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A modeling assessment of association between East Asian summer monsoon and fate/outflow of α-HCH in Northeast Asia

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ABSTRACT

Using a dynamic numerical atmospheric transport model for organochlorine pesticides (OCPs), the relationship between the East Asian summer monsoon and the fate of α-hexachlorocyclohexane (α-HCH), a banned OCP, in the atmosphere over Northeast Asia was investigated and assessed. The modeled temporal and spatial patterns and variability of α-HCH air concentrations during the summer months of 2005 revealed a strong link between this chemical in the atmosphere over Northeast Asia and the East Asian summer monsoon. At lower atmospheric levels, easterly and southeasterly winds blowing from relatively cold ocean surface convey α-HCH air concentration from southeast China to northeast China. A monsoon front extending from southeast China to Japan, characterized by a strong wind convergence, carried the air concentration to a high elevation of the atmosphere where it was delivered by southerly monsoon flow to northern China and North Pacific Ocean. This summer monsoon associated northward atmospheric transport caused a reversal of the soil/air exchange from outgassing to net deposition during spring–summer period. The modeled wet deposition fluxes of α-HCH agreed well with the changes in the typical summer monsoon rain bands, designated as Meiyu in China, Changma in Korea, and Baiu in Japan. The major wet deposition flux paralleled with the monsoon front as well as the monsoon rain bands. The temporal change in the fluxes exhibits abrupt northward advances, which is associated with a stepwise northward and northeastward advance of the East Asian summer monsoon. The modeled α-HCH outflow in the atmosphere from China occurs mostly in the summer months and through northeast China, featured strongly by the evolution of the summer month. This study suggests that the East Asian summer monsoon provides a major atmospheric pathway and summer outflows to α-HCH over East Asia.

1. Introduction

Extensive studies have been devoted to detect the spatial and temporal variations of OCPs across China in recent years (Fu et al., 2003; Jaward et al., 2005; Lammel et al., 2007; Qiu et al., 2004) which have provided an understanding of the status of these OCPs in China. Of these OCPs, technical hexachlorocyclohexane (HCH) was one of the most heavily used pesticides in China (Li et al., 2001a). Application and persistence of technical HCH in China's environment has given rise to environmental contamination and health problem, and exerted a strong influence on the global HCH budget and trans-Pacific atmospheric transport (Bailey et al., 2000; Ding et al., 2007; Lammel et al., 2007; Li, 1999; Li et al., 1998, 2001a; Li and Bidleman, 2003; among many others). Atmospheric transport of OCPs in China on a local to regional scale has been reported in number of recent articles (Fu et al., 2003; Lammel et al., 2007; Qiu et al., 2004). These studies have analyzed trajectories and atmospheric pathways of these OCPs associated with winds on a regional scale. No studies, however, have been devoted to depict an overall spatial and temporal pattern of OCPs and their association with major atmospheric circulations in China.

The climate in Northeast Asia (China, Japan, Korea) and the adjacent oceanic region is affected substantially by the East Asian

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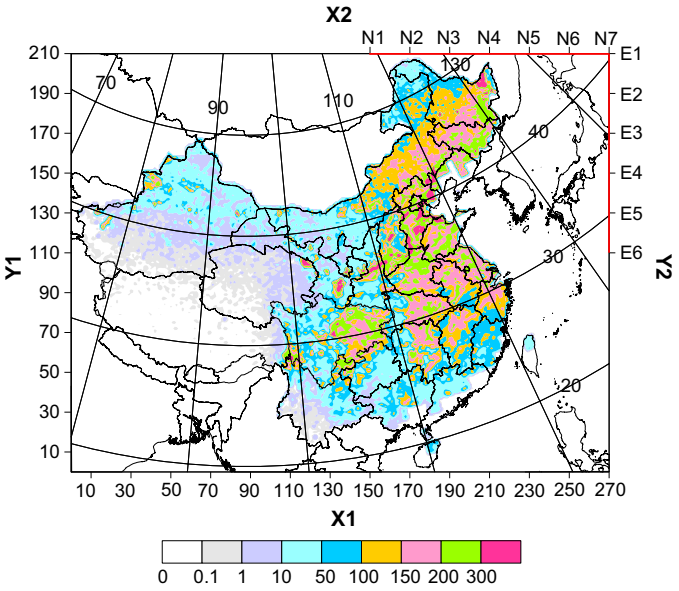

Fig. 1. Soil residue (kg cell^{-1}, 1 cell = 24 km × 24 km) of α-HCH in China in 2005. N1–N7 and E1–E6 in the upper right corner of the map are sectors where α-HCH outflows are estimated (Fig. 9).

monsoon, one of the strongest monsoon systems in the world (Ding and Chan, 2005). Its seasonal change dominates the climate over the continent and its surrounding areas. α-HCH is the major isomer of technical HCH (technical HCH used in China contains 67% α-HCH, Hu et al., 2009) which was extensively used in east China. Because east China has been affected most heavily by the East Asian monsoon regime, understanding the relationship between the multi-compartmental fate of α-HCH in the region and the Asian monsoon is essential to assess the impact of extensive use of OCPs in China on global budget of these toxic chemicals. It has previously been recognized that air pollutants transport in East Asia is strongly influenced by the monsoon regime. Previous studies on long-range transport of air pollutants in East Asia suggested that the winter monsoon regime favored more pronounced long-range and trans-Pacific transport of air pollutants than the summer monsoon regime (Pochanart et al., 2004). Because of the prevailing westerly flow over mid-latitudes of Northeast Asia, the continent exhibits an outflow pattern from land to ocean. However, it is important to note that during the wintertime we always observe low air concentration of OCPs due largely to their weak volatilization associated with low air temperature. Although wind regimes associated with the East Asian winter monsoon favors long-range transport of OCPs, such transport is less significant because the relatively lower air concentration is more readily dispersed in the atmosphere.

The lack of knowledge in the influence of the East Asian monsoon on spatial and temporal variations of OCPs in Northeast Asia poses a challenge for understanding atmospheric transport and outflow of OCPs in this part of Asian continent on a continental or inter-continental (trans-Pacific) scale. The present study aims at exploring the connections between the East Asian monsoon and the spatial/temporal patterns of OCPs in Northeast Asia, and evaluating the impact of the summer monsoon on the outflow of OCPs from their Asian sources. The major portion of the study is illustrated by a numerical simulation of α-HCH in China, with a focus on the impact of the East Asian summer monsoon on the spatial and temporal variations of α-HCH across China and other Northeast Asian countries.

2. Emission and model

2.1. α-HCH status in China

Owing to the extensive use of technical HCH in China between 1952 and 1983, an amount of 3,000,000 t of α-HCH, as a major portion of technical HCH, has entered the China's environment (Li et al., 2001a). A simplified gridded Pesticide Emission and Residue Model (PERM, Li et al., 2004) has been used to compile emission and residue inventories for α-HCH (Li et al., 2000, 2004), β-HCH (Li et al., 2003) on a global scale with a 1° latitude by 1° longitude resolution, for toxaphene on a regional scale with a 1/6° latitude by 1/4° longitude resolution (Li et al., 2001b). This model has been subsequently extended to a higher spatial resolution for α-HCH in China (Tian et al., in press). By inputting the annual usage information of α-HCH (Li et al., 2001a), the extended PERM was applied in this study to simulate emissions to air and residues in soil of α-HCH from 1952 through 2005 in China on a 24 km by 24 km resolution. Fig. 1 illustrates the modeled α-HCH annual soil residues. It was estimated that a total residue of 1500 t of α-HCH remained in Chinese soil in 2005 among which greater values of α-HCH soil residues are found in east China, indicating that east China is a major source of the substance.

2.2. Numerical assessment aspects

Canadian Model for Environmental Transport of Organochlorine Pesticides (CanMETOP, Ma et al., 2003) was employed in this modeling study. CanMETOP is a three-dimensional regional-scale atmospheric dispersion model coupled with a dynamic, three soil layers, Level IV fugacity-based soil/air exchange model (Harner et al., 2001), and a two-film model to estimate water/air gas exchange. In the soil compartment with multiple soil types characterized by soil organic carbon, the soil is treated as three separate well-mixed layers, namely, the exchange (0–0.1 cm), buffer (0.1–1 cm) and reservoir soil layers (1–10 cm). In each layer, chemical loss mechanisms include leaching, degradation and diffusion. At the top-layer loss due to volatilization and gain due to dry and wet deposition are included. For detailed gas/particle, soil/air, water/air, and other partitioning processes, readers are referred to Ma et al. (2003). The horizontal resolution of the model is 24 km and the model domain covers entire China, Japan and Korea. The coupled model has 12 atmospheric levels from the surface to 7 km height. During the summer monsoon the maximum southerly wind in southeast China lays at the height of 2 km whereas the updraft of the monsoon circulation seldom exceeds above 300-hPa (~8–9 km) (Lau and Li, 1984). The meteorological data (wind, air temperature, precipitation) in 2005 driving the CanMETOP were obtained by interpolating the 6-hourly objectively analyzed data from the United States National Center for Environmental Prediction (NCEP) reanalysis (Kalnay et al., 1996). Since the NCEP data are with a 2.5° latitude by 2.5° longitude resolution, they were interpolated into the 24 km × 24 km grids in the CanMETOP by using the Canadian Meteorological Centre's data objective analysis and interpolation

Table 1
Physical and chemical properties of α-HCH at 25 °C used in CanMETOP.

Physical and chemical properties	
Molecular mass (g mol^{-1})	290.85
Molar volume (cm^3 mol^{-1})	243.6
Solid solubility in water (g m^{-3})	1.0
Log KOA	7.26
Degradation half life in soil (day)	800
Degradation lifetime in air (day)	15–120
Washout ratio	2×10^5

Ref: Atkinson et al., 1992; Brubaker and Hites, 1998; Mackay et al., 2000.