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OVERVIEW

Potential sources of unintentionally produced PCB, HCB, and PeCBz in China: A preliminary overview

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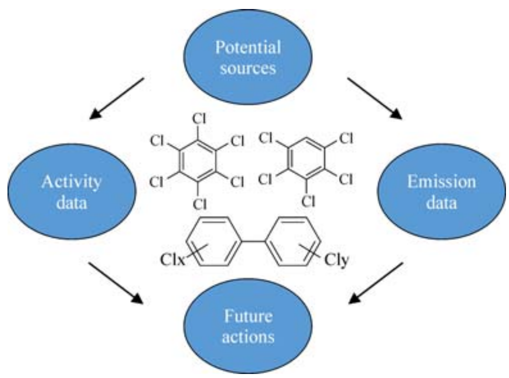
HIGHLIGHTS

- A preliminary analysis of potential sources for unintentionally produced PCB, HCB and PeCBz in China.
- Activity rates of sources for reference years from 2000 to 2015 provided.
- Emissions from a number of sources summarized and compared.
- Implications for future research and regulation discussed.

GRAPHIC ABSTRACT



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ABSTRACT

Under the Stockholm Convention on Persistent Organic Pollutants (POPs), China is required not only to reduce polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF) but also unintentionally produced polychlorinated biphenyls (PCB), hexachlorobenzene (HCB) and pentachlorobenzene (PeCBz). A baseline of the sources in China that generate these unintentional POPs is needed for both research and regulation purposes. In this paper, we have compiled production data of potential sources in China and assessed them in five-year intervals from 2000 to 2015. Most of these activities experienced changes from rapid growth to slow growth. Measured data for PCB, HCB and PeCBz in samples collected from potential sources in China were reviewed. Most information was associated to thermal processes with high potential of emission, including waste incineration and ferrous and non-ferrous metal production. In addition, high levels of PCB, HCB and PeCBz were found as impurities in a few chlorinated products or as by-products in solvent production, which suggested organochlorine industry might be important sources. Finally, based on the studies reviewed, recommendations for future actions in research and policy as well as a few regulatory issues in China are discussed.

1 Introduction

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/PCDF), polychlorinated biphenyls (PCB), and hexachlorobenzene (HCB) are listed in the annex C of the Stockholm Convention on Persistent Organic Pollutants (POPs) as initial unintentionally produced persistent organic pollutants in 2001. Since that, three more POPs were listed into annex C, namely: pentachlorobenzene (PeCBz) in 2009, polychlorinated naphthalenes (PCN) in 2015 (UNEP, 2015), and hexachlorobutadiene (HCBD) in 2017 (UNEP, 2017), so that presently, there are seven

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unintentional POPs. It shall be noted that five groups of POPs listed in annex C- PCB, HCB, PeCBz, PCN and HCBD- are also intentionally produced POPs and listed in the Convention's annex A. It shall be noted that the inventories according to article 5 of the Convention for POPs listed in annex C shall address only those POPs that are "unintentionally formed and released"; thus, transfers of these POPs from intentional production and use are not to be included in these release inventories.

Although in China, PCB and HCB production was completely stopped in the 1970s and in 2004, respectively, the concentrations in environmental media such as air and soil did not show a downward trend (Wang et al., 2010; Hogarh et al., 2012). Historic stockpiles of products and contaminated production sites may still discharge intentionally produced industrial PCB, HCB or PeCBz (Wang et al., 2010), but emissions from unintentional generation are likely to make significant contributions into the environment (Cui et al., 2013, 2015). For the more newly listed unintentional POPs, PCN and HCBD, such information is not yet systematically looked for or assessed and are not included in this study.

China conducted the first release inventory of PCDD/PCDF in 2007 based on activity data in 2004 (SEPA, 2007). The updated release inventory with a reference year of 2013 has been presented as a draft and is not yet published (RCEES, 2014). Both inventories have been developed using the source categorization and emission factors from the UNEP "Toolkit for Identification and Quantification of Releases of Dioxins and Furans" (UNEP, 2005, 2013) amended with nationally developed emission factors based on national measurements in China where appropriate.

It is generally believed that PCDD/PCDF releases are accompanied by releases of PCB, HCB and PeCBz due to similar formation mechanisms in thermal and other processes (Hedman et al., 2006; Pandelova et al., 2006; UNEP, 2013). Several thermal processes identified as sources of PCDD/PCDF, such as waste incineration, iron ore sintering, coking process, cement production, primary and secondary nonferrous smelting, thermal wire reclamation, open and domestic burning of coal, biomass and waste were also reported as sources of PCB, HCB and PeCBz (Lee et al., 2005; Grochowalski et al., 2007, Delepine et al., 2011; Zhang et al., 2011; Antunes et al., 2012; Black et al., 2012). In addition, PCB, HCB and PeCBz can also be formed during the manufacture process of chlorinated chemicals as impurities, by products or waste materials, such as impurities in pigments and pesticides, and wastes in chlorinated solvents production (USEPA, 1998; Weber et al., 2011; Takasuga et al., 2012). This group of sources had large uncertainties and variability in emission compared to the thermal processes, and differences in the scale of concentrations between PCDD/PCDF and the other three unintentional POPs have been observed. For example, dioxin like PCB (dl-PCB) are the major contaminants in pentachloronitrobenzene (PCNB) products contributing 50%–95% of the total toxic equivalent (TEQ) (Huang et al., 2015). In addition, PCNB (also known as Quintozene) degrades to PeCBz; thus, is also a source of PeCBz (Bailey et al., 2009).

China due to its activities and large population is the world's largest emitter of PCDD/PCDF as the dioxin inventory has shown (Fiedler, 2007; Wang et al., 2016). As a party to the Stockholm Convention on Persistent Organic Pollutants, China is required to identify and quantify the releases of all unintentional POPs listed in annex C, and develop action plans to minimize or eliminate them. So far, no systematic investigation has been undertaken about potential sources of unintentionally produced PCB, HCB and PeCBz nor have their releases been quantified in China. To this end, we reviewed the literature for information about unintentional PCB, HCB and PeCBz in China. This paper summarizes first, the sources of unintentional POPs identified in China; secondly, the national activities at 5-year intervals from 2000 to 2015; and thirdly, measured concentrations of these unintentional POPs in China. A synthesis of the available information provides recommendations for future researches and possible regulatory actions.

2 Materials and methods

A systematic literature research about potential source of unintentional PCB, HCB or PeCBz in China was undertaken in late 2016 consulting the following sources: Web of Science (webofknowledge.com), Google® Scholar (www.scholar.google.com) and the Organohalogen Compounds database (www.dioxin20xx.org). Publications and reports in Chinese language were collected by using an electronic search engine from China Knowledge Resource Integrated Database and the WANFANG Data of E-Resources. Information on source activity rates was taken from annual statistic yearbooks or online reports from industry associations or ministries in China as well as the World Bank online databases (data.worldbank.org). Besides, a few literature of sources related to the production of chlorinated products from other countries were also referred to since these products may be manufactured in China.

3 Results

3.1 Overview of sources

The potential sources from literatures were first compiled according to the reporting requirements under the Stockholm Convention using the source groups and subcategories as established in the UNEP Toolkit, version 2013 (UNEP, 2013) and the reporting format under article

15 of the Stockholm Convention. It turned out that all reported data from China about unintentional PCB, HCB or PeCBz were within the scope of source groups in the Toolkit, and only a few differences were seen in the sub-categories. To give an overview, Table 1 summarizes the general information about source identifications. A large proportion of sources in groups 1 and 2 were reported. For some groups, e.g., heat and power generation (source group 3) or transport (source group 5), there are no data at present.

3.2 Activity rates at national level

The activity data were reported on an annually basis. We have grouped them according to the periods of the five-year plans in China, the start and the end year of a plan

Table 1 Grouping of presence or absence of release data for PCB, HCB or PeCBz reported from activities in China according to the ten source groups (1–10) and source categories (a–k) in the UNEP Toolkit

Source	Group	Data from China found	No data from China found
1	Waste incineration	a-Municipal solid waste incineration; c-Medical waste incineration	b-Hazardous waste incineration; d-Light fraction shredder waste incineration; e-Sewage sludge incineration; f-Waste wood and waste biomass incineration; g-Animal carcasses burning
2	Ferrous and Non-Ferrous Metal Production	a-Iron ore sintering; b-Coke production; c-Iron and steel production plants and foundries; d-Copper production; e-Aluminum production; f-Lead production; g-Zinc production; i-Magnesium production; l-Thermal wire reclamation and e-waste recycling	h-Brass and bronze production; j-Thermal non-ferrous metal production (e.g., Ni); k-Shredders
3	Heat and Power Generation		a-Fossil fuel power plants b-Biomass power plants c-Landfill biogas combustion d-Household heating and cooking- Biomass e-Domesting heating- Fossil fuels
4	Production of Mineral Products	a-Cement kilns	b-Lime; c-Brick; d-Glass; e-Ceramics; f-Asphalt mixing; g-Oil shale processing
5	Transport		a-4-Stroke engines b-2-Stroke engines c-Diesel engines d-Heavy oil fired engines
6	Open Burning Processes	b-Waste burning and accidental fires	a- Biomass burning
7	Production and Use of Chemicals and Consumer Goods	d-Chlorinated aromatic chemicals (chloranil; 2,4-D products; PCNB; pigment)	a-Pulp and paper mills; b-Chlorinated inorganic chemicals c-Chlorinated aliphatic chemicals e-Other chlorinated and non-chlorinated chemicals; f-Petroleum refining; g-Textile plants; h-Leather plants
8	Miscellaneous		a-Drying of biomass b-Crematoria c-Smoke houses d-Dry cleaning e-Tobacco smoking
9	Disposal		a-Landfills, waste dumps and landfill mining b-Sewage/sewage treatment c-Open water dumping d-Composting e-Waste oil disposal

(*Continued*)

Source	Group	Data from China found	No data from China found
10	Contaminated Sites and Hotspots		a-Production sites of chlorine b-Production sites of chlorinated organics c-Application sites of PCDD/PCDF containing pesticides and chemicals d-Timber manufacture and treatment sites e-Textile and leather factories f-Use of PCB g-Use of chlorine for production of metals and inorganic chemicals h-Waste incinerators i-Metal industries j-Fire accidents k-Dredging of sediments and contaminated flood plains l-Dumps of wastes/residues from groups 1-9 m-Kaolin or ball clay sites

from 2000 to 2015; thus, for the years 2000, 2005, 2010, and 2015. In addition, activities were compiled for the years 2004 and 2013, the reference years of the China PCDD/PCDF release inventories (SEPA, 2007; RCEES, 2014). Table 2 shows the activity data for relevant sources at national level in China. Also shown in Table 2 are the gross domestic product (GDP) *per* capita and the population.

Most of these data refer to amounts of product manufactured in a given year (e.g., steel, sinter, cement), some are based on amounts of feed material processed (e. g., coal consumption for heating). For some sources such as transport and open burning, activity data were not readily available but could be derived from meta data. Using the base year of 2000, an increase trend for the period from 2000 to 2015, can be seen in all data. An exception is the destructed forest area, which decreased from 884×10^{-6} square meter in 2000 to 129×10^{-6} square meter in 2015. For the 15-year period, the output of secondary aluminum had the highest growth rate of 4170%, followed by the number of passenger vehicles (1807%) and output of brass and bronze products (1098%).

The GDP increased exponentially from 955 USD in 2000 to 7925 USD in 2015, while the increase of population was flat from 1267 million in 2000 to 1375 million in 2015. Different shapes of mostly increase were observed for different activities during the period 2000–2015. These trends have relevance as to the formation and release of unintentional POPs and future release inventories. Some activities have exponential increase, e.g., incinerated municipal waste, generated hazardous waste, sintering ore, coal consumed for power, passenger vehicles, brass and bronze product (Fig. 1(a)). It can be assumed that these sources may continue with sharp increases in the future. Linear trends were observed for crude steel production and magnesium production (Fig. 1 (b)), which may also suggest strong increases in the future. The sources with polynomial trends, all showed declines in the recent years, are shown in Fig. 1(c): e.g., production of pig iron, cement, glass and trucks, coal consumed for heating. Their future development is difficult to predict.

There are some interesting results when comparing primary and secondary metal production (Fig. 2). Both of them have increasing trends but the volumes of metals from primary production were much higher than from secondary production, especially for zinc and aluminum where primary production was 27- or 19-fold over secondary production in the year 2000; the ratios are lower for lead (10%) and copper (3%). In 2015, the primary production was still several times larger than the secondary production but less expressed (5.1% for aluminum, 2.5% for copper, and 2.2% for lead). Only the secondary zinc production remained at similar ratio (18%).

3.3 Concentrations of PCB, HCB and PeCBz from different sources

Quantitative information on releases of unintentional PCB, HCB or PeCBz in either stack emissions, solid residues, such as fly ashes, or in products were found in 27 references. The information was quite scattered and concentrated around stack emissions (total of 172 samples from 23 processes; in addition, raw gas concentrations from three plants) from the production of ferrous and non-ferrous metals, Source Group 2 in the UNEP Toolkit (125 samples from 16 processes). Less information was available from waste incinerators (34 stack samples from municipal solid waste incinerators and medical waste incinerators). In addition, three sets of results were available from a cement kiln. Most information was available for PCB; however, it shall be noted that reporting of PCB was not harmonized: some reported emissions as (i) sum of a number of PCB congeners (often 28 PCB congeners), (ii) sum of (certain) dioxin-like PCB, and (iii) dioxin-like PCB (dl-PCB) as toxic equivalents based on WHO-TEFs (WHO-TEQ_{PCB}) (van den Berg et al., 2006).

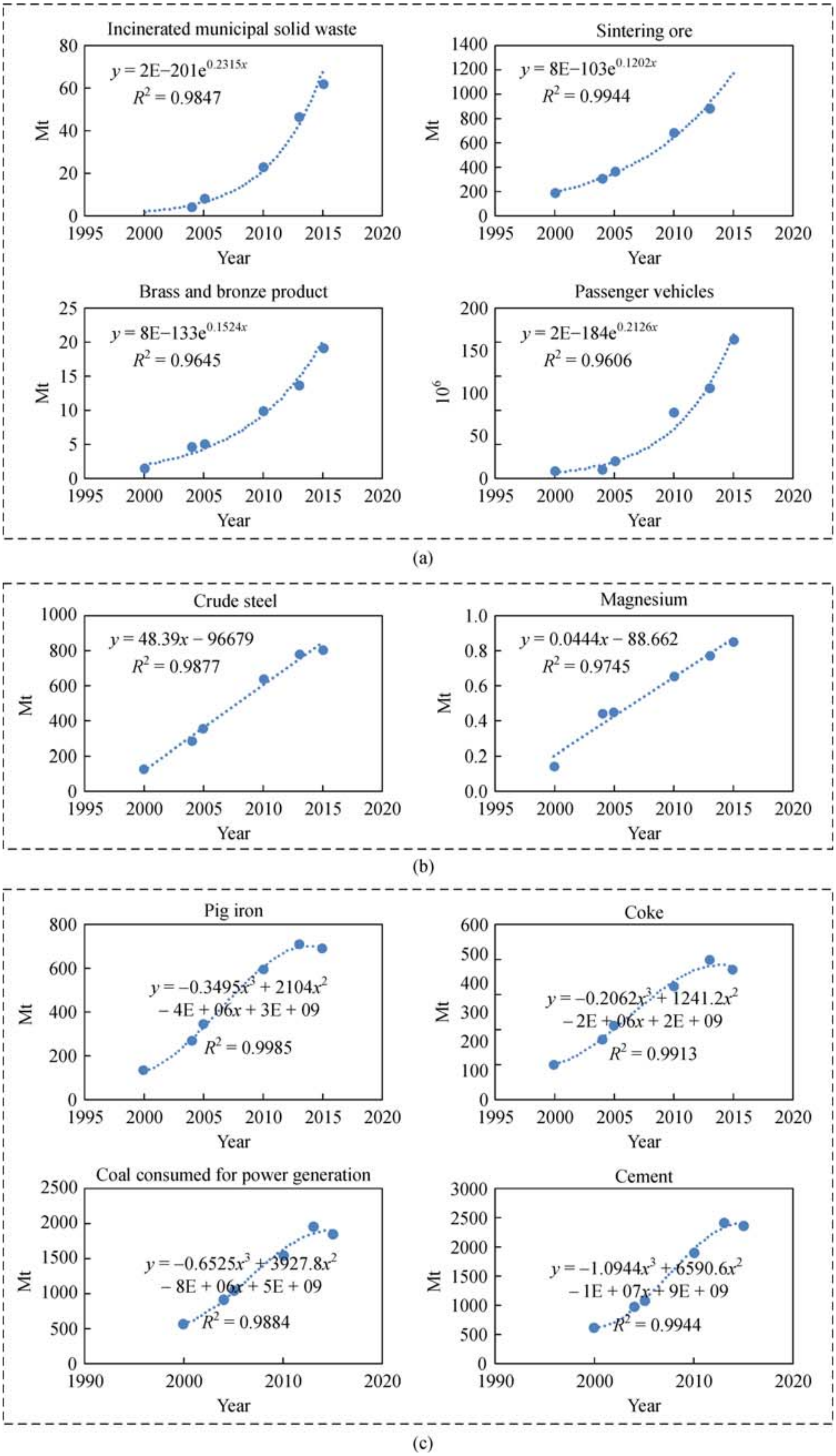

Fig. 1 Examples of increasing trends for activity data during the period from 2000 to 2015: (a) exponential, (b) linear, (c) polynomial

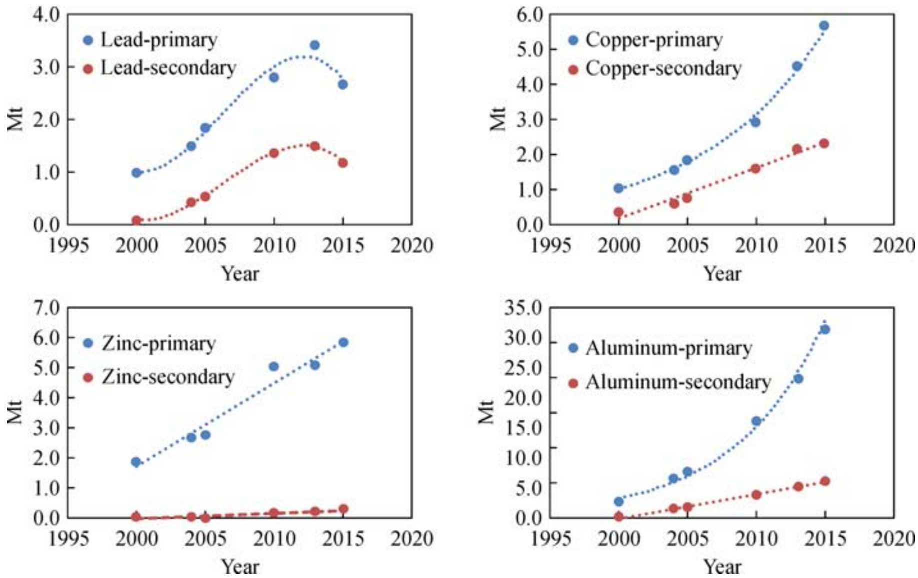

Fig. 2 Comparisons of production volumes (and increasing trends) between primary and secondary non-ferrous metal production

Data on solid residues were available from a total of 102 samples with the majority being for fly ash (86) and 16 for bottom ash. Once again, the majority from processes of the ferrous and non-ferrous metal production (69 data; 17 data for waste incineration). Only six results were found for bottom ashes.

For chlorinated chemicalś production, information on impurities of six products of dl-PCB, HCB and PeCBz from 42 samples was found, and information on carbon tetrachloride byproducts from eight samples was found.

3.3.1 Thermal processes

3.3.1.1 Stack emissions

Table 3 summarizes the concentrations of PCB, HCB and PeCBz in purified stack gas and raw gas from thermal processes in China. Best information was available for concentrations of dl-PCB reported as WHO-TEQ, using the TEF scheme of the World Health Organization as of 2005 (van den Berg et al., 2006); these results are shown in Fig. 3 for the sub-categories of the Source Groups 1, 2 and 4. As can be seen, most of the concentrations are lower than 0.1 ng TEQ/Nm^3. Secondary zinc plants had the highest mean concentration (2.8 ng TEQ/Nm^3), followed by thermal wire reclamation processes (1.4 ng TEQ/Nm^3). Far below 0.1 ng WHO_{2005}-TEQ/m^3 were converter steelmaking processes (0.0014 ng TEQ/Nm^3), secondary lead production (0.002 ng TEQ/Nm^3), cement kilns (0.008 ng TEQ/Nm^3), coking processes (0.008 ng TEQ/Nm^3) and municipal solid waste incinerator (0.009 ng TEQ/Nm^3).

Less information than for dl-PCB was available for HCB and PeCBz. Concentrations of HCB ranged from 0.18 ng/Nm^3 to 754 ng/Nm^3 in purified stack gas, with the highest concentration being found in the iron ore sintering process and the lowest mean concentration being found in coke production process. Concentrations of PeCBz in stack gas were slightly higher than HCB in each respective facility, which ranged from 0.2 to 1501 ng/Nm^3. The highest and lowest concentration was also found in iron ore sintering and coke production process, respectively. It should be stated, secondary copper production released the highest mean concentration of HCB (495 ng/Nm^3) and PeCBz (1181 ng/Nm^3) than other processes.

Assessing the technologies, it must be stated that most measurements were obtained from modern plants with good air pollution control systems (APCS) (Table 3). A few studies have measured concentrations in both raw gas and stack gas from waste incinerators (Table 3). As expected, the unintentional POPs in raw gases from waste incinerators had higher concentrations than after flue gas cleaning devices, such as activated carbon adsorbent or fabric filters (Chen et al., 2009; Yan et al., 2010; Li et al., 2016).

3.3.1.2 Solid residues

Less data than for stack emissions were available for solid residues. Table 4 summarizes the studies and the reported results of PCB, HCB and PeCBz concentrations in fly ash and bottom ash from various thermal processes. Most of these studies only reported concentrations of dl-PCB, which ranged from 0.005 ng/g to 115 ng/g and from 0.00003 ng TEQ/g to 32 ng TEQ/g. The highest concentration of dl-PCB (mean = 71.4 ng/g and 4.17 ng TEQ/g) was found in the fly ash samples from secondary copper production. Iron ore sintering processes also generated high concentrations of dl-PCB in fly ashes.

Table 2 Annual GDP *per* capita, population and activity rate of potential sources of unintentional POPs in China

SG[a]	Category	Unit	2000	2004	2005	2010	2013	2015	Source
	Population	million	1267	1300	1308	1341	1361	1375	China Statistical Yearbook[b]
	GDP *per* capita	$	955	1498	1740	4515	6992	7925	World Bank[c]
	a-incinerated municipal solid waste	Mt	NA	4.5	7.9	23.2	46.3	61.8	China Statistical Yearbook[b]
1	b-generated hazardous waste	Mt	8.3	10.0	11.6	15.9	31.6	39.8	China Statistical Yearbook[b]
	e-generated sewage sludge	Mt		14.0	11.1	21.2	26.4	30.2	China Environment Yearbook[d]
	a-sintering ore	Mt		304.5	369.2	688.2	886.6		China Steel Yearbook[e]
	b-coke	Mt	121.8	206.2	254.1	388.6	479.3	448.2	
	c-pig iron	Mt	131.0	268.3	343.8	597.3	709.0	691.4	China Statistical Yearbook[b]
	c-crude steel	Mt	128.5	282.9	353.2	637.2	779.0	803.8	
	d-copper-primary	Mt	1.0	1.6	1.8	2.9	4.5	5.7[f]	
	d-copper-secondary	Mt	0.3	0.6	0.7	1.6	2.2	2.3[f]	
2	e-aluminum-primary	Mt	2.8	6.7	7.8	16.2	23.2	31.3[f]	
	e-aluminum-secondary	Mt	0.1	1.7	1.9	4.0	5.3	6.4[g]	
	f-lead-primary	Mt	1.0	1.5	1.9	2.8	3.4	2.7[f]	The Yearbook of Nonferrous Metals Industry of China[h]
	f-lead-secondary	Mt	0.1	0.4	0.5	1.4	1.5	1.2[f]	
	g-zinc-primary	Mt	1.9	2.7	2.8	5.0	5.1	5.8[f]	
	g-zinc-secondary	Mt	0.1	0.0	0.0	0.2	0.2	0.3[f]	
	h-brass and bronze products	Mt	1.6	4.7	5.0	9.9	13.6	19.1[f]	
	i-magnesium	Mt	0.1	0.4	0.5	0.7	0.8	0.9[f]	
3	a-coal consumed for power generation	Mt	558	920	1033	1545	1952	1793	China Statistical Yearbook[b]
	e-coal consumed for heating	Mt	88	115	135	153	227	241	
4	a-cement kilns	Mt	597	967	1069	1882	2416	2359	
	d-glass	Mt	9	19	20	33	39	39	China Statistical Yearbook[b]
5	a-passenger vehicles	1.0E + 06	9	11	21	78	106	163	
	c-trucks	1.0E + 06	3	4	10	16	20	21	
	a-cereals	Mt	405	412	428	496	553	572	
6	a-sugarcane	Mt	68	90	87	111	128	117	China Statistical Yearbook[b]
	b-destructed forest area	1.0E + 06 m^2	884	1422	737	458	429	129	

Note: [a]source groups and sub-categories according to the Toolkit classification (UNEP, 2013); [b] (National Statistics Bureau, 2001, 2005, 2006, 2011, 2014, 2016); [c](The World Bank, 2017); [d](Li, 2016; Yang, 2006, 2011, 2014; Yu, 2005); [e](The Editorial Board of China Steel Yearbook, 2005, 2006, 2011, 2014); [f](China Nonferrous Metals Industry Association, 2016); [g] (CNMIABR, 2016);[h](China Nonferrous Metals Industry Association, 2001, 2005, 2006, 2011, 2014)

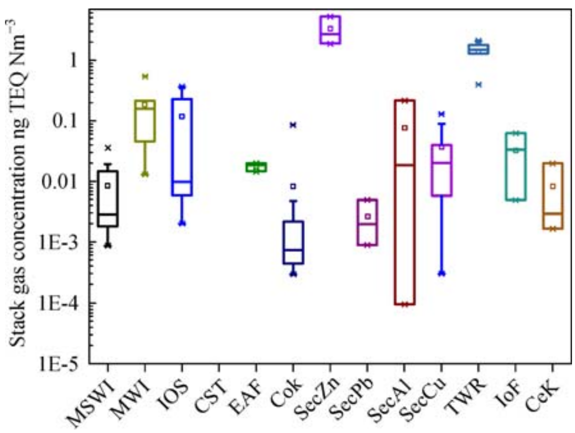

Fig. 3 Concentrations of dl-PCB in purified stack gas from thermal processes in China.
Detailed data are compiled in Table 3. Denotation: MSWI- municipal solid waste incinerator; MWI-medical waste incinerator; IOS-iron ore sintering; CST-converter steelmaking processes; EAF-electric arc furnace; Cok-coking process; SecZn-secondary zinc production; SecPb-secondary lead production; SecAl-secondary aluminum production; SecCu-secondary copper production; TWR-thermal wire reclamation; IoF-iron foundries; CeK-cement kilns

Thermal wire reclamation and medical waste incinerator facilities had high concentrations of dl-PCB in bottom ashes. Iron foundries, converter steelmaking processes and secondary lead production released low concentration of dl-PCB in ash samples.

HCB or PeCBz concentrations are comparable or sometimes lower than those of total PCB in the same ash samples. Concentrations of HCB in ash samples ranged from 0.2 to 87.5 ng/g. The highest concentration was encountered in medical waste incinerator fly ash samples. HCB in the fly ash samples from coking processes and magnesium metallurgy facilities were relatively low with mean values of 0.3 ng/g and 0.2 ng/g, respectively. Concentrations of PeCBz ranged from 0.15 to 51 ng/g in residues with thermal wire reclamation process had the highest concentration and coking process had the lowest. Primary and secondary copper metallurgy facilities also released high concentrations of PeCBz.

3.3.2 Production of chlorinated products

This section covers potential sources that unintentionally generate PCB, HCB and PeCBz as impurities or by-products or related production waste. Some information was available for PCB, HCB and PeCBz in chlorinated pesticides or dyestuff/pigments in China since it still produces quite large amounts of chlorinated chemicals, including aliphatic hydrocarbons (solvents, including chlorinated paraffins), dyestuffs and pigments (see Table 5). Most information was found for HCB impurities, which ranged from non-detect to more than 40,000 ng/g. For dl-PCB, there is also a wide range of concentrations in those chemicals (0.00006–4.2 ng TEQ/g). Less data are available for PeCBz, which ranged from 0.04 to 137 ng/g. One study reported high concentrations of PCB, HCB and PeCBz in the production of chlorinated methane (Zhang et al., 2015). It should be noted, concentrations in products can be also expressed as emission factors (EFs) and the UNEP toolkit has assigned HCB EFs for a few chlorinated products, such as tetrachlorophthalic acid (2000 μg/g), phthalocyanine pigments (1000–200000 ng/g) and chlorinated paraffins (7–8900 ng/g). Although the data sources as well as the origin of products were not clearly specified in the Toolkit, it make sense since China is known as one of the largest producer of these products in the world (UNEP, 2013).

4 Implications for research and regulation issue

This paper provides the first step for future release inventories and assesses emissions of unintentional PCB, HCB and PeCBz in China. Although some potential sources listed in the Toolkit or sources not covered by the Toolkit have not addressed, waste incineration, ferrous and non-ferrous production, cement and a few chemicals production of which the majority were listed in Annex C part II and part III of the Convention are covered in this compilation. Based on the overview of potential sources in this study and updated emission factors in our previous work (Gong et al., 2017a, 2017b), the national release inventories of PCB, HCB and PeCBz of China will be developed in our future work.

Since for emission inventories the activities have the same importance and weight as the emission factors to define the sectoral annual release, the activities in China have been assessed. It was found that the activities exhibited distinct but different patterns ranging from constant strong increases to flattening shape and even declines from 2013 (Fig. 1). The data demonstrate the underlying importance to define a reference year when reporting emission inventories and closely follow the economic and policy developments. For example, some sources might continue to experience strong growth in the future, such as the Chinese government has started initiatives to explore some pilot works of cement kilns co-processing solid waste in 2015 aim to promote its industrialization (MIITC, 2015). According to the thirteenth five-year (2016–2020) construction plan of municipal solid waste disposal facilities, the incineration ratio will be more than 50% by 2020, while in 2014 it was only 29% (NDRC, 2016). A 4% increase *per* year will be a conservative assumption for the incinerated amount in the next few years. In a view on resource management, it can be expected that the share of secondary metal production may increase in the future, most likely accompanied by less steep increases or inverted trends for the primary production. It shall also be noted that a change in

Table 3 PCB, HCB and PeCBz concentrations in stack gas samples from thermal processes in China

SG[a]	Category		Number of samples[b]	Concentration (ng/Nm^3)					APCS[c]	Reference
				PCB	dl-PCB	dl-PCB WHO TEQ	HCB	PeCBz		
1a	Municipal solid waste incinerator	Stack	6	9.9–129[d]	0.1–1.1	0.00087–0.0197	11.2–122.0	18.7–213	BF + SDS + AC	Li et al., 2016
		Raw	2	13.3–44.1[e]	0.93–1.8	0.0105–0.041	28.3–186	195–525	–	
1a	Municipal solid waste incinerator		8	1.34–20[d]					BF +/SDS +/AC +/ SNCR +/QF	Zhao et al., 2015
1a	Municipal solid waste incinerator		5	15.2–127.9[e]	0.55–5.77	0.0023–0.0368			BF +/SDS +/AC + NIDS/+/QF	Liu et al., 2013b
1a	Municipal solid waste incinerator	Stack	9				1.3–11.6	3.9–14.3	not specified	Yan et al., 2010
		Raw	6				51–9594	164–5751	–	
1c	Medical waste incinerator		3	26.5–124[e]	2.1–18.8	0.0133–0.214			BF +/SDS +/AC +/ QF	Liu et al., 2013b
1c	Medical waste incinerator	Stack	3	138–855[d]	3.5–41.0	0.046–0.549			AC + SDS	Chen et al., 2009
		Raw	2	1728–3192[d]	104–192	1.247–2.510			–	
2a	Iron ore sintering		4		12.8–19.1	0.23–0.37	136.2–754.3	763.5–1501.5	ESP	Tian et al., 2012
2a	Iron ore sintering		9	1.56–58.9[e]	0.145–10.44	0.002–0.214			ESP	Liu et al., 2013b
2c	Electric arc furnace		3	159.3–288.1[e]	2.35–5.5	0.015–0.02			BF	Liu et al., 2013b
2c	Iron foundries		5		0.967–20.065	0.05–0.063			BF + WS	Lv et al., 2011a
2c	Converter steelmaking processes		12		0.036–0.75	0.00003–0.00285			BF	Li et al., 2014
2b	Coking process		9			0.0004–0.0877			BF	Liu et al., 2009a
2b	Coking process		9		0.0987–4.648	0.0003–0.0048	0.182–0.816	0.209–0.661	BF	Liu et al., 2009b
2g	Secondary zinc production		3			1.9–5.28			not specified	Ba et al., 2009a
2f	Secondary lead production		3			0.0009–0.005			not specified	Ba et al., 2009a
2e	Secondary aluminum production		< 30[f]			0.0001–0.215			BF +/cyclone	Ba et al., 2009b
2d	Secondary copper production		< 30[f]			0.0003–0.126			BF +/cyclone	Ba et al., 2009b
2d	Primary copper production		9		2.6–30.9		19.6–447	27.6–1035	ESP +/BF	Nie et al., 2012a
2d	Secondary copper production		6		8.1–28.1		441–550	989–1373	BF	Nie et al., 2012a
2d	Secondary copper production		12		0.31–11.6	0.003–0.13			BF[g]	Hu et al., 2013
2j	Thermal wire reclamation		5		187–982	0.4–2.1	41.3–144	103–354	Water-cooling[g]	Nie et al., 2012b
2i	Magnesium metallurgy facilities		9		0.1265–3.78		2.27–22.9	3.21–37.0	ESP/BF	Nie et al., 2011
4a	Cement kilns		3	44.7–143.5[e]	0.44–2.64	0.0017–0.002			BF	Liu et al., 2013b

Note: [a] source group and sub-categories according to Toolkit classification (UNEP, 2013); [b] if the number of samples was not provided; then the number of data was used; [c] BF: bag filter units; SDS: semi-dry scrubber, AC: activated carbon injection, SNCR: selective non-catalytic reduction; ESP: electrostatic precipitator; NIDS novel integrated desulphurization system; QF: quenching facility; WS: wet scrubber; [d] monoCBs to decaCB were analyzed, but the detail of congeners was not provided; [e] triCBs to decaCB were analyzed, but the detail of congeners was not provided; [f] 30 samples in total; [g] One plant was not equipped with any APCS

Table 4 PCB, HCB and PeCBz concentrations in ash samples from thermal processes in China

SG[a]	Category	Number of sample size[b]	concentration (ng/g)					Reference
			PCB	dl-PCB	dl-PCB WHO_{2005}-TEQ	HCB	PeCBz	
	fly ash							
1a	Municipal solid waste incinerator	8	0.524–6.9[c]					Zhao, et al., 2015
1a	Municipal solid waste incinerator	5	1.6–3.4[c]	0.016–1.6	0.00024–0.034	43.1–47.9[d]	9.15–11.6[d]	Li et al., 2016
1c	Medical waste incinerator	4		0.69–15.6	0.012–0.39	0.45–87.5		Yan et al., 2011
2b	Coking process	14		0.26	0.00026	0.29	0.146	Liu et al., 2013a
2a	Iron ore sintering process	4		3.6–38.8	0.2–0.82	8.6–36.7	8.6–26.5	Tian et al., 2012
2c	Iron foundries	13		0.02–8.42	0.00003–0.01			Lv et al., 2011a
2c	Converter steelmaking processes	5		0.005–0.18	0.00003–0.001			Li et al., 2014
2d	Primary copper production	3		0.3–5.7		5.4–5.8	11–14.2	Nie et al., 2012a
2d	Secondary copper production	2		27.7–115		4.4–10.8	12.5–17.1	Nie et al., 2012a
2d	Secondary copper production	<20[e]			0.0003–32.4			Ba et al., 2009b
2e	Secondary aluminum production	<20[e]			0.003–2.3			Ba et al., 2009b
2g	Secondary zinc production	2			0.0329–0.1121			Ba et al., 2009a
2f	Secondary lead production	2			0.00007–0.0003			Ba et al., 2009a
2i	Magnesium metallurgy facilities	4		0.0246–0.054		0.191–0.278	0.273–0.404	Nie et al., 2011
	Bottom ash							
1a	Municipal solid waste incinerator	6	1.00–1.31[f]	0.08–0.52	0.00006–0.0015			Shen et al., 2010
1c	Medical waste incinerator[g]	4		0.0099–38.8	0.003–0.94	5.05–74.8		Yan et al., 2011
2j	Thermal wire reclamation[h]	2		13.6–25.3	0.008–0.2	16.5–23	10.7–50.9	Nie et al., 2012b
2c	Hot dip galvanizing[i]	3		0.233–0.743				Lv et al., 2011b
2c	Iron foundries[j]	1		0.007	0.00001			Lv et al., 2011a

Note: [a] Source group and sub-categories according to Toolkit classification (UNEP, 2013); [b] if the number of samples was not provided; then the number of data was used; [c] monoCB to decaCB were analyzed, but the detail of congeners was not provided; [d] two samples; [e] 20 samples in total; [f] 28 PCB congeners: 8, 18, 28, 44, 52, 66, 77, 81, 101, 105, 114, 118, 123, 126, 128, 138, 153, 156, 157, 167, 169, 170, 180, 187, 189, 195, 206 and 209; [g] the bottom ash was collected at the exit of the slag remover; [h] the ash was collected on the bottom of the furnaces after combustion; [i] the ash was collected on the ground surface around the zinc bath; [j] the ash was collected on the ground surface

Table 5 Concentrations of dl-PCB, HCB and PeCBz in products or by-products in China

Products	Number of sample size	Concentration in product (ng/g)			Reference
		dl-PCB WHO_{2005}-TEQ	HCB	PeCBz	
Chloranil	3	0.0026 (0.0019–0.0033)	32.6	136.6	Liu et al., 2012
2,4-D products	5	0.0219 (0.00006–0.108)	1.7 (ND-2.9)	13.9 (0.37–63.3)	Liu et al., 2013c
PCNB	5	1.5 (0.68–2.5)	31 (3.7–52)	0.2 (0.04–0.3)	Huang et al., 2014
Picloram	3	–	1150 (727–1527)	–	Lu, 2007
Chlorothalonil	1	–	42200	–	Wang et al., 2005
Chlorothalonil	1	–	39900	–	Xiang, 2008
Pigment	24	0.00002–4.21	–	–	Shang et al., 2014
Carbon tetrachloride	8	3.77–1170[a]	–	5950–38800	Zhang et al., 2015

Note: [a] Total of PCB (ng/g)

technologies may be seen with increasing and especially new capacities: For example, the small-sized sintering units (<90 m^2) or small electric arc furnace steelmaking facilities were phased out by 2013 in China (NDRC, 2013). The older and smaller facilities tend to have a higher potential of unintentional POPs release than larger and modern plants. For those have strong increase trend in the future, it will be important that modern technologies be applied to cover the future demand but also to minimize unintentional POPs releases.

Although the contamination levels of PCB, HCB or PeCBz were found in quite a few thermal processes in China, there is still a lack of data for some sources. Since here are no emission standards for PCB, HCB and PeCBz in stack gas in China and there is no Chinese standard for the sampling and analysis of PCB, HCB and PeCBz in flue gas or residues, it is unlikely that many new data will be generated outside of research projects. Some of the thermal processes have to comply with the national emission limits for PCDD/PCDF in stack gas, which is set at with 0.1 ng TEQ/Nm^3, 0.5 ng TEQ/Nm^3, and 1.0 ng TEQ/Nm^3 for municipal solid waste incinerators, hazardous waste incinerators, secondary non-ferrous industries (copper, aluminum, zinc and lead), respectively (SEPA, 2001a, 2001b; 2015). In addition, The standard HJ 77.2-2008 addresses the 2,3,7,8-substituted PCDD/PCDF in flue gas only but does not include dl-PCB (SEPA, 2008). It is recommended to have measured data for the total TEQ, *i.e.* including 17 PCDD/PCDF and 12 dl-PCB. In general, all emission data have to be taken with care since neither the sampling nor the analytical methods have been validated beyond individual laboratories. Only if harmonization of sampling and analytical methods have been agreed, correlations between these compounds can be established.

Less information is available for PCB, HCB or PeCBz generated as impurities in products. In contrast to the PCDD/PCDF, a lot of interest is with the contamination of PCB, HCB or PeCBz. Huang et al. (2014) highlighted, the TEQ concentrations of dl-PCB were higher when compared to PCDD/PCDF in the Chinese PCNB products. In two chlorinated pesticides- chlorothalonil and picloram- high concentrations of HCB were found. These two chemicals are not listed in the UNEP Toolkit and the PCDD/PCDF content therein is not known. There may be other chemical products that have chlorination steps in their production or chlorine in the final molecule to be added. Some products contaminated with HCB are not yet listed in the Toolkit and should be included, such as the production process of dyestuffs having 3,4,5,6-tetrachlorophthalic anhydride/acid as an intermediate. As for control measures, only chlorothalonil has a national standard of HCB impurity with limit of 0.04% (GAQSIQ, 1999), which corresponds to 400,000 ng/g. The two published results (Wang et al., 2005; Xiang, 2008) did not exceed this limit.

Experiences from other countries highlighted HCB in waste and residues during organochlorine productions, such as tetrachloroethylene, trichloroethylene, tetrachloromethane and dichloroethylene. When the wastes or residues had been disposed for some time, highly HCB contaminated sites might be formed (Mumma et al., 1975; Markovec et al., 1984; Weber et al., 2011). However, there are a range of information gaps on PCB, HCB or PeCBz in waste and residues in the chemical industry in China. The only study addressed unintentional POPs generated during the methanol-based production of chlorinated methanes in China (Zhang et al., 2015) indicated the need to evaluate PCB, HCB or PeCBz content and release from organochlorine industry as well as the historic deposits and contaminated sites in the future.

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