Potential source and emission analysis of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in China

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ABSTRACT

The Chinese Government ratified the Stockholm Convention on Persistent Organic Pollutants (POPs) on 25 June 2004. As a party to the Convention, China is preparing its National Implementation Plan (NIP), which must be finished and submitted to the Conference of the Parties before November 11, 2006. The emission inventory and strategy for reducing unintentionally formed products is a very complex and difficult part of the NIP. In this paper, we analyzed the potential sources of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in China and calculated their possible emissions. We included municipal solid waste incineration, hazardous and medical waste incineration, pulp bleaching, sodium pentachlorophenolate production and usage, illegal electrical and electronic waste (“e-waste”) recycling, and ferrous and non-ferrous metal production, and prepared a preliminary analysis of dioxin emissions in China. The results show that ferrous metal production (104.2–6648.3 g TEQ), non-ferrous metal production (2.6–3389.8 g TEQ), and sodium pentachlorophenolate production and usage (120–368.0 g TEQ) should be major potential sources of dioxins and furans. Moreover, municipal solid waste and hazardous and medical waste incineration (35.5 g TEQ) should also be considered high priority sources owing to a rapidly increasing incineration ratio anticipated for the near future.

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1. Introduction

The Chinese Government signed the Stockholm Convention on Persistent Organic Pollutants (POPs) on May 23, 2001 and ratified it on 25 June 2004. China must submit its National Implementation Plan (NIP) to the Conference of the Parties before November 11, 2006. A project entitled “Building the Capacity of the People's Republic of China to Implement the Stockholm Convention on POPs and Develop a National Implementation Plan” was endorsed by the Global Environment Facility and initiated in September 2004. Fig. 1 shows the objective and structure of the NIP project. There are four primary areas of POPs inventory and strategy development: pesticides, stockpiles, polychlorinated biphenyls (PCBs), and unintentionally produced POPs. The last category, especially for the polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs), is very difficult and complex to address in China because it covers so many industries around the entire country.

Dioxin emissions from municipal solid waste incineration (MSWI), sodium pentachlorophenolate (PCP-Na) production and usage, and pulp bleaching were among the first emission issues identified as concerns and studied in China (Bao et al., 1995; Zheng

et al., 1997a; Tian and Ouyang, 2003). Based on the available information and the experiences of developed countries, hazardous and medical waste incineration, illegal electrical and electronic waste (“e-waste”) recycling, and ferrous and non-ferrous metal production are also considered important PCDD/F sources because of their huge volumes and high emission factors (Jun et al., 2004). Therefore, we analyzed all the above sources and their possible emissions in this preliminary study.

2. Waste Incineration

Potential dioxin emission from waste incineration is a topic of great public concern in China. The State Environmental Protection Administration (SEPA) of China has already issued three national standards (SEPA, 2006c) to restrict dioxin emissions from municipal waste, medical waste, and hazardous waste incineration facilities.

2.1. Municipal solid waste Incineration

Until 2003, landfill was still the leading municipal waste disposal method in China, and the incineration ratio was only 4.90% (Ministry of Construction, China, 2004). Most Chinese MSWI facilities are located in eastern and southern provinces, for example in

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Fig. 1. The objectives and structure of the national implementation plan in China.

Shanghai, Zhejiang, and Guangdong. There are two main types of incinerators in China: grate furnaces and fluidized bed furnaces. In total, about 63% of incinerated waste was burned by grate furnaces and 26% was combusted in fluidized bed incinerators now (Wang et al., 2005). Recent studies have shown that the dominant 2,3,7,8 congeners detected were OCDD and 1,2,3,4,6,7,8-HpCDD, followed by 1,2,3,4,6,7,8-HpCDF and OCDF in the flue gases and fly ash of Chinese MSWI facilities (Jin et al., 2003; He et al., 2004). Research has shown that large incinerators in China can likely meet the national standard for dioxin emissions (1 ng TEQ Nm^{-3}) (Li et al., 2001; Zhang et al., 2004).

Based on the increasing trend for use of municipal waste generation from 1993 to 2003 (Ministry of Construction, China, 2004), the estimated amount of total waste generated in China will reach $8.7 \times 10^7 \text{ t}$ in 2006, $9.8 \times 10^7 \text{ t}$ in 2010, and $1.2 \times 10^8 \text{ t}$ in 2015. If the incineration ratios of 5%, 7.5%, and 10% can be assumed in those years, PCDD/F emissions from MSWI facilities will be about 26.2, 44.1, and 66.9 g I-TEQ, on the condition that all the incinerators meet the national emission standard.

2.2. Hazardous and medical waste Incineration

Incineration technologies for flammable hazardous and medical wastes have high priority in the National Program for Hazardous and Medical Waste Treatment Facility Construction Plan ("the Program") (SEPA, 2006b). However, China still lacks the capacity to safely dispose of its hazardous and medical wastes, and as a result large amounts of hazardous waste have been stored temporarily. Only Shenzhen, Tianjin, Shenyang, and Hangzhou have centralized hazardous waste treatment facilities. The volume of hazardous waste has increased rapidly as the average annual growth rate was greater than 30% from 1999 to 2003. About $3.7 \times 10^6 \text{ t}$ of hazardous waste was disposed of in 2003 (SEPA, 2006a).

Considering the large amount of hazardous waste accumulated during these years, a rate of increase of 5% per year will be a safe assumption for the disposal amount in the near future. The amount will be $4.3 \times 10^6 \text{ t}$ in 2006, $5.3 \times 10^6 \text{ t}$ in 2010 and $6.7 \times 10^6 \text{ t}$ in 2015. According to the Program, a hazardous waste incineration ratio of 20% was recently achieved. Assuming that all those incinerators can meet the national emission standards, and that the average flue gas rate is $10,000 \text{ Nm}^3 \text{ t}^{-1}$ waste, the estimated volumes of dioxins emitted from hazardous waste incineration in 2006, 2010, and 2015 will be approximately 4.3, 5.3, and 6.7 g I-TEQ, respectively.

About $6.5 \times 10^5 \text{ t}$ of medical waste was produced in China in 2002, and this quantity is predicted to reach $6.8 \times 10^5 \text{ t}$ in 2010 (SEPA, 2006b). The Program provides that each medium-sized city will have at least one regional medical waste incinerator by 2006. Based on this growth ratio, the estimated annual PCDD/F emissions from medical waste incineration will be about 5.0, 5.1, and 5.3 g I-TEQ in 2006, 2010, and 2015, respectively, assuming that all medical waste is incinerated and that the average flue gas rate is $15,000 \text{ Nm}^3 \text{ t}^{-1}$ waste (UNEP Chemicals, 2005).

The total estimated PCDD/F emissions from Chinese municipal, hazardous, and medical waste incineration combined will be approximately 35.5, 43.5, and 45.4 g TEQ y^{-1} in 2006, 2010, and 2015, respectively. Based on these calculations, PCDD/F emissions from waste incineration will not be so high if all Chinese incinerators meet the national emission standards. However, the actual situation may differ, as shown by the results of a preliminary investigation of MSWI dioxin emissions in 2001 (Tian and Ouyang, 2003). Tian and Ouyang found that emissions from half of the small incinerators exceeded the national standard ($1.0 \text{ ng I-TEQ m}^{-3}$). If the PCDD/Fs collected in the fly ash are also included, the total emissions from waste incineration will be higher than the above estimates.

Table 1
PCDD/F levels in pulp and wastewater samples from pulp mills in China

Process	Chlorine	Extraction	Hypochlorite	Product	Total effluent
Pulp (pg I-TEQ g ⁻¹) (Zheng et al., 2001a,b)	10.3	12.9	9.5	24.7	–
Waste water (pg I-TEQ l ⁻¹) (Zhang et al., 2002)	253.3	85.2–315.6	9.5 (Zheng et al., 1997a)	–	10.11

3. Pulp production

China has a very large pulp and paper industry, which has been considered a large source of PCDD/F emissions since the mid-1990s (Zheng et al., 1997a; Zhang et al., 2000). A typical pulp bleaching sequence includes chlorine, alkaline extraction, and hypochlorite stages, all of which produce PCDD/Fs (Zheng et al., 1996) (Table 1).

Fig. 2 shows the 2,3,7,8 – substituted congener pattern of PCDD/Fs in the various stages of pulp production. The dominant congeners in the chlorine and hypochlorite stages were OCDD and 1,2,3,7,8-PeCDD while the sequence was 2,3,7,8-TCDD, OCDD, 2,3,7,8-TCDF and OCDF in the extraction stage. The PCDD/F concentration in the pulp product was approximately 39.5 pg I-TEQ g⁻¹ (Zheng et al., 2001a,b) representing a value greater than (8.24 pg I-TEQ g⁻¹) reported by the United States Environmental Protection Agency (USEPA) Dioxin Database (USEPA, 2001). The wastewater effluent concentration (10.11 pg I-TEQ L⁻¹) however was lower than the value (56.02 pg I-TEQ L⁻¹) reported in the USEPA database.

The Chinese pulp and paper industries recently discharged more than 1.0×10^7 t y⁻¹ of wastewater annually (SEPA, 2006a). Based on the dioxin concentration in the wastewater effluent as

shown in table 1, the total PCDD/F concentration from pulp will be more than 30 g I-TEQ y⁻¹.

4. Organochlorine pesticides and chemical production

Organochlorine pesticides, such as dichloro-diphenyl-trichloro-ethane (DDT) for pest and disease control and PCP-Na for the control of blood flukes (the source of schistosomiasis) and for wood preservation, were the leading pesticides produced and used from the 1950s to the 1970s in China.

4.1. PCP-Na

High concentrations of OCDD, OCDF, and 1,2,3,4,6,7,8-HpCDD were found in PCP-Na products, ranging from 30 to 92 ng I-TEQ g⁻¹ (Schechter et al., 1994; Bao et al., 1995). This range approached PCDD/F concentrations in Japanese PCP-Na produced by the hexachlorobenzene method (Seike et al., 2003). As for the congener pattern OCDD was the predominant congener, (OCDD > 1,2,3,4,6,7,8-HpCDD > OCDF > 1,2,3,4,6,7,8-HpCDD). This plan differed from that of the PCP-Na produced in Japan.

Because Na-PCP has been widely produced and used principally to control snailborne schistosomiasis in China, residual dioxins in the chemical have been released into the environment and have contributed notably to human exposure in areas where Na-PCP was used (Schechter et al., 1996; Hong et al., 2005). High PCDD/F concentrations were detected in sediment samples from Dongting Lake (Zheng et al., 1997b) and the Pearl River, where PCP-Na was sprayed (Zheng et al., 2001b). The concentrations of PCDD/Fs ranged from 128.3 to 326.9 pg I-TEQ g⁻¹ in the sediment samples. Moreover, high PCDD/F concentrations were also detected in breast milk and blood samples collected around the PCP-Na exposure areas (Schechter et al., 1994). Fig. 3 shows the PCDD/F congener patterns of the sediment samples reported. The patterns of the

Fig. 2. PCDD/F congener patterns in pulp samples at different stages of pulp production.

Fig. 3. PCDD/F congener concentrations detected in Dongting Lake and Pearl River sediment samples.

2,3,7,8-substituted PCDD/F congeners shown in Fig. 3 were very similar to those of PCP-Na samples (Zheng et al., 2003), as OCDD and 1,2,3,4,6,7,8-HpCDD were the dominant 2,3,7,8-substituted congeners found in the sediment.

China produced about 4000 t of PCP-Na annually in recent years (Jun et al., 2004). Based on the PCDD/F concentrations mentioned above, these quantities of PCP-Na resulted in production of approximately 120–368 g I-TEQ of dioxins in each of those years, entering the environment, and creating a great hazard to humans and other living organisms.

4.2. Other chlorine pesticides and chemicals

High concentrations of PCDD/Fs were also found in sediment samples collected near chlorine pesticide and chemical plants, indicating that the wastewater and sludge discharged from the plants might also contain some PCDD/Fs. Fig. 4 shows the PCDD/F 2,3,7,8-substituted congener patterns and concentrations in sediment samples collected near the wastewater discharges of several pesticide and chlorine alkali plants (Wu et al., 1997; Xu et al., 2000; Luksemburg et al., 2001; Hu et al., 2005). Although the average PCDD/F concentration in the effluent was not very high, the large volume produced makes it an important concern.

Ya-Er Lake is located downstream from a pesticide plant. A maximum PCDD/F concentration of 964 pg I-TEQ g⁻¹ was detected in sediment samples collected from the lake (Wu et al., 1997); this concentration is two orders of magnitude higher than the average value for the Yangtze River (Luksemburg et al., 1996). OCDD accounted for more than half of the total PCDD/Fs, and total PCDFs accounted for less than 20% of the total amount of PCDD/Fs in the sediment samples of Ya-Er Lake (Wu et al., 1997). High concentrations of PCDD/Fs were also detected in the Dagu Canal (Luksem-

burg et al., 2001) and Nanpaiwu River (Hu et al., 2005) in Tianjin. The largest pesticide plant in China that produces DDT, PCP, and HCB is located there. The plant's wastewater discharges via smaller ditches to Dagu Canal and then to Nanpaiwu River that empties into the Pacific Ocean at Bohai Bay. Moreover, the high PCDD/F concentration (28 pg I-TEQ g⁻¹) in sediments collected near the Beijing Gaobeidian wastewater treatment plant (Luksemburg et al., 1996), which receives wastewater from several industries, also indicated that other chemical industries were important potential PCDD/F sources in China.

5. Illegal e-waste recycling

Pollution from illegal e-waste recycling is a special problem in China and other developing Asian countries. High concentrations of PCDD/Fs were detected in e-waste samples collected near the open burning sites in the town of Guiyu, Guangdong Province (Fig. 5) (Luksemburg et al., 2002). PCDD/F concentrations in samples collected from the blackened e-waste and burning ash were as high as 469 and 5678 pg I-TEQ g⁻¹, respectively. PCDD/F concentrations in river sediments collected near this e-waste recycling site were 21–39,000 pg I-TEQ g⁻¹ (Luksemburg et al., 2002). The congener patterns of these sediments were quite different from those of the Yangtze River, which had very high HpCDFs, PeCDFs, TCDD, and TCDF, and relatively lower OCDD, similar to the e-waste and ash. Therefore, the PCDD/Fs detected in the local river sediments may have been derived from the illegal e-waste treatment. The results discussed above suggest that e-waste recycling may be an important source of PCDD/Fs in some areas of Guangdong and Zhejiang provinces, where large amounts of e-waste were illegally recycled recently.

Fig. 4. PCDD/F congener concentrations detected in sediments collected near the discharges of pesticide and chlorine alkali plants.

Fig. 5. PCDD/F congener concentrations in e-waste, ash, and sediment samples collected in Guiyu, Guangdong province.

6. Ferrous and non-ferrous metal production

China is one of the world's largest producers of ferrous and non-ferrous metals. Based on the large quantities of metals produced in

2002 in combination with emission factors given in the UNEP PCDD/Fs Toolkit (UNEP Chemicals, 2005), we estimated the potential PCDD/F emissions from metal production (Table 2). We found that ferrous metal production (104.2–6648.3 g TEQ) and non-

Table 2

Potential PCDD/F annual releases from ferrous and non-ferrous production in China, 2002

Subcategories	Production ($\times 10^4$ t y $^{-1}$)	Emission factor ($\mu\text{g TEQ t}^{-1}$)			Subtotal (g TEQ)
		Air	Water	Residues	
Iron ore sintering	21755	0.3–20	–	0.003	65.9–4351.6
Coke production	10073	0.3–3	0.06	–	36.3–308.2
Iron and steel production plants and foundries	19735	0.01–10	–	0–15	2.0–1988.5
Copper	163	0.01–50	–	0–630	0.01–1108.4
Aluminum	451	0.3–35	–	0–400	1.4–1961.8
Lead	123	0.5–80	–	0–5	0.6–104.6
Zinc	215	0.3–100	–	0	0.6–215
Total	106.8–10023.2				

ferrous metal production (2.6–3389.8 g TEQ) are very large potential sources of PCDD/Fs.

7. Conclusion

This preliminary study analyzed potential PCDD/F emissions from municipal, hazardous, and medical waste incineration; pulp bleaching; PCP-Na production and usage; illegal e-waste recycling; and ferrous and non-ferrous metal production in China. By calculating the potentials emissions from those industries, we found that ferrous and non-ferrous metal production and PCP-Na production and usage should have higher priority than other sources, unlike the developed countries. Moreover, municipal solid waste, hazardous and medical waste incineration should also be considered high priority sources owing to the rapidly increasing use of incineration anticipated for the future. Illegal e-waste recycling was also a large potential source of PCDD/F emissions. Moreover, we estimated that PCDD/F releases from waste incinerators and ferrous and non-ferrous metal production are increasing because such incineration and production are increasing without distinct improvement of PCDD/F reduction technologies.

It must be pointed out that notable uncertainties exist in the above estimation, because until now the ferrous and non-ferrous production industries in China did not sample, analyze, or report PCDD/F content in their industrial discharges. Also, it is very difficult to estimate the amount of illegal e-waste recycling that is taking place across the country. For most of the potential emission sources of PCDD/Fs, monitoring projects are urgently needed in order to determine the exact extent of PCDD/F emissions and take effective measures to control dioxin pollution in the near future.

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