (ii) its strong agricultural base and rapid development; (iii) the introduction of bans rather later than in some other countries; (iv) the widespread past use of OCPs (*2*); (v) evidence of ongoing usage and fresh inputs of some OCPs (e.g., DDT and chlordane) (*3, 4*); (vi) ongoing production of some OCPs and intermediates (*3, 5*). China is ranked first for the usage of technical HCH and fifth for DDT (*5*). Recent surveys and monitoring have reported relatively high levels of OCPs in Chinese air, soils, water bodies, and human biota samples (*6*−*9*).

Recent developments in passive air samplers (PAS) (*10*) have made it possible to conduct large-scale spatial surveys of POPs distributions in the atmosphere, with the purpose of building up information on compound distribution and sources. Examples include a continental scale survey across Europe and deployments across global background sites (*11, 12*). Indeed, an initial survey of polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), hexachlorobenzene (HCB), and DDTs in China was conducted in 2004, using the PAS approach (*13*). This paper presents a more detailed follow-up to that work, by presenting data for DDTs, chlordanes, HCB, and endosulfan during the four seasons of 2005, from 37 cities and three background stations. Cities were the focus here, for logistical reasons and because they can be important sources of OCPs, such that levels and human exposures may actually exceed those in rural, agricultural areas (*14, 15*). The data are shown here and used to explore source information about the chemicals.

Materials and Methods

Site Locations and Air Sampling. Thirty seven cities (provincial capitals and major regional centers) and three background stations were selected (see Supporting Information Figure S1 and Table S1). In the cities, the PAS were deployed in open areas, >3 m above the ground. The seasonally averaged wind fields across China with back-trajectories at three background sites in the year of 2005 were determined by analyzing meteorological data from the National Centre for Atmospheric Research (NCAR), Japan (see Supporting Information Figure S2).

The PUF disk PAS apparatus has been used and described several times previously (e.g., (*13, 16, 17*)). All the PUF disks including 116 samples, 54 field blanks, and 12 laboratory blanks were pre-extracted with dichloromethane (DCM) and acetone at Guangzhou Institute of Geochemistry and transferred by express mail service to sampling sites in glass jars. Four separate deployments of 8 weeks were made in 2005: winter (February 1 to March 28), spring (April 1 to May 28), summer (July 5 to August 30) and autumn (September 15 to November 11), respectively. Sample loss occurred occasionally during sampling, transport, and storage. A total of 200 PUF disks inclusive of 54 field blanks were dispatched, of which 174 were returned and analyzed for OCPs.

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10.1021/es802371n CCC: $40.75
Published on Web 01/26/2009

TABLE 1. Summary Data of DDTs, Chlordanes, Endosulfans, and HCB in the Atmosphere of 37 Chinese Cities and Seven Background Sites (pg·d^{-1})[a]

	winter					spring					summer					autumn				
Season	min	max	mean	std	Bg	min	max	mean	std	Bg	min	max	mean	std	Bg	min	max	mean	mtd	Bg
o,p′-DDT	97	2200	315	426	531	33	5080	833	1170	46	n.d.	6620	1390	1580	290	25	4050	761	1070	14
p,p′-DDT	31	1050	210	211	166	11	1600	474	491	16	n.d.	2730	515	551	65	20	2370	464	626	12
p,p′-DDE	47	568	169	140	135	5.9	3340	453	625	17	24	3650	712	775	93	n.d.	3110	423	635	20
Hep	n.d.	1240	120	235	18	n.d.	2220	174	396	34	n.d.	788	144	174	42	n.d.	422	114	128	15
TC	43	733	182	176	56	24	5620	322	995	47	35	3690	586	755	337	34	4290	688	936	23
CC	20	4720	1270	1020	253	25	4760	839	895	34	35	3940	909	698	351	90	5300	1250	1240	54
TN	12	137	47	34	12	8.9	1140	70	201	24	9.2	1000	143	189	159	10	1240	170	262	6.6
CN	n.d.	107	21	21	4.6	n.d.	1100	49	199	1.4	n.d.	147	31	33	15	n.d.	165	24	37	n.d.
α-Endo	54	1190	197	215	47	3	224	64	46	7.8	n.d.	170	40	38	17	9.5	592	98	115	10
β-Endo	5.3	422	49	86	20	1.8	328	41	62	3.6	n.d.	404	63	86	8.3	n.d.	126	20	28	4.5
HCB	123	6510	2080	1410	455	207	5450	1277	1230	868	48	3790	1180	778	916	490	4300	1380	936	804
TC/CC	0.04	5.7	0.45	1.1	0.22	0.04	3.8	0.38	0.68	1.5	0.07	1.6	0.59	0.40	1.1	0.05	2.1	0.59	0.53	0.43
op′/*pp′*-DDT	0.21	14	2	2.6	3.2	0.16	7.1	2	1.7	3.3	n.d.	5.4	2.6	1.4	5	0.37	3.6	1.7	0.75	1.3
p,p′-DDT/ *p,p′*-DDE	0.18	7.1	1.7	1.5	1.2	0.04	23	2.2	4.1	1.3	n.d.	7.4	1.1	1.3	0.66	0.38	5.2	1.2	0.99	0.65

[a] n.d.: Not detected. Bg: Average values at background sites.

Analysis and QA/QC. Detailed analytical procedures and QA/QC data were provided in the Supporting Information (S1: Analysis and QA/QC). Briefly, after adding surrogates, the PUF disks were Soxhlet extracted with DCM. Clean-ups were done by column chromatography (*18*), and the measurement was conducted using an Agilent 5975N GC-MSD (*19*). All reported values were corrected by the field blanks and surrogate recovery.

Results and Discussion

Introductory Remarks on the Data Set. Data derived from PAS can be presented in different ways. Commonly, this is the compound mass detected per sampler for a given deployment (e.g., ng/sampler), or a time-normalized mass per sampler (e.g., ng sampled/day). Either of these approaches highlights spatial and seasonal differences. Alternatively, the data can be converted to an estimate of the compound mass per unit volume of air (e.g., ng·m^{-3}), using information from uptake calibration studies and permeation compounds (*17*). Depending on the configuration of sampler, uptake rates using the sampler design here are typically 3−5 m^3·day^{-1} (*12, 17*). In this paper where the focus is on spatial and seasonal comparisons, the data are presented as ng·day^{-1}, to allow slight differences in deployment time between locations and sampling occasions. However, some estimates of typical air concentrations are made, at an average sampling rate of 3.5 m^3·day^{-1} (*13*), to give necessary international context to the levels measured. Derived concentration data are generally within a factor of 2 compared to those by conventional active samplers (*12, 17*).

Table 1 presents summary data for concentrations and isomer ratios of OCPs obtained by PAS in the Chinese cities and background sites (pg·day^{-1} per disk) for the winter, spring, summer, and autumn of 2005. Detailed site information is given in Supporting Information Table S2.

In general, the OCP concentrations derived here (as pg·m^{-3}, Supporting Information Table S3) were similar to those reported by other recent studies in China (*2, 20*). The concentrations of DDTs, chlordanes, and HCB in the Chinese cities were much higher than in those in Europe and North America (*12, 17*), and also higher than other parts of Asia previously reported (*11, 13, 21*). By contrast, endosulfans occurred at relatively low concentrations compared to North America and Europe (*11*).

HCHs were also measured in the PAS samples (refer to Supporting InformationTable S2 for HCH data). However, the data are not discussed here because of the page limit.

Spatial and Seasonal Distribution of DDTs. Figure 1 presents the DDT distributions for the four sampling seasons. Relatively high levels of DDTs (summation of *o,p′*-DDT, *p,p′*-DDT and *p,p′*-DDE) were observed mostly in the cities of southern, southwestern and central parts of China.

(i) In winter the highest DDT levels were observed in Guangzhou (2780 pg·d^{-1}), Zhuzhou (2330 pg·d^{-1}) and Xi'an (1410 pg·d^{-1}). At other sites the values were generally lower than 900 pg·d^{-1}.

(ii) In spring most elevated DDT levels were observed in the southeast to southwest parts of China, namely Guangzhou (7650 pg·d^{-1}), Zhuzhou (3560 pg·d^{-1}), Changsha (2950 pg·d^{-1}), Fuzhou (3340 pg·d^{-1}), Chongqing (6170 pg·d^{-1}), Chengdu (2620 pg·d^{-1}); and the levels were also high in northwest and northern China, such as at Kelamayi (1620 pg·d^{-1}), Urumuchi (1440 pg·d^{-1}) and Dalian (2500 pg·d^{-1}).

(iii) In summer the DDT levels were high in most of the sampling sites except for some cities in southwestern and northwestern China. Across the country the highest DDTs were observed in Guangzhou (5600 pg·d^{-1}), Zhuzhou (6550 pg·d^{-1}) and Chongqing (11 000 pg·d^{-1}).

(iv) In autumn the DDT levels were much higher in southern China and the coastal regions such as Beihai (5370 pg·d^{-1}), Guangzhou (2900 pg·d^{-1}), Xiamen (6600 pg·d^{-1}), and Fuzhou (9530 pg·d^{-1}). Also relatively high levels were observed at Chongqing (4270 pg·d^{-1}) in southwest, Xi'ning (2840 pg·d^{-1}) in northwest, and Beijing (1250 pg·d^{-1}), and Shenyang (1140 pg·d^{-1}) in northern China. The spatial pattern for autumn in 2005 was similar to the previous survey in 2004 (*13*).

Sources of DDT in Chinese Cities. DDT was produced and widely used in China from the 1950s to its official production ban in 1983. Only smaller amounts have been used since then. Approximately 460 000 tonnes of technical DDT was produced in China until 1993 (*5*). Recent production is estimated to be a few thousand tonnes a year (*5*). The majority of this (∼80%) is used for dicofol production (*3*). DDT residues in dicofol are required to be low (<0.1%), but can reach ca. 20% in some dicofol products because of improper production technology (*3, 22*). The known sources of technical DDT are for malaria control, mosquito repellency, and as an active additive in antifouling paints (*23, 24*).

Ratios of DDT isomers can provide useful source information. However, environmental processes such as evapo-

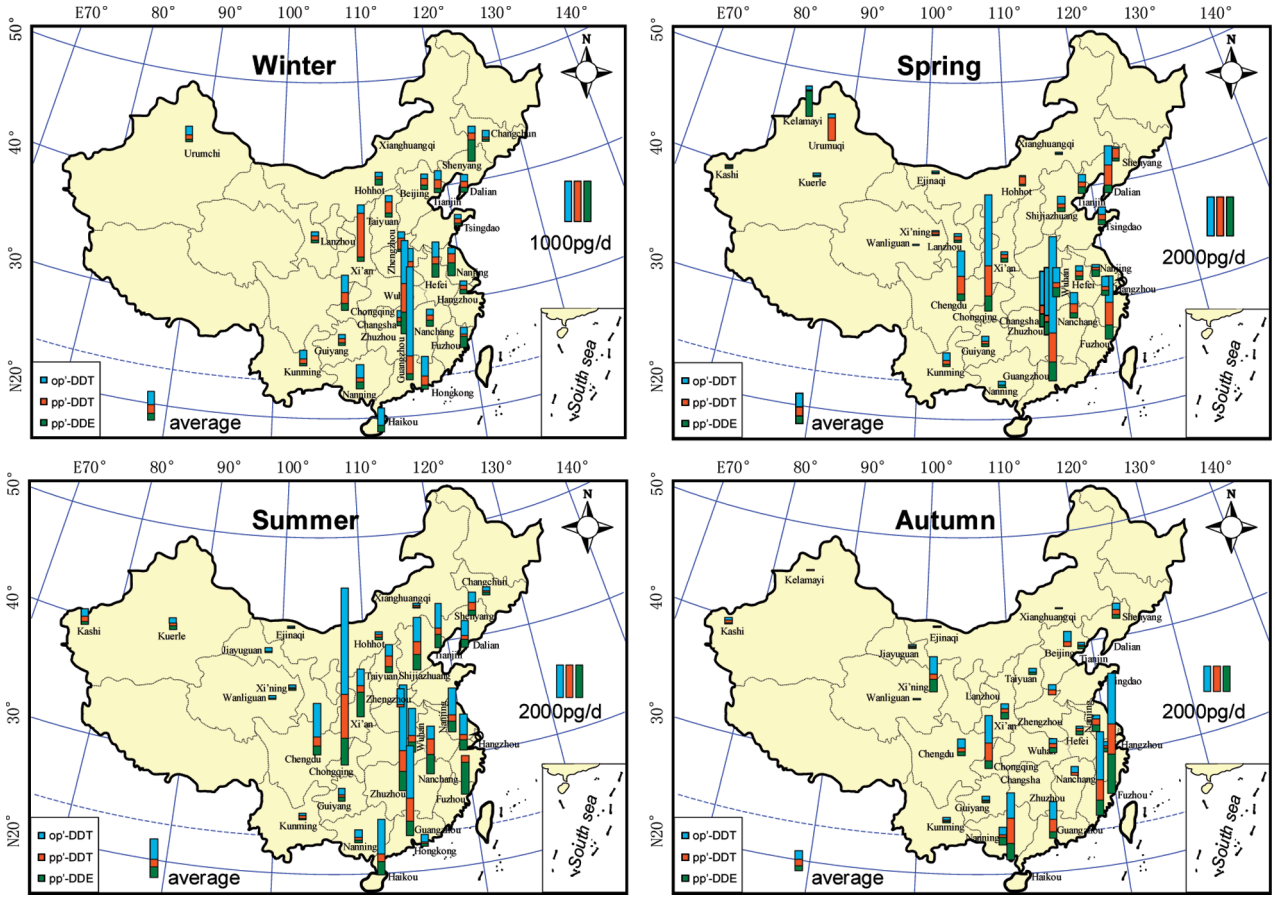

FIGURE 1. Spatial−temporal distribution and compositional patterns of DDTs in the atmosphere of 37 Chinese cities in 2005.

ration and air−surface exchange can lead to fractionation of the isomers and hence change the ratios. The difference of isomer ratios between soil and atmosphere profiles can be predicted by the compounds' subcooled liquid vapor pressure (p_L^0) which governs the vaporization of chemicals from soil to atmosphere. Assuming that only two types of DDT sources dominate their emissions to the environment, if air−soil equilibrium is achieved, the ratios between air and soil can be written as follows (*25*):

$$R_{air(a/b)} = (R_{S1(a/b)} * x + R_{S2(a/b)} * y) * p_{L\,(a/b)}^{0} \quad (1)$$

$$x + y = 1 \quad (2)$$

Where $R_{air(a/b)}$ is the ratio of a/b in the air, $R_{S1(a/b)}$ and $R_{S2(a/b)}$ are the ratios of a/b in source 1 and source 2, respectively; and x and y are the contributions from the two sources, which can be calculated as follows:

$$x = R_{S1} = [R_{air(a/b)} * (p_{L\,(a)}^{0}/p_{L\,(b)}^{0}) - R_{S1(a/b)}]/[R_{S2(a/b)} - R_{S1(a/b)}] \quad (3)$$

$$y = R_{S2} = [R_{air(a/b)} * (p_{L\,(a)}^{0}/p_{L\,(b)}^{0}) - R_{S2(a/b)}]/[R_{S1(a/b)} - R_{S2(a/b)}] \quad (4)$$

Several recent studies suggested that primary/fresh DDT emissions still exist, given the observed high DDT concentrations along with high DDT/DDE ratios. In this study, the *p,p′*-DDT/*p,p′*-DDE ratios for Chinese cities was 0.2−7.1 (mean 1.7 ± 1.5), 0.04−23 (2.2 ± 4), 0−7.4 (1.1 ± 1.2), and 0.4−5.2 (1.2 ± 1) for winter, spring, summer, and autumn, respectively. Taking isomer fractionation during the air−soil exchange processes into consideration, from eq 1, the average *p,p′*-DDT/*p,p′*-DDE in soil, which is more comparable to industrial products than that in the air, was calculated to be 11.8, 14.9, 7.7, and 8 for winter, spring, summer, and autumn, respectively. These high ratios obviously suggest a fresh DDT input in the Chinese cities (Table 1), similar to previous studies (*8, 26, 27*).

The ratios of *o,p′*-DDT/*p,p′*-DDT can be used to distinguish technical DDT from "dicofol- type DDT". Normally a higher proportion of *o,p′*-DDT than *p,p′*-DDT can be found in dicofol residues. The *o,p′*-DDT/*p,p′*-DDT ratio was reported to be 0.2∼0.26 in technical DDT and ∼7.5 in dicofol products (*3*). According to eq 1, these product ratios would result in a calculated ratio of 0.74−0.96 for technical DDT and ∼28 for dicofol-type DDT, respectively, in the atmosphere.

An average ratio of 7 has been reported for *o,p′*/*p,p′*-DDT in the air around the Taihu Lake district in East China (*3*). For this value, according to eqs 3 and 4, the contributions from technical DDT and dicofol-type DDT will be ∼75 and ∼25%, respectively. This is different from the previous reports (*3, 25*) in which most of DDT in the Taihu Lake was suggested to be "dicofol-type", based on a direct comparison of the ratios in the air with those in the products.

According to our data, technical DDT still dominated in the air of most of the cities in China. The *o,p′*/*p,p′*-DDT ratios were 0.21−14 (mean, 2.01 ± 2.59), 0.16−7.1 (mean, 2 ± 1.69), 0−5.4 (mean, 2.64 ± 1.42), and 0.4−3.6 (mean, 1.66 ± 0.75) in the winter, spring, summer, and autumn, respectively (Table 1). Thus it can be estimated that the current DDT sources in the Chinese cities was composed of ∼95% of technical DDT and ∼5% of dicofol-type DDT.

Technical DDT may be introduced to the atmosphere of the Chinese cities via (i) direct use in cities for killing mosquito and health control programs, (ii) emission from DDT producing factories (e.g., in Tianjin), and (iii) release from DDT-activated antifouling paints. As mentioned above, at least 520 tonnes per year of technical DDT is used in China,

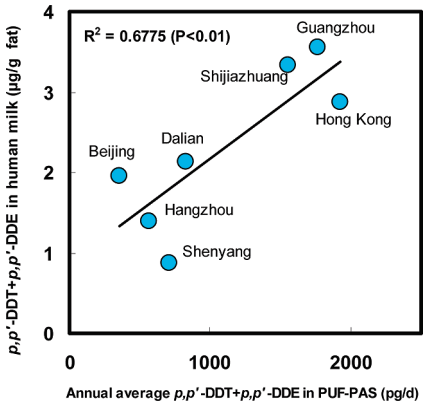

FIGURE 2. Correlations of *p,p′*-DDT+*p,p′*-DDE in the passive air samples and those in human breast milk in seven Chinese cities. Data of human breast milk was from refs *9, 14, 32*−*34*. PAS data for Hong Kong was from ref *28*.

exclusive of that for malaria control (*24*). Whereas from 1988 to 2002 there was ~625 tonnes "dicofol-type DDT" released into the environment every year (*3*). It was also estimated that 5000 tonnes of DDT-activated antifouling paint was used for fishing ships, corresponding to ~250 tonnes of technical DDT (*19, 24, 28*) used every year, since the 1990s. The technical DDT released from DDT-activated antifouling paint may be helpful to explain the observed high DDT levels in the coastal sites in autumn (refer to Figure 1 and Supporting Information Tables S1 and S2).

However, during spring and summer time, several cities did show higher *o,p′*-DDT/*p,p′*-DDT ratios signifying a dicofol contribution (*3*) (e.g., Guangzhou, Zhuzhou, and Changsha in spring, and certain cities in summer) (Figure 1 and Supporting Information Table S2), which may be attributed to dicofol use for horticultural purposes, and/or, possibly lesser use for vegetables and fruits.

Comparison with Human Breast Milk. Atmospheric POPs can enter into ecosystems via several pathways (*29*−*31*). High levels of atmospheric DDT may result in high human exposure and body burdens, both via direct inhalation and—more importantly—higher general environmental and foodstuff levels. As shown in Figure 2, the DDT concentrations in several Chinese cities seem to match well with the reported DDT concentrations in human breast milk ($p < 0.01$) (*9, 14, 28, 32*−*34*). Humans are exposed to DDTs principally through diet, much less via inhalation (*35*). However, the fresh DDTs at high concentrations in the atmosphere as observed in this study would reflect the existence of an active and bioavailable DDT pool in the urban/regional environment, which can be eventually related to the extent of human exposure.

Chlordanes. Chlordane has been widely used for termite control in China, to treat building foundations, dams, grasslands and forests (*19*). Table 1 and Supporting Information Figure S3 show the chlordane patterns and spatial distribution in the Chinese cities for the four sampling seasons. High chlordane levels were observed. The total chlordane levels were 1640 ± 1320, 1450 ± 2100, 1810 ± 1670, and 2500 ± 2390 pg·d^{-1} for winter, spring, summer, and autumn, respectively. CC dominated the chlordane family, accounting for 77, 58, 50, and 56% of the total chlordane in winter, spring, summer, and autumn, respectively. TC accounted for 11, 22, 32, and 31%, respectively. TN and CN generally accounted for 1−12%.

(i) Winter: the average chlordane level was 1640 pg·d^{-1}. Remarkably, CC (1270 pg·d^{-1}) dominated the chlordane composition in the atmosphere in most of the Chinese cities in winter, implying a control by secondary or "weathered" chlordane sources (i.e., low TC/CC ratios, see further discussion below).

(ii) Spring: the chlordane levels were relatively low in spring except at Guangzhou (1450 ng.d^{-1}).

(iii) Summer: the average chlordane level was 1810 ng·d^{-1}, relatively high concentrations along with high TC proportions were observed in southern, southwestern and eastern China, indicating a fresh chlordane input.

(iv) Autumn: autumn showed the highest level (2500 ng·d^{-1}) in the year. Higher levels and relatively higher TC/CC ratios were found in northern, eastern, and southern China, signifying a fresh use of chlordane in these regions (*11, 28, 36*).

Good correlations between TC and CC ($R^2 = 0.674$, $p < 0.01$), TC and TN ($R^2 = 0.958$, $p < 0.01$), CC and TN ($R^2 = 0.686$, $p < 0.01$). However, CN and heptachlor showed no significant correlation with other chlordane components.

TC/CC ratios <1 were widely reported in the atmosphere of the Arctic, Great Lakes, U.S., and UK, and they were considered to be indicative of inputs from "weathered" chlordane sources (*36*−*41*). The annual average TC/CC ratio in the atmosphere of the Chinese cities was 0.43 ± 0.27, delineating a dominant "weathered" secondary chlordane source in China. However, high TC/CC ratios >1 were also observed in summer and autumn in some eastern, coastal, and southern regions (see Table 1), indicating fresh use of chlordane in the cities. Around 400−800 tonnes per year of chlordane was used in China recently (*42*).

Low TC/CC Ratios in Winter to Spring. The TC/CC ratios in the Chinese cities displayed a significant seasonality, lower in winter (mean 0.25 ± 0.33) and spring (0.38 ± 0.68) and higher in summer (0.59 ± 0.4) and autumn (0.61 ± 0.53). This result is similar to the recent PAS campaign conducted in the Pear River Delta of South China (0.27 ± 0.04 and 0.79 ± 0.13 in winter and summer, respectively) (*28*). However, previous studies in the Arctic showed higher TC/CC in winter to spring and lower TC/CC in summer to autumn (*36*−*38*). We suggest that the low TC/CC ratios in winter may be used as a fingerprint/tracer of Chinese chlordane emission.

Theoretically, a lower TC/CC ratio may be expected in summertime, because TC is generally more susceptible to degradation than CC (*36, 38*). The low TC/CC in Chinese urban air from winter to spring may be due to the re-emission from "weathered" chlordane sources.

Emission of "weathered" chlordane from construction foundations in wintertime would be a potential secondary source. It is compulsory in China for house and building owners to take termite prevention measurements, in particular in cities. Chlordane was widely used as a preventive termiticide for construction foundations. A historic production of 9000 tonnes of chlordane was recorded in China. The annual chlordane production (843 tonnes) peaked in 1999 (*42*). A large amount of chlordane was used for termite control and treatment in urban construction foundations. In wintertime when there is little fresh chlordane application, emissions of "weathered" chlordanes characteristic of low TC/CC ratios are expected to dominate in the urban atmosphere. Therefore, it is suggested that the potential chlordane emissions from urban construction foundations need further investigation in the near future.

HCB. The HCB concentrations in the Chinese cities were in the range of 48.4−6510 pg·d^{-1} up to 2 orders of magnitude higher than those reported in Europe and North America (*12, 43*) (see Supporting Information Table S2). It seems that China is an important current global source region for HCB, based on our data.

HCB is highly persistent in the environment. The potential sources of HCB in developing countries is still not clear (*44*). HCB had been widely used as a fungicide, but, remarkably, it has never been registered as a pesticide in China. From

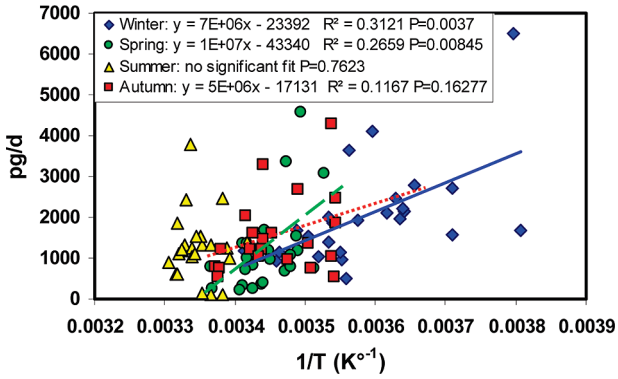

FIGURE 3. The relationship between atmospheric HCB levels and the reciprocal of temperature (K, seasonal average) in 37 Chinese cities in the year of 2005.

1988 to 2003 the total production was about 66 000 tonnes in China, of which >95% was used to produce pentachlorophenol (PCP) and Na−PCP (*42*). HCB can be released as a byproduct of chlorination processes such as pesticide production, as well as fuel combustion and waste incineration (*23, 44*−*46*). Application of PCP, Na−PCP, and chlorothalonil containing HCB residue (*44*) may also introduce HCB to the environment. It is noteworthy to point out that, according to our investigation, most of the chlorothalonil products on the market in China are imported/originated from the UK.

Unlike the relatively uniform distributions of HCB in the UK and North America (*12, 43*), the spatial patterns of HCB in China displayed distinctive regional and seasonal variations. A high H/L (highest/lowest level) ratio up to 135 denoted that there were obvious HCB primary emissions across China during all the four sampling seasons. In summer and autumn, relatively high HCB concentrations were observed in cities located in the central parts of China, and evaporation from HCB-containing PCP/Na-PCP for schistsome control (*13*) and chlorothalonil (*44*) may be the potential sources during these seasons.

However, in winter, the spatial distribution of HCB was similar to that of PAHs across China (*18*), with relatively high levels observed in North, Northwest and Southwest China (refer to Supporting Information Figure S4), where it is colder than other regions. In Figure 3 the observed HCB levels were plotted with the annual average air temperatures of the cities. While no significant spatial trend was found for summer and autumn samples, the colder cities did display higher atmospheric HCB levels than the warmer cities in winter and spring. HCB emissions from PCP/Na-PCP and chlorothalonil cannot explain this temperature-dependent distribution, as most of their application is in warmer South China and East China, and if it is in North China, in summer and autumn when there is no snow and vegetation grows. Higher wind speed can increase the PAS sampling rates (*47*). Although the wind speed in winter and spring was generally higher, the average wind speed in North China where higher HCB levels were observed was not significantly higher than other parts of China, in winter and spring in the campaign year (refer to Supporting Information Figure S2). The fact that no organochlorines other than HCB displayed such a trend also seems to exclude wind speed as a potential controlling factor here. We have reported quite similar seasonality and temperature-dependence of PAHs in the same batch of PAS samples, for which more fuel combustion and less combustion efficiency in colder cities in winter and spring was suggested to be responsible (*18*). Therefore, it is suggested that attention must be paid to combustion derived HCB in China in the context of the implementation of the Stockholm Convention, and further investigation is needed to determine the emission inventories of HCB from different combustion sources in China.

Endosulfans. Endosulfan concentrations (α-endosulfan ranged 0−1190 pg·d^{-1} and β-endosulfan ranged 0−422 pg·d^{-1}) were generally lower than those reported for North America, South America, Europe, and Africa by the GAPS project (*11*). Endosulfan was mainly used for controlling cotton bollworm in China, and geographically, the high concentrations of total endosulfan occurred in the cotton production areas in China as depicted in Supporting Information Figure S5.

Acknowledgments

The study was funded by National Natural Scientific Foundation of China (NSFC 40590391, 40673076, and 40518002). We thank all the friends and colleagues for their help in field sampling. G.Z. wishes to thank the U.K. Royal Society for supporting his work in LU. We are greatly in debt to the three anonymous reviews for their pertinent and constructive comments.

Supporting Information Available

Three tables showing the background information, concentrations of compounds at each sampling site and comparison data, and five figures depicting site locations, air back trajectories and wind field, and spatio-temporal distribution of TC, CC, HCB, and endosulfan. This material is available free of charge via the Internet at http://pubs.acs.org.

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ES802371N