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SCIENCE DIRECT®
Ecotoxicology and Environmental Safety 63 (2006) 196–203
Ecotoxicology and Environmental Safety
www.elsevier.com/locate/ecoenv

Rapid communication

Evaluating the fate of *p*,*p*′-DDT in Tianjin, China using a non-steady-state multimedia fugacity model

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Received 17 February 2005; received in revised form 5 June 2005; accepted 5 June 2005
Available online 31 August 2005

Abstract

Studies showed that DDT levels were still high in Tianjin, China, even though its use was banned in 1983. To estimate current risk of DDT to human health in Tianjin area, a non-steady-state (Level IV) multimedia fugacity model was used to simulate the fate and transfer of *p*,*p*′-DDT before and after the ban. The ordinary linear equations of Level IV model were solved with a matrix approach. The calculated *p*,*p*′-DDT concentration in air, water, soil, and sediment reached a maximum in the 1980s and then decreased, and agree well with those measured. The biggest bulk sinks of *p*,*p*′-DDT were soil and sediment, which accounted for 90% of total amount of *p*,*p*′-DDT in the environment. Air deposition and diffusions through the interfaces of water–air and water–sediment were the major intermedia transfer processes, while the degradation in soil and sediment were the key eliminating routes for *p*,*p*′-DDT in the environment.

Keywords: *p*,*p*′-DDT; Environmental fate; Multimedia fugacity model; Non-steady-state

1. Introduction

The organochlorine pesticide DDT (Dichlorodiphenyltrichloroethane) is a chemical used in large quantities during the past 50 years to control insects that harm agricultural crops and carry diseases like malaria. The commercial pesticide DDT consists of 70–90% *p*,*p*′-DDT and 10–20% of the isomer *o*,*p*′-DDT (Li et al., 1999). Like other persistent organic pollutants (POPs), DDT has been found worldwide due to its widespread use, environmental persistence, and atmospheric long-range transport. Although most nations either imposed a ban on DDT or restricted its use as pesticide since the 1970s, significant levels of DDTs are still being detected worldwide in air, water, soil, plants and wildlife, and even in Polar Regions.

As the largest developing country, China has produced and used 400 thousand tons of DDT since 1952, accounting for about 20% of the total world production (Hua and Shan, 1996). In 1983, China banned the application of DDT in agriculture. Now China has only two plants producing DDT as an intermediate for dicofol production and control of malaria. The Tianjin Chemical Plant is the larger one with an annual production of 2500–3500 tons DDT (Hu and Liu, 2003). It is believed that besides the emissions from croplands, where DDT was used in the past, the discharge of wastewater from the Tianjin Chemical Plant was also a source for DDT in the environment of the Tianjin area.

Tianjin is located in the northeastern China near the Bohai Bay. Since the first use of DDT in 1952, the

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doi:10.1016/j.ecoenv.2005.06.003

pollution of DDT in this area became a serious environmental problem. The monitoring results on JiYun River in 1976 showed that the river was heavily polluted; the concentration of total DDT reached 21,900 ng L^{-1} (Li, 1985). The use of DDT in Tianjin ended in 1983; however, recent studies on POP pollution in Tianjin area show that the pollution levels are still high. The concentrations of total DDTs in air particles, river waters, soils, and sediments in Tianjin Region were 1.87 ng m^{-3} (Wu et al., 2003), 18.1 (1.5–349) ng l^{-1} (He, 2003), 11.7 (0.7–972) ng g^{-1} (Gong et al., 2004), and 63.4 (1.65–5350) ng g^{-1} (He, 2003), respectively, much higher than the total DDTs concentrations in other regions in China and other countries. It is therefore important to estimate current risk of DDT to human health in Tianjin area and to evaluate the effectiveness of the ban of DDT use in 1983 on the reduction of DDT concentration in the environment.

To assess the environmental fate of *p*,*p*′-DDT, the concentrations and fluxes of *p*,*p*′-DDT among environmental compartments are needed. The available monitoring data, however, are very limited because most monitoring efforts do not provide representative data in time and space. Model simulation, therefore, provides a useful tool to fill the gap. A number of models have been used to estimate the fate of chemicals in the environment (Van Jaarsveld et al., 1997; Harner and Mackay, 1995). Among these, fugacity model, mostly Level III steady-state fugacity model (Mackay and Paterson, 1991), has been widely used in different regions to estimate the fate of different chemicals. For example, Equilibrium Criterion Model (EQC) used to assess environmental risk of pesticides in Xiamen area (Zhang et al., 1999), ChemFrance model used to estimate the environmental fate of organic chemicals such as lindane in France (Bintein and Devillers, 1996), ChemCAN model applied to assess the environmental fate and exposure of benzene and chlorobenzenes in Canada (MacLeod and Mackay, 1999) and to study benzo[a]pyrene in wastewater-irrigated area (Wang et al., 2003) and phenanthrene in Tianjin Region (Tao et al., 2003).

Level III fugacity model is based on a steady-state assumption and is relatively easy to use. However, it does not simulate the dynamic processes towards the steady state. A dynamic "Level IV fugacity model" was introduced to simulate the non-steady-state behavior, when the input of the chemical into the environment changes, for example as a result of a newly implemented regulation or policy (Bru et al., 1998). Level IV fugacity models are less commonly used than the steady-state ones because they are mathematically more complicated. Research so far on Level IV fugacity models mainly concentrates on numerical solutions (Bru et al., 1998; Hertwich, 2001), except a few recent studies on the historical behavior of chemicals in some regions, such as the study on long-term fate of two hexachlorocyclohexane isomers in the Baltic Sea environment (Breivik and Wania, 2002) using the POPCYCLING-Baltic Model, and the study on the fate of polychlorinated biphenyls in the United Kingdom over a 60-year period (Sweetman et al., 2002).

In this study, Level IV fugacity model was used to simulate the transfer and fate of *p*,*p*′-DDT in the Tianjin area before and after the ban of DDT use as a pesticide since 1984. The differential equations of Level IV fugacity model were solved with matrix approach. Distribution of *p*,*p*′-DDT concentration in air, water, soil, and sediment and the transfers between these compartments were estimated. The effect of the ban of DDT use in agriculture on *p*,*p*′-DDT residues in the environment was also discussed.

2. Fugacity model

The concept and application of fugacity models have been described by Mackay and co-authors (Mackay and Paterson, 1991; Mackay et al., 1992, 1996; Mackay, 2001). For Level IV fugacity models, the non-steady-state mass balance of a chemical in a compartment is described by the following ordinary differential equation:

$$V_i Z_i \mathrm{d}f_i/\mathrm{d}t = E_i + A_i + \sum(D_{ji} f_j) - f_i \sum D_{Ti} \qquad (1)$$

where V_i is the volume of the compartment i, Z_i is fugacity capacity of the compartment i, E_i is emission rate into the compartment i, A_i is advection input to the compartment i, D_{ji} is transfer coefficient from compartment j to compartment i, D_{Ti} is the total output from compartment i.

Air, water, soil, and sediment (indicated by 1, 2, 3, and 4, respectively) are usually the four bulk compartments included in a fugacity model. Each of the four bulk compartments is composed of air, water, and solid phases as sub-compartments. For instance, bulk air comprises pure air plus aerosol particulates, and bulk water comprises pure water plus suspended solids. Eq. (1) can be written in a matrix form:

$$\mathrm{d}f/\mathrm{d}t = \mathbf{A}\mathbf{f} + \mathbf{U}, \qquad (2)$$

where $\mathbf{f}$ is the solution vector and $\mathbf{U}$ is the emission rate vector:

$$\mathbf{f} = \begin{pmatrix} f_1 \\ f_2 \\ f_3 \\ f_4 \end{pmatrix} \quad \mathbf{U} = \begin{pmatrix} (E_1 + G_1 C_{B1})/V_1 Z_1 \\ (E_2 + G_2 C_{B2})/V_2 Z_2 \\ E_3/V_3 Z_3 \\ 0 \end{pmatrix}.$$