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Distribution and accumulation of hexachlorobutadiene in soils and terrestrial organisms from an agricultural area, East China

Zhenwu Tang [a], Qifei Huang [b,*], Jiali Cheng [c], Dan Qu [a], Yufei Yang [b], Wei Guo [a]

[a] MOE Key Laboratory of Regional Energy and Environmental Systems Optimization, Resources and Environmental Research Academy, North China Electric Power University, Beijing 102206, China
[b] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[c] National Institute for Nutrition and Food Safety, Chinese CDC, Beijing 100021, China

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ABSTRACT

Hexachlorobutadiene (HCBD) is a potential persistent organic pollutant that has been found in abiotic environments and organisms. However, information on HCBD in soils and its accumulation in terrestrial food chains is scarce. This study investigated the accumulation of HCBD in soils, plants, and terrestrial fauna in a typical agricultural area in Eastern China, and drew comparisons with organochlorine pesticides (OCPs). The HCBD concentrations in soils were <0.02–3.1 ng/g dry weight, which were similar to α-endosulfan concentrations but much lower than the concentrations of some other OCPs. The HCBD soil–plant accumulation factors, 8.5–38.1, were similar to those of *o,p′*-DDT and higher than those of HCHs and *p,p′*-DDT, indicating that HCBD is strongly bioaccumulated by rice and vegetables. HCBD concentrations of 1.3–8.2 ng/g lipid weight were found in herbivorous insects, earthworms, and Chinese toads. The biomagnification factor, the ratio between the lipid-normalized concentrations in the predator and the prey, was found to be 0.16–0.64 for different food chains of Chinese toads, so HCBD was found not to biomagnify, which is in contrast with OCPs. Further research into whether HCBD is biomagnified in high trophic level organisms or through the entire terrestrial food web is required.

1. Introduction

Hexachlorobutadiene (HCBD), a halogenated aliphatic hydrocarbon, has been used as an intermediate in the production of a variety of chemicals and as an ingredient in transformer, hydraulic, and heat-transfer liquids (Lecloux, 2004; POPRC (Persistent Organic Pollutants Review Committee), 2013). It is also unintentionally generated during the production of chlorinated hydrocarbons, particularly perchloroethylene, trichloroethylene, and carbon tetrachloride (Brüschweiler et al., 2010). Hexachlorobutadiene is generally moderately acutely toxic and has been found to be genotoxic and carcinogenic in some laboratory animals (Rabovsky, 2000; Brüschweiler et al., 2010; POPRC (Persistent Organic Pollutants Review Committee), 2013). Adverse effects on marine and freshwater species have been observed from exposure to relatively low HCBD concentrations (POPRC (Persistent Organic Pollutants Review Committee) 2013) and HCBD has been detected in various abiotic and biotic media such as surface water, sediments, ambient air, and organisms (Lee and Fang, 1997; Vorkamp et al., 2004; Li et al., 2008; Juang et al., 2010; Miège et al., 2012). Thus, HCBD has been classed as a priority pollutant in many countries, including Canada, China, the UK, and the USA. In 2012, HCBD was listed as a candidate persistent organic pollutant by the Stockholm Convention for its toxicity, persistence, and potential for bioconcentration and long-range transport in the environment (POPRC (Persistent Organic Pollutants Review Committee), 2012).

The potential for HCBD to bioconcentrate in aquatic organisms has been experimentally confirmed. For fish bioconcentration factor (BCF, which is the ratio of the chemical concentration in an organism to the concentration in water) values from 1 to 19,000 L/kg on a whole body basis are reported by Environment Canada (1999). The International Programme on Chemical Safety (IPCS (International Programme on Chemical Safety), 1994) stated that HCBD bioaccumulation factors (BAFs), calculated using wet weight concentrations, in plankton, crustaceans, molluscs, insects, and fish in surface waters were comparable to those observed in the laboratory, and were in the range of 33–11,700 L/kg. Some previous studies have showed the levels of HCBD in aquatic organisms to be relatively low perhaps owing to the mild pollution in the waters studied (Environment Canada, 1999; Miège et al., 2012; Jürgens et al., 2013). At the same time, relatively higher

* Corresponding author. Fax: +86 10 8491 3903.
E-mail addresses: zwtang@ncepu.edu.cn (Z. Tang), huangqf@craes.org.cn (Q. Huang), jlcheng77@163.com (J. Cheng), qudan527@sina.com (D. Qu), cqyyf@163.com (Y. Yang), gwfybj@gmail.com (W. Guo).

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concentrations of HCBD have been reported in the aquatic species surrounding industrial areas (IKSR (Internationale Komission zum Schutz des Rheins), 2002; Environment Canada, 1999). A previous report found that the level of HCBD in the mussels that were caged in the river near to three industrial areas of the St. Clair River for three weeks was 36 ng/g wet weight (Environment Canada, 1999). However, recent HCBD monitoring data, especially for HCBD concentrations in terrestrial organisms are scarce and there is limited information available on HCBD bioaccumulation in terrestrial species.

Because of its lipophilicity (log K_{ow}=4.78) and ability to bioaccumulate (POPRC (Persistent Organic Pollutants Review Committee), 2013), HCBD is likely to biomagnify through food chains and food webs. Based on the BCF according to the European Commission (2003), The Netherlands (2012) estimated the HCBD biomagnification factor (BMF) as being 3 kg/kg, indicating that HCBD has the potential to be biomagnified. In contrast, Kelly et al. (2007) performed bioaccumulation modeling studies and found HCBD BMF values of <1 for all the organisms modeled, including water-respiring and air-breathing organisms. No field-based food chain studies have been performed, so the modeled BMF values (especially for terrestrial food chains) cannot be verified in natural food chains. Thus, the biomagnification potential of HCBD remains undetermined and investigations are urgently needed.

Many bioconcentrative chemicals, especially organochlorine pesticides (OCPs), have been investigated in depth and shown to bioaccumulate strongly, and to biomagnify through both aquatic and terrestrial food webs (Hoekstra et al., 2003; Weber et al., 2010; Foster et al., 2011; Silva Barni et al., 2014). It is known that HCBD and OCPs have many similar chemical properties and characteristics. In this study, the occurrence of HCBD in soils and terrestrial organisms in a typical agricultural area in Eastern China was investigated and the accumulations of HCBD and some OCPs in terrestrial plants were assessed to determine whether HCBD acts in a similar way to OCPs; being efficiently transferred from the abiotic environment into plants, and then entering the terrestrial food chain. The transfer of HCBD and OCPs through terrestrial food chains and the potential for HCBD to be biomagnified was also determined. This research will enable better understanding of the environmental behavior and fate of HCBD.

2. Materials and methods

2.1. Study area

Taicang, a city in Eastern China, with a population of 470,000 and an area of 810 km^2, is close to Shanghai. With the initiation of the "Reform and Open" policy in 1978, Taicang entered the fastest industrialization period ever experienced, and this has been sustained until today. Being one of the most developed industrial cities in the Yangtze River Delta, Taicang has many chemical synthesis plants and other industrial enterprises. It is possible that HCBD has been released from some local industrial sources into the surrounding farmlands. At the same time, the agriculture in this region is comparatively advanced and produces mainly vegetables and rice, because of the better weather and soil conditions. There has been intensive historic use of OCPs for supporting or improving the agricultural productivity in this area. In the Taicang area suburban farms, a variety of terrestrial plants and animals are widely distributed making it a suitable area for studying pollutant accumulation and transport in terrestrial species at different trophic levels.

2.2. Sample collection

In October 2011, soil and organism samples were collected from an agricultural area in the suburbs of Taicang (Fig. 1). Soil samples (0–10 cm deep) were collected from a total of 23 sites (five replicate samples from each site), in the east and south of Xintang, a village in a suburb of Taicang, using a stainless steel shovel. Each soil sample was placed in a pre-cleaned aluminum box and freeze dried before being ground, homogenized, and stored at -20 °C until analysis.

Rice (*Oryza sativa*) and some vegetables, including cabbage (*Brassica chinensis*), cowpea (*Vigna unguiculata*), pumpkin (*Cucurbita moschata*), mustard (*Brassica juncea*), and cauliflower (*Brassica oleracea*), were collected at the same time as soil sampling. The rice samples were collected from near the soil sampling sites 11–23 and the other plants were collected from vegetable plots at sites 6 and 7 (shown in Fig. 1). The rice was sampled by collecting all aboveground parts and the aboveground stems and leaves were collected for every vegetable sample. Each composite sample consisted of five subsamples. The numbers of composite samples are given in Table S1. The plant samples were washed with tap water and cut into lengths <1 cm and mixed well. Then the samples were freeze dried and ground, homogenized, and stored at -20 °C.

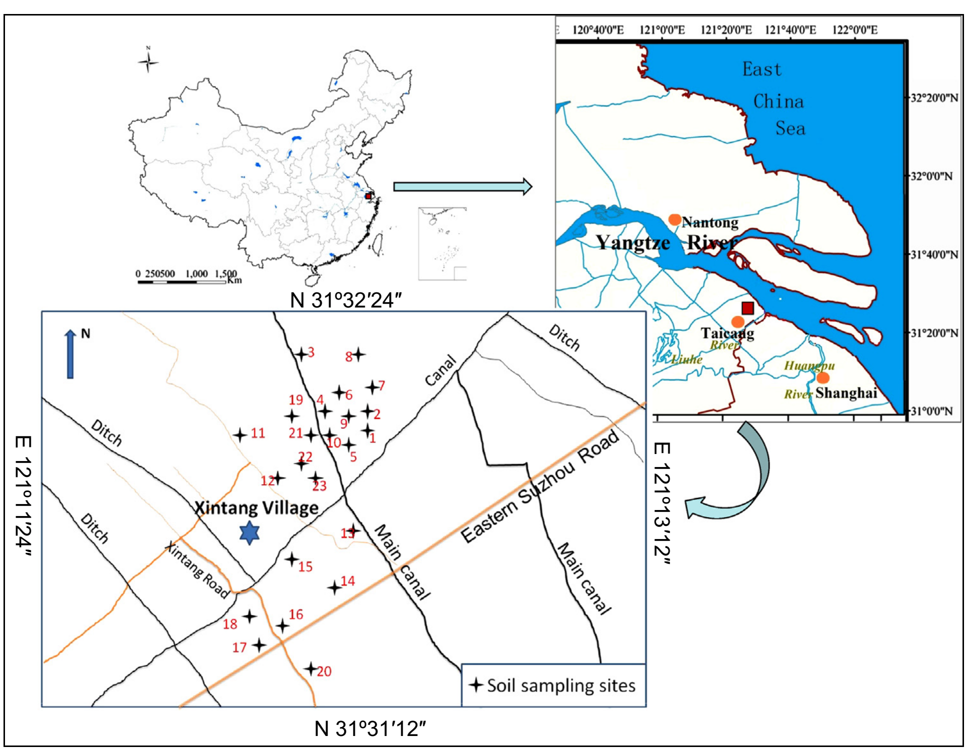

Fig. 1. Map showing the soil sampling sites in suburban Taicang, East China.