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A preliminary compilation and evaluation of a comprehensive emission inventory for polychlorinated biphenyls in China

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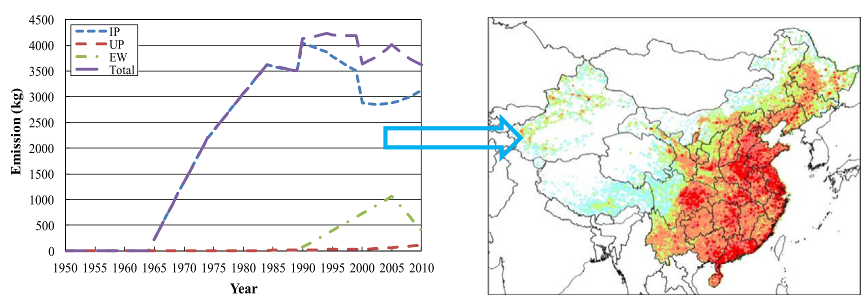
HIGHLIGHTS

- The first comprehensive gridded PCB emission inventories including IP-, UP-, and EW-PCBs.
- The IP-indicator PCB emissions in China were estimated at 130.1 t from 1965 to 2010.
- The UP-indicator PCB emissions in China were estimated at 8.56 t from 1950 to 2010.
- The total EW-PCB emissions in China were estimated at 103.5 t from 1990 to 2010.
- Modeled IP-PCB concentrations significantly correlated with monitoring data at urban sites.

GRAPHICAL ABSTRACT



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ABSTRACT

Emission inventories for polychlorinated biphenyls (PCBs) are crucial input data for atmospheric transport modeling and for the study of source–receptor relationships and the environmental behavior of these chemicals. Three types of primary PCB sources are considered in this study: intentionally produced PCBs (IP-PCBs), unintentionally produced PCBs (UP-PCBs), and PCB emissions from two e-waste sites (EW-PCBs). This study presents the historical emissions of all IP-, UP- and EW-PCBs into the air in China and the gridded Chinese emission inventories at a resolution of 1/6° latitude × 1/4° longitude from 1950 to 2010. The UP-TPCB emissions from 1950 to 2010 were re-estimated to be 8.56 t from eight emission sources comprising 96.3% of the Chinese UP-TPCB emissions. The EW-TPCB emissions from 1990 to 2010 were estimated to be 103.5 t, of which 7.1 t and 12.3 t were EW-PCB28 and EW-7PCB congeners (i.e., indicator-PCB28, 52, 101, 118, 138, 153, 180), respectively. The IP-PCB28 and IP-7PCB congener emissions from 1965 to 2010 were estimated to be 57.4 t and 130.1 t, respectively. A significant correlation was found between congener PCB28 and 7PCBs ($R^2 = 0.988$ and $P = 0.000$), which suggests that PCB28 is a good marker congener for describing the emission trends of all 7PCB emission sources. The gridded emission data were compared with published measured atmospheric concentrations for 2004 and 2008, and a significant correlation was found between the modeled emissions and monitoring data. To our knowledge, this study presents the first comprehensive gridded emission inventories that include all IP-, UP-, and EW-PCBs on a national scale.

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1. Introduction

Polychlorinated biphenyls (PCBs), which are synthetic, chlorinated, organic chemicals, are one of the twelve persistent organic pollutants (POPs) originally targeted by the Stockholm Convention (UNEP, 2001). These chemicals exhibit extraordinary stability, high toxicity, extremely high long-range atmospheric transportability, and the potential to bioaccumulate in the food chain, all of which threaten human health and environmental ecosystems (Tian et al., 2013).

POPs may be emitted from primary and secondary sources. *Primary emission* into the atmosphere occurs from direct sources of POPs. After PCBs were banned, reservoirs in the soil, sediment, and water continued to emit these chemicals to the environment as *secondary emissions*. Emission inventories for PCBs from primary sources are important input data for models of atmospheric transport and source receptors for studying the fate of POPs in the environment. In this study, *three general categories of primary PCB emissions* to the atmosphere are identified for China: primary emissions of intentionally produced PCBs (*IP-PCBs*), emissions of unintentionally produced PCBs (*UP-PCBs*), and emissions from discarded electrical and electronic equipment (e-waste) at two Chinese e-waste sites (*EW-PCBs*). EW-PCBs cannot be easily classified as IP- or UP-PCBs because they are from imported discarded products containing PCBs and can release PCBs when subjected to waste treatment processes (e.g., dismantling and open burning). Thus, EW-PCBs were classified as a third category of primary emissions because they can include both IP-PCBs and UP-PCBs. The reduction of UP-PCB emissions was addressed under the Stockholm Convention on Persistent Organic Pollutants, Annex C (unintentional production) (UNEP, www.pops.int). EW-PCBs are emitted due to uncontrolled discarding or inappropriate waste management/recycling, which generates significant amounts of PCB-containing hazardous materials (UNEP, 2009).

Among the emission inventories for the three different PCB sources defined above, those for IP-PCBs have been studied much longer. For example, Breivik and coworkers pioneered the study on global emission inventories for IP-PCBs more than 10 years ago and estimated that the global IP-PCB emissions of 22 selected PCB congeners for 1930–2010 were 7709 t, with a range of 440 to 91 722 t (Breivik et al., 2002b, 2007).

Meanwhile, the study of the emission inventories for UP- and EW-PCBs began only recently, which is important because the production of IP-PCBs was banned worldwide many years ago. Thus, the emissions of UP- and EW-PCBs have become more important. Our previous study (Cui et al., 2013) estimated the historical annual emissions of UP-PCBs and compiled the first gridded emission inventories for UP-PCBs in China for 1950–2010.

Recent studies have revealed high concentrations of PCBs at e-waste recycling sites, particularly in some developing countries, such as China, India, Pakistan, and Vietnam (UNEP, 2005), where the volumes of e-waste generated domestically and from material illegally imported from developed countries are rapidly increasing (Wong et al., 2007; Breivik et al., 2011, 2014). Emissions of PCBs from EW-PCBs in China have become a new source of PCB emissions that cannot be ignored. These emissions may not entirely account for the potential increase in atmospheric PCBs across China, but their influence could be very important in the area where dismantling is performed and surrounding regions, particularly in the river–sea boundary zone, which may be the potential source of PCBs to the world's oceans, including coastal areas (i.e., seawater and sediment) (Yang et al., 2012).

The objectives of this study are to compile the emissions from the IP-PCBs using a methodology similar to that used by Breivik et al. (2002a,b); to re-estimate the UP-PCB emissions using the emission factors newly published by Liu and coworkers (2013); to calculate the PCB emissions from the two major e-waste sites in China; and to produce gridded PCB emission inventories for all three sources (IP-, UP-, and EW-PCBs) for China with a resolution of 1/6° latitude × 1/4° longitude (24 × 24 km^2) from 1950 to 2010. In the near future, we will use these gridded emission inventories as input to our newly developed model, the Gridded Emission and Residue Model for Industrial POPs (GERM-InPOPs), to study the environmental behavior of PCBs in China, including their transfer and transport in different matrices, their secondary emissions, and their fate in the Chinese environment.

2. Methods and input data

2.1. Gridded IP-PCB emission inventories

To create IP-PCB emission inventories for China, we first compiled IP-PCB usage data (Zhang et al., 2010) and then calculated the emissions of PCBs from these IP-PCBs that were used in China with the help of the modified emission factors that were originally introduced by Breivik et al. (2002a,b). Breivik's research provides usage, disposal, and accidental factors for PCB congeners, but these emission factors have limitations. For example, the emission factor for usage only considered a temperature of 20 °C, and those for landfills, disposal and accidents only considered temperatures of 5, 10, and 20 °C. To better assess the amount of emissions for PCB congeners, we recompiled these factors as functions of temperature. The gridded temperature data from 1950 to 2010 were obtained by interpolating objectively analyzed daily data from the National Centers for Environmental Prediction (NCEP) reanalysis at standard atmospheric pressure.

2.1.1. Usage inventories

PCBs were manufactured in China for 10 years from 1965 until 1974, when they were banned. During this time, approximately 10 000 t of PCBs was produced (~0.8% of the total global PCB production), of which 9000 t was trichlorobiphenyl, known as Number 1 PCB, and 1000 t was pentachlorobiphenyl, known as Number 2 PCB (Xing et al., 2005). Trichlorobiphenyl was used primarily in electrical capacitors and transformers, while pentachlorobiphenyl was used mainly as a paint additive (China SEPA, 2003). We therefore assumed that 1000 t of PCBs was used annually for 10 years from 1965 to 1974 (Xing et al., 2005), as shown in Fig. A1 of the Supporting Information (SI), along with the usage reported in other sources (Jiang et al., 1997). The gridded Chinese PCB usage inventory for 1965–1974 at a resolution of 1/6° latitude × 1/4° longitude (Zhang et al., 2010) is presented in Fig. A2. This usage inventory was produced using the gridded population data at the same spatial resolution as a surrogate. Fig. A2 shows that the major usage of Chinese PCBs was in the eastern part of the country, which is host to highly developed industries and large populations.

2.1.2. Congener profile

The congener profile is important for calculating the amount of each congener in the PCBs used. We analyzed the Chinese transform oils and obtained the congener profile for the Number 1 PCB (trichlorobiphenyl) (Jiang et al., 2007; Zhang et al., 2010) (Fig. A3(a)), in which low-chlorinated biphenyls were the major components. For the Number 2 PCB (pentachlorobiphenyl), the congener profile of Aroclor1254 (see Fig. A3(b)) was used as a surrogate because the profile of this product is not available. Because the Chinese-produced PCBs consisted of 90% Number 1 PCB and 10% Number 2 PCB, the congener profile of the Chinese PCB production can be considered well represented by the profiles of Number 1 (Fig. A3(a)) and Number 2 PCBs (Fig. A3(b)) using the production-weighted factors (0.9 for Number 1 and 0.1 for Number 2 PCBs). The results of this calculation are shown in Fig. A3(c). 7PCB congeners PCB28, 52, 101, 118, 138, 153, and 180 are called indicator PCB congeners because they can represent the characteristics of PCBs in the environment. For the 7PCB congeners, the annual usages from 1965 to 1974 were 37.5 t for PCB28, 36.7 t for PCB 52, 9.5 t for PCB101, 15.3 t for PCB118, 7.4 t for PCB138, 3.6 t for PCB153, and 1.7 t for PCB180.

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