An inventory of Potential PCDD and PCDF Emission Sources in the mainland of China

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Introduction

Polychlorinated dibenzo-p-dioxin (PCDD) and polychlorinated dibenzofurans (PCDF) are widespread environmental pollutants. A number of countries have developed national inventories of PCDD/F emission, such as USA, EU Nations and Japan [1]. However, due to the lack of PCDD/F data measured in China and the uncertain nature of the documentation available on emission factors, the report on inventories of dioxin emission is absent. With the municipal population growth, economic development and living-standard improvement, China faces many severe environment issues including potential problems related to PCDD/F. The country is aware of potential dioxin sources such as: incineration, iron and steel industry, chemical industry, fires, coal power plant, foundries, PCB in capacitors and transformers, sintering, traffic emission. In 2001, China signed the Stockholm Convention on Persistent Organic Pollutants in Stockholm. Therefore, there is a need for information regarding dioxin emission from these sources for taking actions to reduce and/or eliminate the release of dioxins in China, and reduce human exposure. In this study, we identify those potential PCDD/F emission sources and work out the first inventory on PCDD/F emission into the environment in China.

Methods

The dioxin inventory is developed according to an approach used by most of the agencies, such as USEPA.
Emission of source (g I-TEQ/yr)
= Emission factor (g I-TEQ/kg) × "Activity rate" (kg/yr or t/yr) (1)
The PCDD/PCDF emission per year is given in grams I-TEQ per year. According to equation (1), the annual flux is calculated by multiplying the release of PCDD/PCDF (*e.g.* in µg I-TEQ) per unit of feed material processed or produced (*e.g.* ton or liter) with the amount of feed material processed or produced (ton per year).

Results and discussion

Various quantities of PCDD/F released from different sources into the environment are estimated. The resulting PCDD/F emission inventory is presented in Table 1. The total emissions to the environment range from 7144 to 13575 g I-TEQ/yr, in which the central estimate of 2773 g I-TEQ PCDD/F emits into atmosphere. The large margins as the overall uncertainty of the inventoried emission estimates are indicated in this study. Comparison of the central estimate of PCDD/F annual fluxes into air (g I-TEQ/yr) between a few countries [1] and China is shown in Figure 1. Details of PCDD/F emission from major sources are discussed as followed.

Municipal Solid Waste incinerators: China produces about 29% of the world's municipal solid waste (MSW) each year, and with the economy continuing to grow rapidly, it is clear that China bears what may be the heaviest MSW management burden in the world. Over 1 billion people in China generate in excess of 200 million tons household solid waste each year. According to our analysis, the rate of MSW incineration is about 2%. In China, incineration of MSW has been technically proven as an effective waste treatment approach. Therefore, many big- and medium-size cities in China have constructed, are in the processing of constructing and/or are planning to

Figure 1: PCDD/F annual fluxes into air (g I-TEQ/yr); reference year 1995; For France, reference 1998.

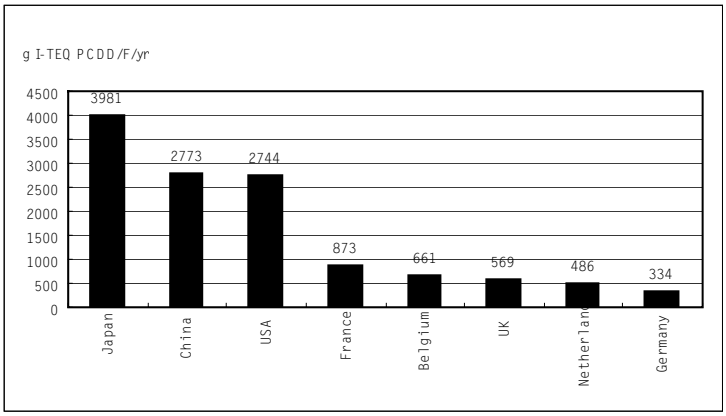

Table1: PCDD/F emission inventory in China; reference year 2002

Process	Emission Factor (μg I-TEQ ton $^{-1}$)	Estimated National quantity produced/utilized	Emission (g I-TEQ/yr)
Municipal Solid Waste incinerators	0.8-231	4 ×10^6 tons	3.20-924
Cement kilns	0.29	725 ×10^6 tons	210.25
Coal Power Plants	0.087	576.88×10^6 tons	50.19
Residential coal combustion	2.1-9.3	7.8×10^6 tons	16.38-72.52
Coke production	0.3	142.8×10^6 tons	42.84
Sinter Plant	1.2-9.0	120×10^6 tons	144-1080
Iron and steel	0.7-10	200×10^6 tons	127.4-1820
Non-ferrous metal	5-35	10.12×10^6 tons	50.6~354.2
Asphalt mixing	0.014	220×10^6 tons	3.08
Crematoria	2.4-80(per body)	4.15(million cremations)	9.96-332
Chloralkali Industry (electrode sludge)	21650	0.220~0.305 (reference 1998)	4400-6600
Pentachlorophenol/phenate	142.92 ng I-TEQ/g	4000 tons	571.7
Bleached chemical wood pulp and paper mills	315.6 ng I-TEQ/ (m^3 wastewater)	4800 (million m^3)	1514.88
Total			~ 7144-13575 g I-TEQ/yr

install MSW incineration devices [2]. Therefore, the ratio of incineration of MSW will increase rapidly in the near future. China EPA has enacted PCDD/F emission standard from waste incinerator. The limit value of 1.0 ng I-TEQ/m³ for municipal waste incinerators and 0.5 ng I-TEQ/m³ for hazardous waste incinerators are enacted. This limit is less stringent than 0.1 ng I-TEQ/m³ regulated by the developed countries such as USA, UK, and Japan [2]. Even so, most of existing municipal waste incinerators in China could not reach this standard request. Under such a background, an emission factor of 0.8~231 μg I-TEQ ton $^{-1}$ (Eduljee,1996)[3] is used because MSW incinerators in China are divided into two categories: new plant and old plant. Thus, the estimated PCDD/F annual emission is 3.20~924 g I-TEQ. However, to apply such an emission factor with great span to estimate the PCDD/F emission from all facilities seems debatable. Additionally, it should be noted that the annual PCDD/F emission quantity from this sector would be unavoidable to increase with growing number of MSW incinerators in the near future.

Cement kilns: In this study, PCDD/F released from cement kilns were also calculated. By using USEPA's PCDD/F emission factor of 0.29 ng I-TEQ/kg of clinkers produced (USEPA, 1998) [4] and annual cement production of 725 million tons in 2002 [5], the PCDD/F emission of this sector were 210.25 g I-TEQ yr^{-1}.

Coal Power Plants: China is the second largest energy consumer in the world following the United States. China's electricity is generated overwhelmingly by coal (about 70%). Because of China's extensive domestic coal resources and its wish to minimize dependence on foreign energy sources, it is expected that coal will remain the main energy source for electricity generation of China for the foreseeable future. In China, there are no reports on PCDD/F emission from coal power plant. In this study, we applied an emission factor of 0.087 ng I-TEQ/kg of coal consumed for this source (USEPA, 1998) [4]. In 2001, 576.88 million tons of coals were used for power generation in China [5]. By using USEPA's emission factor, we estimated the PCDD/F annual emission of this sector to be 50.19 g I-TEQ, which accounts for 0.40%~0.72% of the total emission.

Residential coal combustion: A total of 7.8 million tons coal was consumed by residents in 2001. Of this quantity, 13% was lignite, 12% was anthracite and 75% was bituminous. Edulijee[3] assumed the following emission factors: 2.1 µg I-TEQ /tons for anthracite coal and 5.7-9.3 µg I-TEQ /tons for bituminous. In this study, we applied 2.1-9.3 µg I-TEQ /tons as emission factor for this sector. Thus, 16.38-72.52 g I-TEQ PCDD/F was released into air annually. However, there is currently no data on PCDD/F emission from other combustion facilities using coal, such as industrial boiler or furnace. This may result in a highly uncertain estimation for coal combustion source category.

Coke production: Edulijee[3] reported an emission factor of 0.3 µg I-TEQ/ ton of coke produced. Production statistic for the China indicated that 142.8 million tons were produced in 2002 [5]. Thus, base on these data, the PCDD/F annual emission from this source was 42.84 g I-TEQ.

Sinter plant: PCDD/F emission factor range of 1.2-9.0 µg I-TEQ/ ton are available from the UK [3]. The quantity of sinter material produced in 2002 was 120 million tons [5]. Based on these data, PCDD/F emission from this source was 144-1080 g I-TEQ per annum.

Iron and Steel: China has taken the first place for crude steel production for consecutive eight years. Crude steel production in China reached 182 million tons in 2002. It is foreseeable that metal processing represents the most significant primary measured source sector in China at the present time. By process, the production of converter steel was 151.58 million tons, accounting for 83.22% of the total crude steel output, the production of electric arc furnace steel was 30.49 million tons, 16.73% of the total, and the steel output of open hearth furnace and other furnace was 83.1 thousand tons, taking up only 0.05% of the total [5]. However, PCDD/F emission standard for electric furnace has not been enacted in China. Tysklind[6] reported emission factors ranging from 0.2 to 20 µg I-TEQ ton^{-1} for electric furnaces, relating to emission after the bag-house. USEPA estimated an emission factor of 1.15 ng/kg for electric furnaces [4]. Eduljee[3] took a range 0.7~10 µg I-TEQ ton^{-1}, as an emission factor, representative of no chlorine and high chlorine operational conditions. Since the PCDD/F emission may vary considerably with the technology applied, we took 0.7~10 µg I-TEQ ton^{-1} as emission factor for crude steel production. According to 182 million tons steel were produced in 2002, 127.4~1820 g I-TEQ PCDD/F was released into environment.

The data suggest that the release from ferrous metal processing category accounts for large portion of the dioxin emission in China. Meanwhile, the large data gaps existed for this sector show that the emission estimate had quite a high degree of uncertainty. Since the uncertainty of the whole PCDD/F inventory is highly relative to this sector, there is an urgent need to obtain reliable PCDD/F data from iron and steel plant.

Non-ferrous metal: In China, despite a lack of appropriate PCDD/F data to estimate the PCDD/F emission, we try to provide an order of magnitude indication of PCDD/F emission from this industry sector in this study due to enormous output. The total production of 10 kinds of nonferrous metals in China has already occupied the second place in the world for 6 years in succession. The quantity of ten kinds of non-ferrous metal was 10.12 million tons in 2002 [5]. Eduljee[3] estimated a common emission factor range of 5-35 µg I-TEQ ton^{-1} for all non-ferrous metals thermal operations. In this study, we applied the same emission factor without relation to the actual technology and estimated 50.6~354.2 g I-TEQ emission from this sector per year.

Asphalt mixing: China production statistic indicated that 220 million tons asphalts were produced in 2002 [5]. 3.08 g I-TEQ PCDD/F was released into the environment with an emission factor of 14 ng I-TEQ/ton product [7].

Crematoria: China Ministry of Civil Affairs reported that 4.15 millions cremations were undertaken in 2002 [5]. The cremating rate was 50.6%. An emission factor range of 2.4-80 µg I-TEQ $body^{-1}$ was assumed for this study [3]. We estimated the PCDD/F emission of this category to be 9.96-332 g I-TEQ/yr.

Chloralkali Industry: China is one of major Chloralkali Industry production country in the world. There are about 300 Chloralkali enterprises in China, their productivity rank second in the world. In 1998, the quantity of alkali was 5.08 million tons. Wu [8] investigated that the PCDD/F level in electrode sludge of one Chloralkali enterprise was 21.65µg I-TEQ/kg. Generally, 40~60 kg electrode sludge will be produced as one ton alkali made. According to these data, 4.40~6.60 kg I-TEQ PCDD/F will be released into the environment without any treatment, which accounts for over 50% of the total PCDD/F emission.

These data indicated that Chloralkali Industry contributed substantial amounts of PCDD/F emission in the environment than expected, possibly due to aged process or inadequate pollution control devices of these facilities. Therefore, implementation of advanced industrial technologies and PCDD/F abatement technology must be applied and will help to reduce total amount of PCDD/F emission in China.

Pentachlorophenol/phenate (PCP/PCP-Na): In China, the output of PCP is approximately 4000 tons every year as wood preservative and insecticide for killing snail-borne schistosomiasis. Substantial amounts of sodium pentachlorophenol (Na-PCP) salts have been sprayed in certain areas of central China since the 1960s for control of snail-borne schistosomiasis. The level of PCDD/Fs in the PCP was 142.92 ng I-TEQ/g [9]. According to this level, there are 571.7 g I-TEQ PCDD/F emission into soil and water every year. Schecter et al. [10] determined PCDD/F in sediment samples and human blood and milk samples collected from a district where snail-borne schistosomiasis is prevalent. The PCDD/F level in the blood from exposed persons was 9 ~ 16.3 pg

I-TEQ /g lipid, while the concentration in blood samples from the general population was 4.8 ~6.4 pg I-TEQ /g lipid. Although human PCDD and PCDF tissue levels in China were low compared with those in samples from more industrialized countries, the higher levels in exposed persons were cause for concern.

Bleached chemical wood pulp and paper mills: In China, the output of paper production was 29.6 million tons in 1999. China is the largest producer of non-wooden paper pulp in the world. Non-wood plant fibers such as cereal, rice and reeds are extensively used as raw materials for paper production in China. The production of non-wood pulp was 16.0 million tons in 1998. Differences in bleaching processes for pulp are crucial for PCDD/F formation. In USA or some advanced countries, elemental chlorine-free bleaching processes had been required by laws to replace traditionally chlorine-based one in order to reduce PCDD/F formation. However, bleaching of non-wood plant fibers in China usually requires a single step of hypochlorite bleaching process. Zheng [11,12] investigated the level of PCDD/F (33.5 ~43.9 pg I-TEQ/g dry pulp) in pulp from five bleached kraft pulp and paper mills. At the same time, Zheng studied the PCDD/F level of the wastewater form bleaching process was 315.6 ng I-TEQ/ m^3. At present, China has enacted the discharge standard of wastewater pollutants for paper industry. The highest wastewater discharge standard for non-wood pulp bleaching is 300 m^3/ton. Thus, we assumed that the total 4.8 billions m^3 wastewater were discharged from non-wood pulp mills. Based on these data, the annual PCDD/F emission into water is 1.51 kg I-TEQ. However, with the advanced bleaching technology employed and wastewater treatment, the PCDD/PCDF contamination in effluents, products, and sludge from this sector will be reduced.

PCBs: Approximately eight thousand tons of PCBs were produced in China during the 1960's and 1970's. The PCBs production was banned in the late 1970s. The typical models of Chinese commercial PCBs are #l PCB (the chlorine content is 42%, which is similar to Aroclor 1242) and #2PCB (the chlorine content is 56%, which is similar to Aroclor 1254). Most of these PCBs products were used as dielectric fluid in electronic capacitors, and a small part of which were used as additives in paints. Only very low level of OCDD was detected in #1PCB, whereas the high level of PCDF resulted in TEQ value of 217 ng I-TEQ/g. No PCDD were detected in #2PCB, but elevated levels of PCDF congeners generated 417 ng I-TEQ/g. In order to dispose of PCBs, an experimental PCBs incinerator has been manufactured in China. The level of PCDD/Fs is 47.23 ng TEQ/g in stack ash and 87.86 pg TEQ/g in bottom ash [13,14]. Although there were PCDD/F emission from used PCBs leakage and PCBs incinerator annually, this inventory presented in this study didn't cover this sector.

Trichlorobenzene: Trichlorobenzenes (TCB) are used as solvents in industry and are intermediates in the production of several chemicals. TCB are also components of the dielectric fluids used in transformers and capacitors. The manufacturing enterprises of TCB are distributed in Hebei, Jiangsu, Hunan and Liaoning provinces of China. The production technology of TCB is relatively backward. Wu [8] investigated the content of PCDD/F in waste residues is up to 10%. UNEP [1] reported the PCDD/F concentration in TCB products was 0.023 µg I-TEQ/kg. Nevertheless, the quantity of PCDD/F emission from this sector wasn't estimated in the inventory because no data were available regarding production of TCB in China.

Other sources: Uncontrolled combustion activities, such as wood combustion, fallen leaves burning, festival firecracker and harvest-and-burn at some tillage, are still prevalent in urban and rural area in China. In addition, there are 16.09 million automobiles running on the road with an increase rate of 16% per year. Most of the engines are equipped with very limited pollution control devices and leaded gasoline is not completely prohibited in China. Contribution of PCDD/F from these sources is unknown. Future work is required to assess and quantify PCDD/F emission from these sources.

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