



Review article

Occurrence, fate and ecological risk of chlorinated paraffins in Asia: A review



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ABSTRACT

Chlorinated paraffins (CPs), complex mixtures of polychlorinated alkanes, are widely used in various industries and are thus ubiquitous in the receiving environment. The present study comprehensively reviewed the occurrence, fate and ecological risk of CPs in various environmental matrices in Asia. Releases from the production and consumption of CPs or CP-containing materials, wastewater discharge and irrigation, sewage sludge application, long-range atmospheric transport and aerial deposition have been found to be most likely sources and transport mechanisms for the dispersion of CPs in various environmental matrices, such as air, water, sediment, soil and biota. CPs can be bioaccumulated in biota and biomagnified through food webs, likely causing toxic ecological effects in organisms and posing health risks to humans. Inhalation, dust ingestion and dietary intake are strongly suggested as the major routes of human exposure. Research gaps are discussed to highlight the perspectives of future research to improve future efforts regarding the analysis of CPs, the environmental occurrence and elimination of CPs, the total environmental pressure, and the risks to organisms and populations.

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1. Introduction

Chlorinated paraffins (CPs) are highly complex technical mixtures of alkanes with varying chain lengths and degrees of chlorination (in a

range of 30%–70% by mass). According to their carbon chain length, CPs can be classified as short-chain chlorinated paraffins (SCCPs, C₁₀–C₁₃), medium-chain chlorinated paraffins (MCCPs, C₁₄–C₁₇), and long-chain chlorinated paraffins (LCCPs, C₁₈–C₃₀) (Tomy et al., 1998a). In the European area, they have been manufactured as additives in lubricants and cutting fluids for metal working (70% of the total use), flame retardants in the rubber industry (10%), paints (9%), sealing materials (5%), and other applications (e.g., leather and textiles, 5%) since the 1930s (Štejnárova et al., 2005). With high production volumes and widespread applications, CPs are inevitably released into the environment during their production, transportation and usage, and they have been detected in a wide range of environmental media, such as air, water, sediments, soils, biota and humans worldwide (Bayen et al., 2006; Feo et al., 2009). Compared to the other two groups of CP mixtures, there is growing concern over SCCPs worldwide due to their persistence, greater potential for long-range atmospheric transport (LRAT), bioaccumulation in food webs and higher toxicity to aquatic organisms (Reth et al., 2005). In December 2009, SCCPs were added to Annexes I and II of the POPs (Persistent Organic Pollutants) protocol of the Convention on Long Range Transboundary Air Pollution (LRTAP). The OSPAR Commission for the Protection of the Marine Environment of the North-East Atlantic adopted a decision on SCCPs in 1995 – establishing a ban on uses in all areas of application (exemptions for dam sealants and underground conveyor belts until 2004). SCCPs have been under the review by the POP Review Committee (POPRC) since 2006. In 2015, the POPRC decided that SCCPs “are likely, as a result of long-range environmental transport, to lead to significant adverse human health and environmental effects such that global action is warranted” (Persistent Organic Pollutants Review Committee, 2012). A risk management evaluation for SCCPs is currently being prepared under the POPRC. However, there is no regulation of the utilization of CPs in China, which is the largest producer and consumer of CPs worldwide; therefore, a large amount of SCCPs are still produced and used in industrial activities (Chen et al., 2011).

However, the quantification of CPs in environmental samples is challenging, particularly because of the lack of suitable standards, their low response in various detection systems, and their highly complex nature, leading to great difficulties in chromatographic separation among CP congeners and between CPs and other organohalogen compounds (van Mourik et al., 2015). Consequently, information on CPs in the environment is still scarce compared to other organochlorine pollutants (such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs)). Almost all data in the reviews of CPs are available only for Europe and North America; there are currently no comprehensive overviews of CPs in Asia, even though China is the world's largest producer and consumer of CPs. Nevertheless, information on CPs in a wide variety of environmental matrices, such as air, water, wastewater, sewage sludge, sediment, soil and biota samples in Asia, has been increasing in the last decade. Abundant data have been accumulated from monitoring activities throughout Asia, providing an opportunity to assemble an integrated report on the emissions, occurrence, fate and toxicity of CPs, particularly SCCPs and MCCPs.

Therefore, the present review focuses on the occurrence and distribution of CPs in various environmental matrices in Asia, including results from abiotic media to show the source, transport and fate of CPs, as well as from biota to suggest a certain extent of bioaccumulation and biomagnification through food webs. Moreover, the toxicity and human exposure to CPs is evaluated. Finally, research gaps are discussed to define the directions and primary emphases of future studies.

2. Emission sources

2.1. Physicochemical properties

The physicochemical properties of CPs vary within large ranges and are governed by two factors: the carbon chain length and the degree of

chlorination. The melting point of CPs increases with the number of carbon and chlorine atoms; thus, CPs range from colorless to yellowish liquids at approximately 40% chlorination to white solids at 70% chlorination at room temperature. However, the vapor pressure and calculated Henry's law constant values of CPs appear to decrease with an increasing carbon-chain length and chlorination degree. Low vapor pressures are with ranges of 2.8×10^{-7} – 6.6×10^{-2} , 1.7×10^{-8} – 2.50 and 6.3×10^{-15} – 7.9×10^{-7} Pa for SCCPs, MCCPs and LCCPs, respectively, and their respective water solubilities are 0.49–1260, 0.029–14.0 and 1.6×10^{-6} – 0.086 µg/L (Environment Canada, 1993a,b). There are no large differences in the values of Henry's law constant, which have ranges of 0.34–14.7, 0.01–51.3 and 0.003–54.8 Pa m³/mol for SCCPs, MCCPs and LCCPs, respectively (Environment Canada, 1993b; Willis et al., 1994). The values of vapor pressure and the calculated Henry's law constant are similar to those for PCBs and some OCPs (Mackay et al., 1992, 1997), which are known to be transported in the atmosphere. The calculated octanol–air partition coefficients (log *K*_{oa}) vary from 8.20 to 9.80 for SCCPs with 50% to 60% chlorination (Feo et al., 2009). As a consequence of partitioning to the atmosphere, SCCPs and MCCPs can be remobilized from water to air or from moist soils to air, thus facilitating atmospheric partitioning and transport. Notably, with octanol–water partition coefficients (log *K*_{ow}) generally >5 (4.48–8.69, 6.83–8.96 and 8.70–12.7 for SCCPs, MCCPs and LCCPs, respectively) (Muir et al., 2000), lipophilic CPs have the potential to biomagnify in food webs (Zeng et al., 2011b; Ma et al., 2014b). These extremely large ranges of log *K*_{ow} values suggest the widely varying physical–chemical properties of CPs, which is one of the reasons why they are so challenging to analyze.

CPs are generally considered to persist in the environment. Direct photolysis, hydrolysis and oxidation by visible or near-UV radiation are suggested to be insignificant routes of transformation at ambient temperature (Mukherjee, 1990; Environment Canada, 1993b; Willis et al., 1994). According to Atkinson's OH radical reaction model (Atkinson, 1986), the theoretical atmospheric half-lives of CPs would be inversely proportional to their carbon chain length, with ranges of 1.2–1.8, 0.85–1.1 and 0.5–0.8 days for SCCPs, MCCPs and LCCPs, respectively (Willis et al., 1994). Nevertheless, the atmospheric half-lives of SCCPs were estimated to be 0.81–10.5 days and 1.2–15.7 days, respectively, meeting the persistence criteria for POPs (Persistent Organic Pollutants Review Committee, 2012). The hydrolysis and oxidation of CPs in natural waters are considered negligible at ambient temperature (Willis et al., 1994); however, hydrolysis or oxidation might occur in the aquatic environment in the presence of catalysts (Reiger and Ballschmiter, 1995). The biodegradation of CPs is inhibited by greater chlorine contents and longer carbon chains (Tomy et al., 1998b). Half lives in aerobic sediments were 12 ± 3.6 days and 30 ± 3.6 days for C₁₂–SCCPs with 56% and 69% chlorination, respectively (Persistent Organic Pollutants Review Committee, 2012), while they became 12 and 58 days for C₁₆–MCCPs with 35% and 69% chlorination, respectively (Fisk et al., 1998b). The half lives for SCCPs (65% chlorine) in freshwater and marine sediments were estimated at 1630 and 450 days under aerobic conditions, respectively, whereas little or no mineralization was noted in anaerobic sediments (UK Environment Agency, 2007). Specifically, the environmental residence time of C₁₆H₂₄Cl₁₀ is estimated to be 520 days compared to 210 days for C₁₂H₂₀Cl₆ (Tomy et al., 1998a). The historical records of CPs in dated sediment cores demonstrate that SCCPs might persist for >50 years in anaerobic lake sediments (Tomy et al., 1999; Iozza et al., 2008).

2.2. Production and consumption

In China, the industrialized production of CPs began in 1978, with a production volume of 3400 tons. Because of the high demand from the plastic industry, the annual production of CPs increased sharply from a few thousand tons in the early 1990s to 0.6 million tons in 2007 and to 0.8 million tons in 2010, which made China as the largest producer

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