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Atmospheric DDTs over the North Pacific Ocean and the adjacent Arctic region: Spatial distribution, congener patterns and source implication

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ABSTRACT

During the 2003 Chinese Arctic Research Expedition (CHINARE 2003) from Bohai Sea to the high Arctic (37°N–80°N), air samples were collected and analyzed for DDTs. ∑DDTs (sum of six congeners) ranged from 0.52 to 265 pg m^{-3} with an average of 13.1 pg m^{-3}. Higher DDT concentrations were observed in Bohai Sea and near eastern Russia. The congener patterns were obviously different between the Far East Asia and the higher latitudinal regions that *p,p'*-DDT and *o,p'*-DDT were dominated in the former; while *o,p'*-DDT and *o,p'*-DDE were dominated in the latter. The source contributions of technical DDT and dicofol type DDT were estimated. Results showed that technical DDT was the dominant source (>94%) which was fresher in the Far East Asia compared to the North Pacific Ocean and the Arctic. For dicofol type DDT, the estimated contribution was minor. The "new" *o,p'*-DDT observed should have relatively more contribution from dicofol type DDT in the North Pacific Ocean and the Arctic.

1. Introduction

DDTs are a group of chlorinated pesticides (Table 1) that have long been a great concern due to their susceptibility to atmospheric transport, persistence in the environment, bio-accumulative potential and harmful effects especially on wildlife (Stokstad, 2007). Technical DDT, containing ~85% *p,p'*-DDT and ~15% *o,p'*-DDT, was heavily used in agriculture as well as in the control the spread of vector-born human diseases, like malaria, since 1940s (Metcalf, 1973; Qiu et al., 2004). Although technical DDT was banned worldwide for agricultural use from 1970s, restricted usage in disease vector control continues to this day in certain parts of the world, especially in Africa (Lubick, 2007). DDT can be degraded into DDE via oxidative dehydrochlorination and into DDD via reductive dechlorination. After the ban of agricultural use, ambient concentration of *p,p'*-DDT was decreasing gradually (Bignert et al., 1998). Its major metabolites, *p,p'*-DDE and *p,p'*-DDD, became increasingly dominant in the environment (Hung et al., 2002; Mai et al., 2002). Unfortunately, these metabolites are also highly persistent and toxic to wildlife. DDE, for example, was found to induce eggshell thinning in birds by disrupting calcium absorption (Lundholm, 1997). Despite its obvious toxicity to animal, DDT is still a controversial issue in its effects on human health. DDT is classified as "moderately hazardous" by WHO (2005) and has no significant correlation with cancer (López-Cervantes et al., 2004; Brody et al., 2007). Since DDT was banned mainly for ecological reasons (Rogan and Chen, 2005), considering its low cost, high effectiveness, persistence and low acute toxicity to human beings, WHO even announced that it supported the return of DDT in the fight against malaria on 16 September 2006 (Lubick, 2007).

In addition, "new" *o,p'*-DDT input recently reported in the environment made the situation more complicated. High concentration of *o,p'*-DDT was observed in the air across Asia (Qiu et al., 2004; Jaward et al., 2005). Dicofol, containing high contents of *o,p'*-DDT congeners as impurities, is regarded as a main contributor to ambient *o,p'*-DDT (Gillespie et al., 1994; Qiu et al., 2004). This pesticide is used to protect cotton, fruit trees, and vegetables from mites (Qiu et al., 2005) and was temporarily banned in United States and England until DDT contents were reduced (Rasenberg and van de Plassche, 2003). The successive input of *o,p'*-DDT would lead to the variation of DDT composition in ambient sample. In the Arctic, an increasing temporal trend of *o,p'*-DDT was observed during a five-year observation (Hung et al., 2002). Considering source inputs, transport, degradation and exchanges between various media, the congener patterns and spatial distribution of DDTs in the environment would be very complex.

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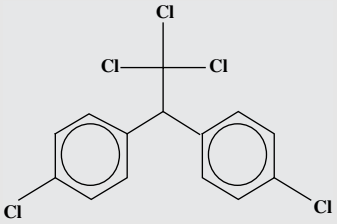
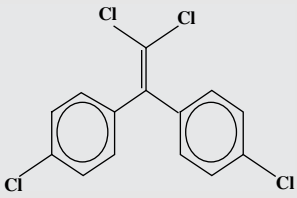
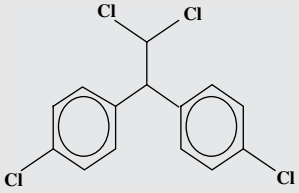
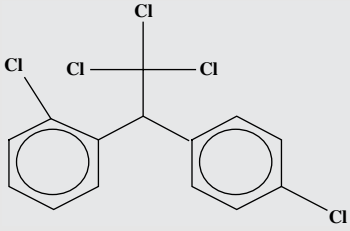
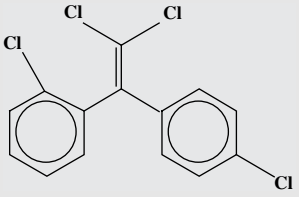
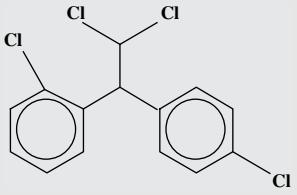
Table 1
Information of DDT congeners.

Abbreviations	CAS-Numbers	IUPAC Names	Structure
p,p'-DDT	50-29-3	1-chloro-4-[2,2,2-trichloro-1-(4-chlorophenyl)ethyl]benzene	
p,p'-DDE	72-55-9	1-chloro-4-[2,2-dichloro-1-(4-chlorophenyl)ethenyl]benzene	
p,p'-DDD	72-54-8	1-chloro-4-[2,2-dichloro-1-(4-chlorophenyl)ethyl]benzene	
o,p'-DDT	789-02-6	1-chloro-2-[2,2,2-trichloro-1-(4-chlorophenyl)ethyl]benzene	
o,p'-DDE	3424-82-6	1-chloro-2-[2,2-dichloro-1-(4-chlorophenyl)ethenyl]benzene	
o,p'-DDD	53-19-0	1-chloro-2-[2,2-dichloro-1-(4-chlorophenyl)ethyl]benzene	

To get a better understanding of the fate of DDTs, large-scale surveys are necessary to gain the information of source areas, compositional characteristics and long-range atmospheric transport (LRAT). In a previous cruise during 1989–1990 Iwata et al. (1993) first revealed the global distribution of DDTs in the ocean air. Simonich and Hites (1995) later also made a survey of DDT levels around the world based upon tree bark samples. Compared to other places (Oehme et al., 1996; Hung et al., 2002; Jaward et al., 2004a; Shen et al., 2005), Asia is one of the hot spots of DDTs where multiple sources of DDT can be found, including dicofol usage for cotton protection (Qiu et al., 2004), and technical DDT usage for disease control (Sharma, 2003), antifouling paints for fishing ships (Li et al., 2007), and even illegal agriculture purpose (Jaward et al., 2005). Previous studies showed that LRAT of persistent toxic