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Short-chain chlorinated paraffins in the soils of two different Chinese cities: Occurrence, homologue patterns and vertical migration

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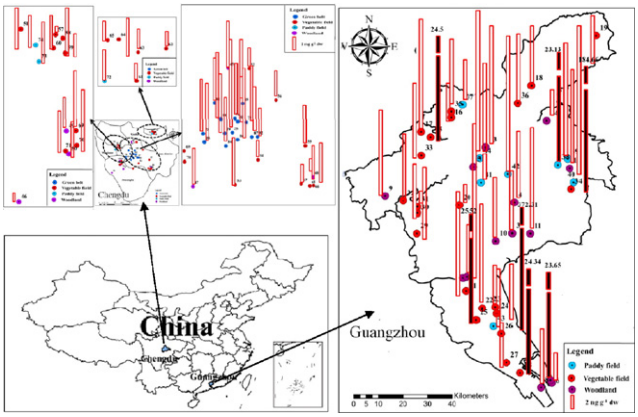
HIGHLIGHTS

- SCCP levels in topsoils from Guangzhou and Chengdu were relatively lower.
- SCCP concentrations in topsoils from Chengdu were lower than those from Guangzhou.
- Atmospheric transport was an important pathway of SCCPs to topsoils.
- Cl_{5-6} SCCPs may be more prone to move to deep-layer soils than Cl_{9-10} SCCPs.
- SCCP levels displayed little dependence on organic matter for most topsoils.

GRAPHICAL ABSTRACT



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ABSTRACT

Short-chain chlorinated paraffins (SCCPs) are candidate persistent organic pollutants (POPs) that are under review by the Stockholm Convention. China is currently the largest producer and consumer of chlorinated paraffins (CPs). To study the environmental behavior and fate of SCCPs in the soils of urban and suburban regions, the SCCP concentrations in 88 topsoils and 15 soil columns from land of different use types (e.g., woodland, vegetable field, paddy field and greenbelt) from Guangzhou and Chengdu have been determined. The SCCP concentrations in topsoils from Guangzhou (range: 1.45–25.5 ng g^{-1} dry weight (dw), average: 10.3 ng g^{-1} dw) were much higher than those from Chengdu (range: 0.218–3.26 ng g^{-1} dw, average: 1.43 ng g^{-1} dw). When compared to previously reported SCCP levels for topsoils from other areas, the SCCP concentrations measured in the present work were quite low. Much higher SCCP concentrations were observed in the greenbelt topsoils from Chengdu relative to the values measured from woodlands and vegetable and paddy fields. The composition profiles suggest that $C_{10}Cl_{6-10}$ and $C_{11-13}Cl_{6-8}$ were the major groups of SCCPs in topsoils from the woodlands and vegetable and paddy fields in Guangzhou and Chengdu. Vertical variations of the SCCP concentrations in the soil columns suggest that less chlorinated SCCPs (Cl_{5-6}-SCCPs) are more capable of migrating to the deeper-layer soils than more chlorinated ones (Cl_{9-10}-SCCPs). The SCCP concentrations displayed little dependence on organic matter (OM) for most topsoils ($p > 0.05$), indicating that OM is not the controlling factor in the distribution of SCCPs in the soils.

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Capsule: This study analyzed the occurrence, homologue patterns and vertical migration of SCCPs in the topsoils of two Chinese cities with different industrial structures and climate conditions.

1. Introduction

Short-chain chlorinated paraffins (SCCPs) are chlorinated paraffins (CP) with 10–13 carbon atoms (Bayen et al., 2006). Currently, SCCPs are listed as candidate persistent organic pollutants (POPs) under review by the Stockholm Convention because of their stability, bioaccumulation, long-range transport potential and toxic character (POPRC, 2015). SCCP mixtures have been extensively used in applications including metalworking lubricants, paints, adhesives, sealants, leather fat liquors, plastics, rubber, flame retardants, textiles and polymeric materials (POPRC, 2015). Further, the increasing demand for CPs has made China the world's largest producer and user of CPs, with the total CP production reaching 1,050,300 tons in 2013 (WCC, 2014). The major CP products in China are CP-42, CP-52 and CP-70 (Sun et al., 2005; Tang and Yao, 2005), with CP-52 accounting for nearly 90% of the total 2012 output (WCC, 2013). The SCCP mass fractions in CP-42, CP-52, and CP-70 were determined to be 3.7%, 24.9%, and 0.5%, respectively (Gao et al., 2012). Large quantities of SCCPs can be released into the environment during the production, storage, transportation and use of CPs (POPRC, 2015). It was reported that the total emission of SCCPs in China reached 1789 tons in 2011 and will increase to 2563 tons in 2016 in the absence of any measures to reduce SCCP emission, with applications in metalworking fluids, flame retardants and plasticizer being the major emission sources (Xu et al., 2014).

Soils are a principal receptor and environmental reservoir for many persistent semi-volatile organic contaminants (SVOC) (Ockenden et al., 2003; Li et al., 2006; Gevao et al., 2011; Wang et al., 2012). Similar to other POPs, SCCPs can also accumulate in soils (Zeng et al., 2011). However, the current limitations of analytical methods make scarce information on SCCP pollution and their evolution in soils (Nicholls et al., 2001; Zeng et al., 2011; Gao et al., 2012; X.-T. Wang et al., 2013; Y. Wang et al., 2013; Wang et al., 2014; Halse et al., 2015). A study conducted in the Liaohe River Basin, where CPs were industrially produced in large quantities, found that local industrial activity was the major source of soil SCCP contamination (Gao et al., 2012). Similar results were observed for SCCPs in soils from the Pearl River Delta area (Y. Wang et al., 2013). In addition to emissions from local industrial sources, releases from sewage treatment plants, sludge application and wastewater irrigation were also suggested to be important sources of SCCPs in soils (Zeng et al., 2011; Wang et al., 2014).

Guangzhou is the largest city in southern China, whereas Chengdu is the largest city in western China (Fig. 1). As the capital of Guangdong Province, Guangzhou is located in the Pearl River Delta (PRD), an area that is well known as an industrial base and manufacturing center. The gross product of Guangzhou reached 16,707 billion yuan in 2014, ranking third nationwide (Zeng, 2015). Leading industries in Guangzhou include automobile, electronic and petrochemical. A large amount of CPs may have been used in these industries in China, as CP products are largely used as metal cutting fluids, flame retardants and plasticizers (Xu et al., 2014). Guangzhou is subjected to the subtropical maritime monsoon climate, with the southeast and southwest monsoons prevailing in spring and summer and the northeast monsoon prevailing in autumn and winter (Chen et al., 2013a). Chengdu is the capital of Sichuang Province and is the most economically developed city in western China, with electronic information, pharmaceutical, aircraft, and tobacco being the main industries. In 2014, its gross product was 10,057 billion yuan (Yin, 2015). Chengdu is subjected to the subtropical humid monsoon climate. Although Chengdu is located in the monsoon region, the wind in Chengdu remains gentle throughout the year (CBS, 2011).

The different industrial structures between Guangzhou and Chengdu may lead to differences in the usage of CP products and cause further discrepancies in the status of SCCP pollution in the soils. In addition, the

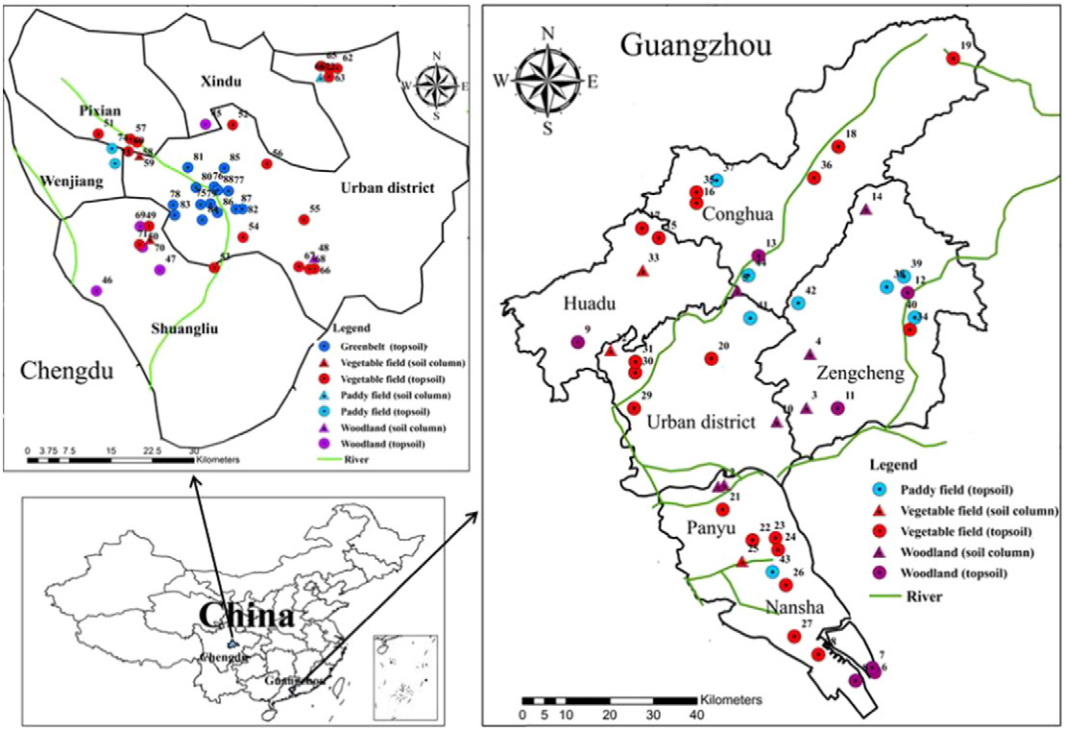

Fig. 1. Sampling sites of soils in Guangzhou and Chengdu.