



Application of a level IV fugacity model to simulate the long-term fate of hexachlorocyclohexane isomers in the lower reach of Yellow River basin, China

Jiangting Ao, Jingwen Chen*, Fulin Tian, Xiyun Cai

Key Laboratory of Industrial Ecology and Environmental Engineering (MOE), Department of Environmental Science and Technology, Dalian University of Technology, Linggong Road 2, Dalian 116024, China

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ABSTRACT

A level IV multimedia fugacity model was established to simulate the fate and transfer of hexachlorocyclohexane (HCH) isomers in the lower reach of the Yellow River basin, China, during 1952–2010. The predicted concentrations of HCHs are in good agreement with the observed ones, as indicated by the residual errors being generally lower than 0.5 logarithmic units. The effects of extensive agricultural application and subsequent prohibition of HCHs are reflected by the temporal variation of HCHs predicted by the model. It is predicted that only 1.8 tons of HCHs will be left in 2010, less than 0.06% of the highest contents (in 1983) in the study area, and about 99% of HCHs remain in soil. The proportions of HCH isomers in the environment also changed with time due to their different physicochemical properties. Although β -HCH is not the main component of the technical HCHs, it has become the most abundant isomer in the environment because of its persistence. The dominant transfer processes between the adjacent compartments were deposition from air to soil, air diffusion through the air–water interface and runoff from soil to water. Sensitivity analysis showed that degradation rate in soil, parameters related to major sources, and thickness of soils had the strongest influence on the model result. Results of Monte Carlo simulation indicated the overall uncertainty of model predictions, and the coefficients of variation of the estimated concentrations of HCHs in all the compartments ranged from 0.5 to 5.8.

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1. Introduction

Organochlorine pesticides, such as hexachlorocyclohexane (HCH), have been recognized as one of the major classes of environmental pollutants (Bidleman and Olney, 1974; Tanabe and Tatsukawa, 1980; Atlas and Giam, 1981). Technical HCHs contain only 10–12% of the active isomer γ -HCH and are predominantly composed of non-insecticidal α -isomer (60–70%), β -isomer (5–12%) and δ -isomer (6–10%) (Iwata et al., 1993). Although only γ -HCH exhibits significant insecticidal activity, all the isomers are acutely and chronically toxic (Metcalf, 1955). β -HCH isomer may be the most toxicologically significant HCH due to its high persistence and estrogenic effects (Willett et al., 1998). Exposure to HCHs could be one of the significant risk factors for human beings.

HCHs were used extensively in China for many decades, because it is both effective and inexpensive. Technical HCHs were first introduced for use as pesticide in 1952, and on April 1, 1983, the Chinese government banned the production and use of HCHs (Chinese Ministry of Agriculture, 1989). The total amount

of technical HCHs produced in China was about 4.46 million tons from 1952 to 1983. During the 1970s and early 1980s, China was the largest producer and user of technical HCHs in the world (Li et al., 1998). In China, HCHs were mainly used in agriculture, although a small portion was also used in forestry and public health (Cai et al., 1992). The lower reach of the Yellow River basin is one of the major agricultural production bases in China, and thus was also one of the agricultural areas with the highest usage of HCHs in history (Li et al., 1998).

To date, information on the dynamic environmental fate and transfer of HCH isomers has not been available in the Yellow River basin. The methodology of multimedia environmental fugacity model was proven to be effective in simulating the fate of persistent organic pollutants in different spatial scales (Mackay and Paterson, 1991; Hertwich, 2001; Mackay, 2001), and a level IV fugacity model can be used for dynamic modeling addressing the changes in the past and predicting environmental fate of a pollutant in the future.

In this study, the dynamic changes of HCH isomers concentrations and transfer fluxes between adjacent compartments were simulated in the lower reach of the Yellow River basin during 1952–2010, by establishing a level IV fugacity model. Distribution

* Corresponding author. Tel./fax: +86 411 8470 6269.
E-mail address: jwchen@dlut.edu.cn (J. Chen).

of HCHs concentrations in the air, water, soil, and sediment was estimated. Model validation, sensitivity analysis and uncertainty analysis were performed to estimate the reliability of the model outputs. The results of the modeling can be used to assess the environmental quality of the lower reach of the Yellow River basin, and provide fundamental data to assess the potential ecotoxicological effects.

2. Methodology

2.1. Study area

The Yellow River is the second largest river in China, with a total length of 5464 km. The lower reach of the Yellow River extends 786 km from Huayuankou through the flat alluvial plains of Henan and Shandong provinces to the river mouth, with a basin area of $2.261 \times 10^4 \text{ km}^2$ (Liu et al., 1997; He, 2000; Wang et al., 2007), accounting for 3% of the whole area of the Yellow River basin. The lower basin is a humid area, with annual average temperature of 12–14 °C, and annual average precipitation of 670 mm (Wang et al., 2007). The light and heat resources are very abundant in the basin, and the natural conditions make it an agriculturally fertile area for many kinds of crops.

2.2. Model framework and calculation

A level IV fugacity model based on a non-steady-state assumption was applied to simulate the temporal trends of concentrations and inter-compartmental transfer fluxes of HCHs from 1952 to 2010. Air (pure air and particulates), water (pure water and suspended solids), soil (air, water and solids), and sediment (water and solids) were the four bulk compartments included in the fugacity model. The processes taken into consideration in the model for HCHs included agricultural application, advection air/water flow in/out of area, exchange between inter-compartment, and degradation in the four bulk compartments.

The comprehensive and detailed descriptions of the fugacity model are available in the monograph of Mackay (2001). By considering all the above processes, the non-steady-state mass balance of HCH isomers in the four compartments was described with the following system of linear differential equations:

$$V_A Z_A \frac{df_A}{dt} = E_A + G_A C_A + D_{W-A} f_W + D_{S-A} f_S - (D_{A-W} + D_{A-S} + D_{A(A)} + D_{R(A)}) f_A \quad (1)$$

$$V_W Z_W \frac{df_W}{dt} = G_W C_W + D_{A-W} f_A + D_{S-W} f_S + D_{Sed-W} f_{Sed} - (D_{W-A} + D_{W-Sed} + D_{A(W)} + D_{R(W)}) f_W \quad (2)$$

$$V_S Z_S \frac{df_S}{dt} = E_S + D_{A-S} f_A - (D_{S-A} + D_{S-W} + D_{R(S)}) f_S \quad (3)$$

$$V_{Sed} Z_{Sed} \frac{df_{Sed}}{dt} = D_{W-Sed} f_W - (D_{Sed-W} + D_{R(Sed)}) f_{Sed} \quad (4)$$

where the subscript $i = A, W, S$ and Sed , represents bulk compartment of air, water, soil and sediment, respectively. V_i is the volume of the compartment i , Z_i is fugacity capacity of the compartment i . E_i is the emission rate into the compartment i . G_i is advection flow rate of compartment i . C_i is background inflow concentration of adjacent region in compartment i . D_{ij} is transfer rate coefficient from compartment i to compartment j . $D_{A(i)}$ and $D_{R(i)}$ represent advection flow rate coefficient and degradation rate coefficient of compartment i , respectively.

The model was programmed using Matlab 7.0, and the system of the linear differential equations was solved at hourly time steps. The initial values of the fugacity were set at zero in 1952, and the calculated values of fugacity from previous phase served as the ini-

tial values for the succeeding phase. For each value of t , the values of the fugacities $f_i = f_i(t)$ are multiplied by the capacity of fugacity Z_i to determine the value of concentration $C_i = C_i(t)$ of HCH isomers in the bulk compartment. Consequently, the transfer fluxes between the adjacent compartments can also be calculated using the values of fugacity and transfer rate coefficient.

2.3. Model parameters

The input parameters to the model comprised environmental parameters describing compartment composition, inter-compartment exchange rates, etc., physicochemical properties of HCH isomers and emission data. Since the parameter values usually span a wide range, geometric means and standard deviations could be derived. For the parameters with only a single value available, their coefficients of variation (CV) were artificially assigned (Cao et al., 2004). The geometric means and standard deviations were used for model calculation and uncertainty analysis.

As many parameters as possible that represent the environmental characteristics of the study area were collected from literatures (Wu et al., 1999; He, 2000; Cao et al., 2005a; Liu et al., 2004; Shi et al., 2005; Zhao et al., 2005). Default values were taken from Mackay and Paterson (1991) and Mackay (2001) in absence of reliable literature data. These values of relevant environmental parameters are listed in Table 1. The values of physicochemical properties of HCH isomers used here were the mean values derived from previous studies or most frequently used in the model calculation (Mackay et al., 1997; Mackay, 2001; Cao et al., 2005a, 2007), and these values are summarized in Table 2.

Few anabranch and sewage merged into the lower reach of the Yellow River (Dong, 1992). Therefore, the impact of this factor on the concentration of HCHs can be neglected. The inputs of HCHs into the environment of the study area were from agricultural use and air/water advection. Emission data are not always easily accessible or largely uncertain, and have to be estimated (Scheringer and Wania, 2003; Li et al., 2006). The amount of HCHs used in the local agriculture is estimated by the annual average usage of HCHs in the Shandong and Henan provinces, and the agriculture area in the basin (Dong, 1992; Hua and Shan, 1996; Li et al., 1998, 2001). The agricultural use of HCHs increased since 1970s in the basin and reached a peak in 1980, decreased thereafter to zero in 1984 (Li et al., 2001). Therefore, the historical agricultural use of HCHs in the basin could be roughly divided into three phase: (1) during 1952–1969, the estimated HCHs agricultural use was about 530 tons per year; (2) 1470 tons per year during 1970–1983; and (3) none since 1984.

According to Li et al. (2003), the amount of β -HCH emitted to the atmosphere due to the spraying event was 30% of the total application. Considering the vapor pressures of the other isomers are higher than that of β -HCH, we presumed that 40% of the total use of the other isomers entered into air and 60% entered into the soil. No monitoring data were available for the air advection flow concentrations of HCH isomers in the upper-wind of the study area. The air advection flow concentrations (C_a) of HCH isomers before 1983 were estimated from concentrations of pine needle HCHs in North China in early 1980s, by assuming equilibrium between the air and pine needles phases (Ockenden et al., 1998; Xu et al., 2002), and C_a after 1983 were estimated from the monitoring results about HCHs in 1996 and 2002 in North China (Wu et al., 2003; Cao et al., 2005a). Due to the lack of monitoring data about the inflow water concentrations of HCH isomers before 1983, the inflow water concentrations were estimated from the measurement of HCHs concentrations in up-stream of the lower reach of the Yellow River in 1986–1987 (Bao et al., 1991), according to the variation of water concentrations derived from the model results, as also did by Cao et al. (2005b).

Table 1
Environmental parameters of the study area^a

Symbol	Unit	Values		Symbol	Unit	Values	
		Mean	Std.			Mean	Std.
A_w	m ²	3.8×10^8	3.1×10^6	r_{pw}	kg m ⁻³	1.8×10^3	6.8×10^2
A_s	m ²	2.2×10^{10}	1.8×10^8	r_{ps}	kg m ⁻³	2.4×10^{3b}	1.5×10^2
h_a	m	5.0×10^2	1.8×10^2	r_{psed}	kg m ⁻³	2.4×10^{3b}	2.7×10^2
h_w	m	5.0×10^0	1.7×10^0	f_{pw}		3.2×10^{-3}	1.6×10^{-3}
h_s	m	1.0×10^{-1}	3.5×10^{-2}	f_{ps}		8.0×10^{-3}	4.5×10^{-3}
h_{sed}	m	5.0×10^{-2}	1.8×10^{-2}	f_{psed}		4.3×10^{-3}	1.3×10^{-3}
V_{pa}		3.7×10^{-10}	6.3×10^{-10}	Q		2.0×10^{5b}	8.0×10^4
V_{pw}		2.7×10^{-3}	6.9×10^{-3}	U_r	m h ⁻¹	7.5×10^{-5}	4.8×10^{-6}
V_{as}		2.0×10^{-1a}	4.0×10^{-2}	U_p	m h ⁻¹	1.1×10^{-b}	9.4×10^{-1}
V_{ws}		3.0×10^{-1a}	5.0×10^{-2}	U_{ww}	m h ⁻¹	3.0×10^{-5}	1.9×10^{-6}
V_{ps}		5.0×10^{-1a}	1.1×10^{-1}	U_{sw}	m h ⁻¹	2.3×10^{-8b}	1.5×10^{-9}
V_{wsed}		3.0×10^{-1a}	1.4×10^{-1}	U_{dp}	m h ⁻¹	6.8×10^{-7}	5.2×10^{-7}
V_{psed}		7.0×10^{-1a}	6.0×10^{-2}	U_{rsed}	m h ⁻¹	2.7×10^{-7}	2.1×10^{-7}

^a Default values were from Mackay (2001).

^b Default values were from Mackay and Paterson (1991).

^{*} A_w, A_s area of water, soil; h_a, h_w, h_s, h_{sed} depth of air, water, soil and sediment; $V_{pa}, V_{pw}, V_{ps}, V_{psed}$ volume fraction of solid in air, water, soil and sediment; V_{as} volume fraction of air in soil; V_{ws}, V_{wsed} volume fraction of water in soil and sediment; r_{pw}, r_{ps}, r_{psed} densities of solids in water, soil and sediment; f_{pw}, f_{ps}, f_{psed} contents of organic carbon in solids in water, soil and sediment; Q scavenging rate; U_r rain rate; U_p dry deposition velocity; U_{ww} water runoff rate from soil; U_{sw} soil runoff rate from soil; U_{dp} sediment deposition rate; U_{rsed} sediment resuspension rate.

Table 2
Physicochemical properties of HCHs isomers^{*}

Symbol	Unit	α -HCH		β -HCH		γ -HCH		δ -HCH	
		Mean	Std.	Mean	Std.	Mean	Std.	Mean	Std.
H	Pa m ³ mol ⁻¹	6.3×10^{-1}	5.5×10^{-1}	1.2×10^{-1}	2.8×10^{-2}	3.2×10^{-1}	1.6×10^{-2}	8.3×10^{-3}	3.7×10^{-2}
P_s	Pa	2.7×10^{-1}	6.1×10^{-2}	5.1×10^{-5}	3.5×10^{-5}	5.5×10^{-2}	2.6×10^{-2}	5.4×10^{-3}	1.6×10^{-2}
K_{oc}		1.6×10^3	8.8×10^2	1.5×10^3	9.1×10^2	1.4×10^3	1.1×10^3	2.0×10^3	1.1×10^3
k_a	h ⁻¹	8.0×10^{-4}	1.6×10^{-3}	2.7×10^{-4}	5.3×10^{-4}	8.7×10^{-4}	2.8×10^{-3}	2.7×10^{-4}	1.6×10^{-3}
k_w	h ⁻¹	8.1×10^{-4}	1.6×10^{-3}	8.1×10^{-4}	7.0×10^{-4}	1.3×10^{-4}	6.6×10^{-4}	3.6×10^{-4}	7.6×10^{-4}
k_s	h ⁻¹	7.8×10^{-5}	1.6×10^{-4}	3.0×10^{-5}	4.7×10^{-5}	7.0×10^{-5}	3.2×10^{-5}	7.8×10^{-5}	6.1×10^{-4}
k_{sed}	h ⁻¹	5.2×10^{-5}	1.0×10^{-4}	2.6×10^{-5}	4.6×10^{-4}	1.1×10^{-4}	4.1×10^{-4}	5.2×10^{-5}	4.8×10^{-4}

^{*} P_s vapor pressure; K_{oc} organic carbon partition coefficient; k_a, k_w, k_s and k_{sed} degradation rate constants in the air, water, soil and sediment. The values were derived from Mackay et al. (1997), Mackay (2001), and Cao et al. (2005a; 2007).

3. Results and discussion

3.1. The dynamic changes of concentration

Temporal trends of concentrations of HCH isomers in air, water, soil, and sediment during 1952–2010 in the study area are shown in Fig. 1. Changes in the usage pattern with time were rapidly reflected in temporal changes of HCH isomer concentrations. A relatively steady state was reached for the concentration of HCH isomers in the study area in about 12 years after the application of HCHs in 1952. This steady state continued till 1969 when the application of HCHs began to increase. The concentrations of HCH isomers in the study area reached maximum in 1983. Due to the ban of HCHs pesticides, HCH isomers concentrations decreased sharply in 1984, and the downward trend decelerated until recent years. As compared with 1983, concentrations of HCHs in air, water, soil and sediment in 1984 reduced by about 99%, 74%, 33% and 30%, respectively. According to model prediction, only about 0.06%, 0.05%, 0.05% and 0.09% of the highest concentrations in air, water, soil and sediment, respectively, will be left in 2010.

The time needed for reaching the steady state in different environmental compartments changed greatly. HCHs reached their steady state the most quickly in air because of the mobility and relatively fast degradation in air. The concentrations of HCHs in soil and sediment changed slowly and decreased continuously after the ban, due to their slow response time (large fugacity capacity, and slow evaporation and degradation). The time needed for reaching the steady state in water was between that of air and soil.

The proportions of HCH isomers in the environment also changed with time due to their different physicochemical properties and corresponding environmental behavior. As the main component of technical HCH, α -HCH entered the environment far more than the other isomers, thus much higher concentration of α -HCH was found in all the compartments before 1983. After the ban in agriculture, the percentage of α -HCH in HCHs also kept a relative high level in all the compartments. The percentage of β -HCH in HCHs increased gradually in all the compartments except for in the air, and even has exceeded α -HCH in soil and sediment since early 1990s because it is the more persistent in the environment. It indicates that β -HCH has become the most abundant isomer in HCHs till now. In the air, the concentrations of β -HCH and δ -HCH were very low in comparison with α -HCH. The major reasons are the differences of their physicochemical properties. The Henry's law constants and vapor pressures of β -HCH and δ -HCH are much less than that of α -HCH, which relate to soil–air and water–air exchange. γ -HCH entered the environment a little more than β -HCH, while the concentrations of γ -HCH decreased far more quickly than β -HCH in the water, soil and sediment because γ -HCH was prone to volatilize and was degraded faster in the environment. The concentrations of γ -HCH and δ -HCH maintained relatively constant since early 2000s in the study area.

3.2. Transfer and distribution

Before 1983, the major source of HCHs in the study area was agricultural application of pesticides. Since 1984, air advection

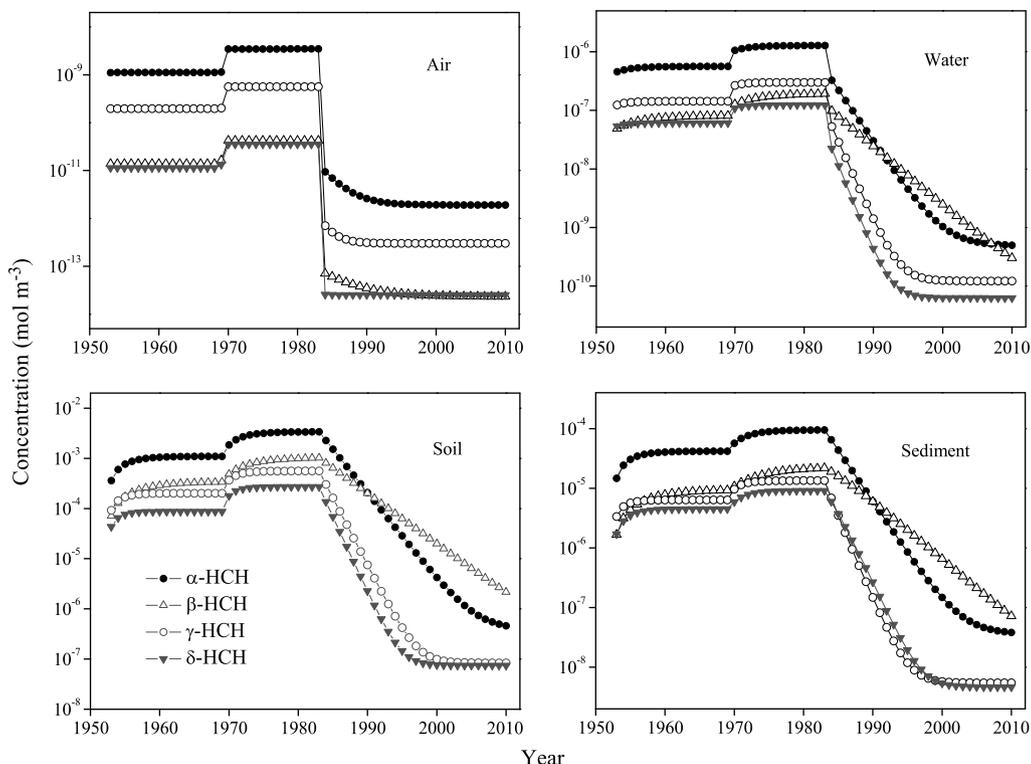


Fig. 1. Temporal trends of the concentration of HCH isomers in various compartments from 1952 to 2010.

input has become the dominant source of HCHs in the study area. The pattern of the dynamic change of the transfer flux was similar to that of concentration change because the transfer flux and concentration were both calculated from fugacity. According to the calculation, degradation was the main pathway for HCHs to vanish from the study area, and degradation in the soil was the major loss process, accounting for about 90% of the total degradation amount, followed by degradation in the air. During 1952–1983, about 749 t a⁻¹ HCHs disappeared from the study area through degradation in the soil, while the value decreased quickly after 1983 due to the prohibition of HCHs.

Transfer fluxes of HCHs between the adjacent compartments are shown in Fig. 2. The dominant transfer processes of HCHs in the study area were deposition from air to soil, air diffusion through the water–air interface and runoff from soil to water. The prohibition of HCHs also resulted in a sharp decline in the transfer fluxes of HCHs between the adjacent compartments. All

the transfer fluxes of HCHs between the adjacent compartments in 2010 are expected to decrease to less than 0.09% of those in 1983.

The contents of HCHs in the environmental compartments were calculated using the concentrations derived from model calculation and volumes of the bulk compartments. In 1983, the total amount of HCHs reached the highest value, and their reserve in the environment of the study area reached 3383 t. From 1984 to early 2000s, the total amount of HCHs has experienced a rapid descending, and only 1.8 t will be remained in 2010. The distribution characteristic of HCHs in the four bulk compartments changed a little with time. The proportion of HCHs in air, water, soil and sediment are 0.40%, 0.04%, 99.53%, 0.03% in 1969, 0.40%, 0.03%, 99.56%, 0.02% in 1983 and 0.41%, 0.03%, 99.52%, 0.04% in 2010, respectively. It can be seen that soil is the dominant sink. This implies that the rate of disappearance of HCHs in the environment of the study area mainly determined by the slow quantified degradation rate in the soil environment.

Cao et al. (2005a) simulated the long-term transfer and fate of HCHs in Beijing, and Li et al. (2006) evaluated the fate of *p,p'*-DDT in Tianjin using a non-steady-state multimedia fugacity model. They also found that the concentration variation of the pollutants was dependent on the agricultural applications. Moreover, air–soil deposition was the most primary transfer process between the adjacent compartments, and degradation in the soil was the dominant eliminating pathway for the pollutants.

3.3. Model validation

Measured data associated with the study area were collected from previous studies (Hu and Li, 1986; Bao et al., 1991; Wu et al., 1999; Zhao et al., 2005; Sun et al., 2007). No measured levels of HCHs in the basin air were available, which were obtained in the neighboring areas (Wu et al., 2003; Cao et al., 2005a; Lammel et al., 2007). Comparisons between modeled and observed

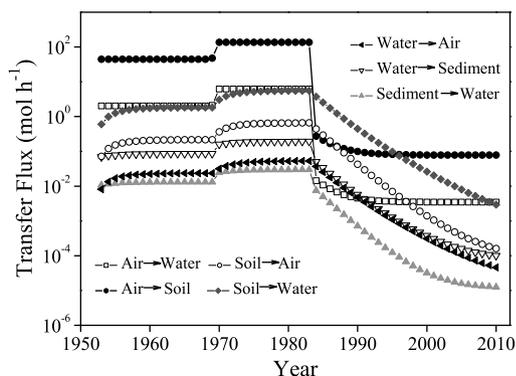


Fig. 2. Transfer fluxes of HCHs between the adjacent compartments during 1952–2010.

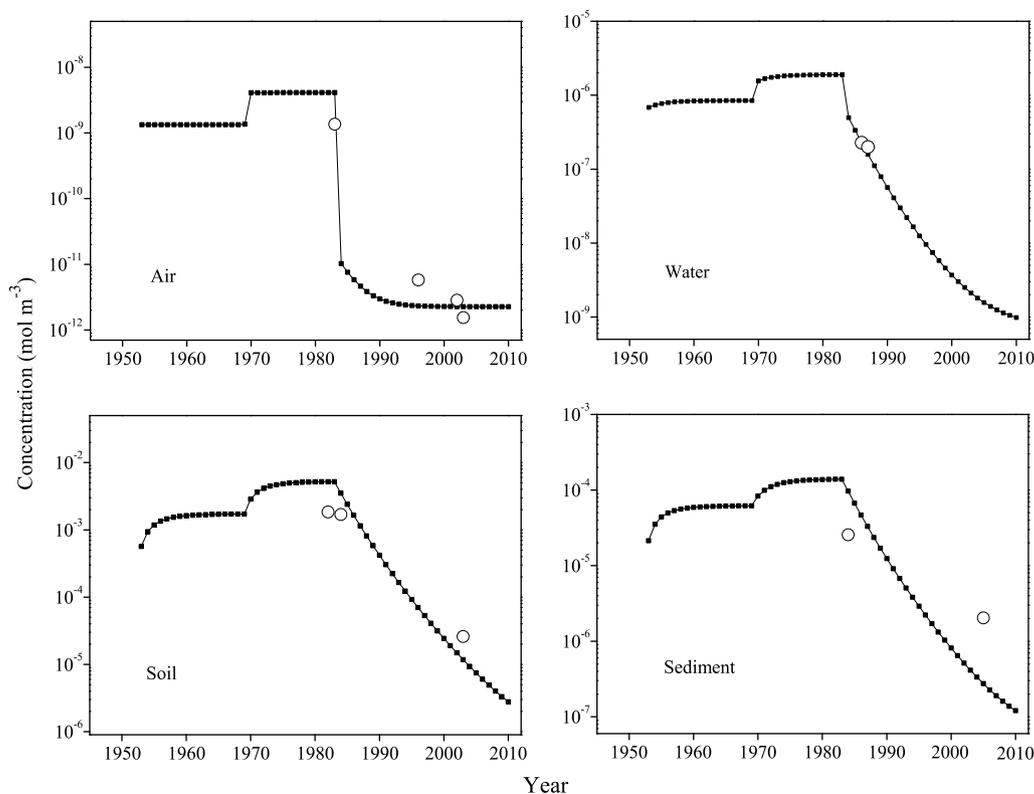


Fig. 3. Comparison between the calculated (curve) and observed (dot) concentrations of HCHs in various compartments in different periods.

concentrations showed good agreement. The differences between modeled and observed concentrations were generally from 0.01 to 0.48 logarithmic units (Fig. 3), except for the concentrations in the sediment. The largest variation between modeled and observed concentrations was found in sediment of 2005, with a difference of 0.87 logarithmic units. This can be explained by the follows: (1) the calculated HCHs concentrations represent the average of the whole study area, while the measured data were taken from limited sampling sites; (2) the uncertainty of the input parameters, such as the degradation rate in the sediment that was from the literature not from specific measurements for the purpose of modeling, may lead to prediction errors.

3.4. Sensitivity analysis

Sensitivity analysis as well as model validation is important means for evaluation of multimedia fugacity models. The main purpose of sensitivity analysis is to identify the major uncertainties and variabilities in the input data (Scheringer and Wania, 2003). The sensitivity coefficient was defined as the ratio of the relative variation of the estimated concentration to that of the input parameter

$$SC_i = \frac{\Delta Y_i / Y_i}{\Delta X_i / X_i} \quad (5)$$

where SC_i represents the sensitivity coefficient of input parameter i . X_i and Y_i represent the input parameter i and corresponding estimated concentration, respectively.

The sensitivity coefficients of input parameters to the calculated concentrations of HCHs in the four bulk compartments in 1969 (steady-state), 1983 (steady-state) and 2010 (after-steady-state, prediction) exceeding 0.7 are shown in Fig. 4. The influences of input parameters to the estimated HCHs concentrations in different compartments were greatly different. For example, only air and

soil were sensitive to temperature (T), while sediment was strongly affected by degradation rate in sediment (k_{sed}) and sediment thickness (h_{sed}). The sensitivity of most input parameters also changed with time, especially the degradation rate in soil (k_s) and the parameters related to major source. The degradation rate in soil becomes the most influential to the model outputs for 2010. The major input of HCHs from agricultural use decreased to zero in 1984, and degradation in soil is the dominant eliminating pathway for HCHs. So the influence of k_s on the model outputs gradually increased after the ban, and k_s will become the dominant parameter determining the fate of HCHs in 2010. The emission rates to air and soil (E_a and E_s) were the most influential parameters in 1969 and 1983, but have little influence on the model results for 2010 due to the ban. The influences of air advection flow rate and concentration (G_a and C_a) on the model results will become powerful in 2010. Among all the other parameters, volume fractions of solids in water and soil (V_{pw} and V_{ps}), sediment deposition rate (U_{dp}), sediment resuspension rate (U_{rsed}) and thickness of soil and sediment (h_s and h_{sed}), have a strong and relatively consistent influence over time on the uncertainty in the model outputs.

3.5. Uncertainty analysis

Uncertainty analysis was performed using Monte Carlo simulation. Normal or lognormal distribution was adopted for the key parameters identified in the sensitivity analysis, considering the probability distributions of these parameters (Cao et al., 2004, 2007). The simulation carried out repeatedly 2000 times with values randomly selected based on their probability distributions.

Results of Monte Carlo simulation for HCHs in the four bulk compartments were shown in Fig. 5, with semi-interquartile range (25–75th percentile range) marked. As shown in Fig. 5, the dispersions of the concentrations for each media were moderately large, and generally covered 0.5–2.4 orders of magnitude. Most of the CV

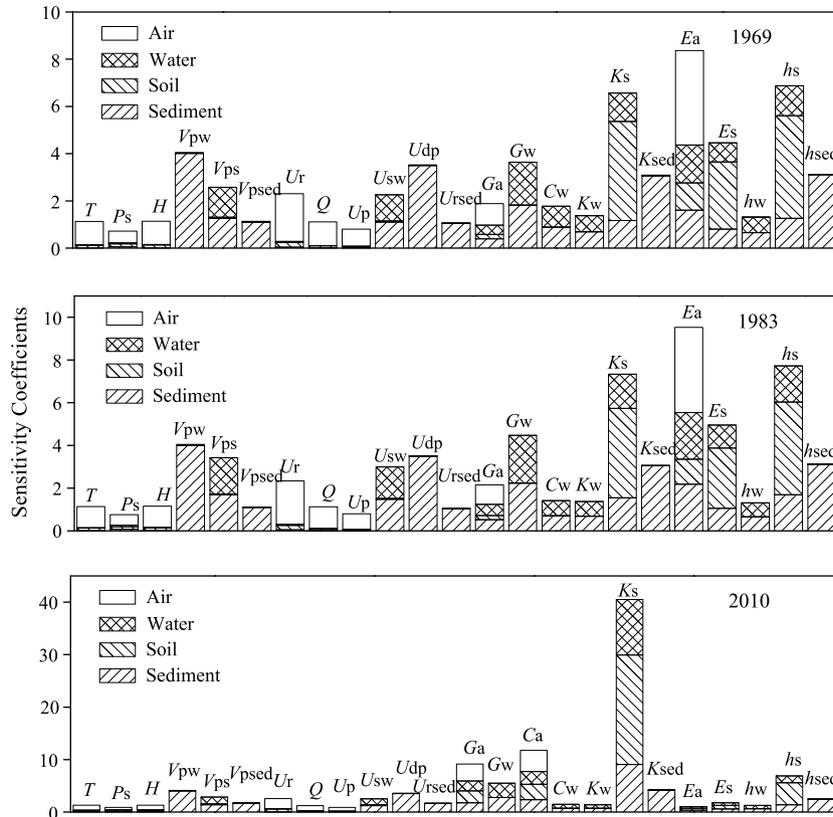


Fig. 4. Sensitivity coefficients of input parameters to concentrations of HCHs in various compartments in 1969, 1983, and 2010.

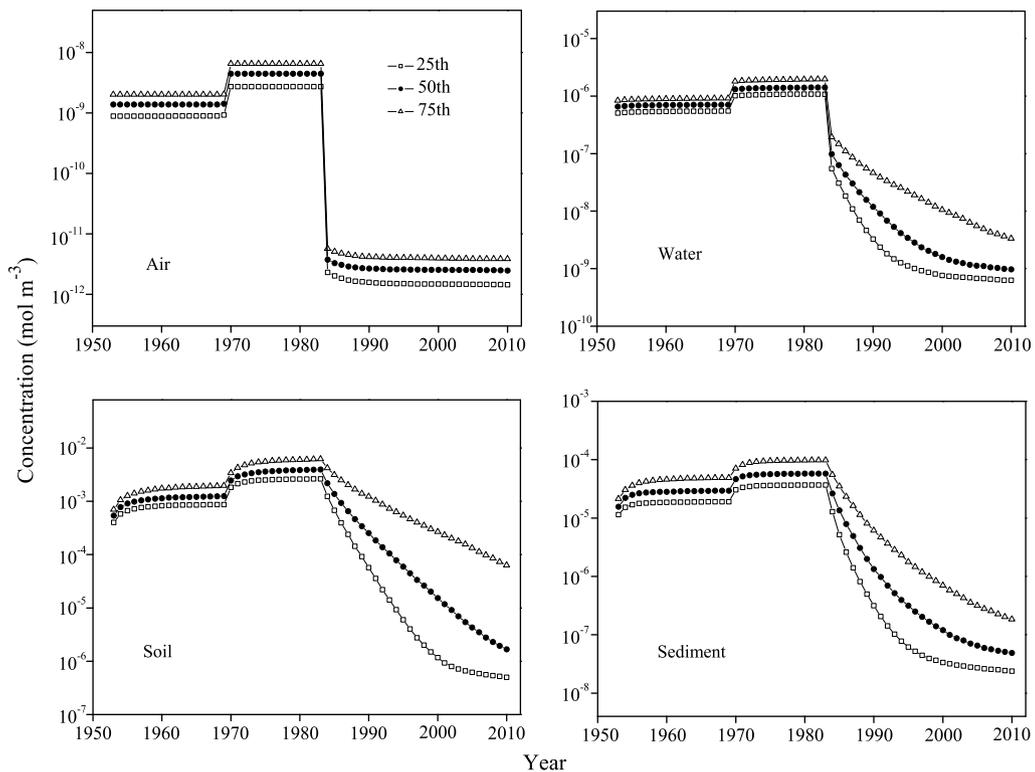


Fig. 5. The dispersions of HCHs concentrations in various compartments during 1952–2010 derived from Monte Carlo simulation. The results are presented as the 25th (at the bottom), the 50th (in the middle), and the 75th (on the top) percentiles.

values of the key input parameters are less than 1 except for degradation rate constants, yet the CV values of the estimated concentrations ranged from 0.5 to 5.8. The CV values of concentrations

also changed with time. In general, the relatively higher CV values of concentrations were found after 1983. The estimated concentrations in soil and sediment have the larger uncertainties than in

other compartments, which are possibly related to the most influential parameters (k_s , k_{sed} and E_s).

4. Conclusions

The level IV fugacity model was successful in reproducing the long-term trend of HCH isomers in the multimedia environment of the lower reach of the Yellow River basin. Direct emission of agricultural use within the basin was the decisive factor controlling the temporal patterns of HCHs in the area. The concentrations of HCHs reached a maximum in the early 1980s and then decreased sharply due to the prohibition of HCHs pesticides. The proportions of HCH isomers also changed with time. The pattern of the dynamic change of the transfer flux between the adjacent compartments was similar to that of concentration change. Degradation rate in soil, parameters related to major sources, and thickness of soil were main factors influencing the model results. Monte Carlo simulation showed the CV values of the estimated concentrations of HCHs in all the compartments ranged from 0.5 to 5.8.

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