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RESEARCH ARTICLE

Environmental behaviors and potential ecological risks of heavy metals (Cd, Cr, Cu, Pb, and Zn) in multimedia in an oilfield in China

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Abstract The environmental behaviors of five heavy metals (Cd, Cr, Cu, Pb, and Zn) in a Chinese oilfield were investigated using a steady-state multimedia aquivalence (SMA) model. The modeling results showed good agreement with the actual measured values, with average residual errors of 0.69, 0.83, 0.35, 0.16, and 0.54 logarithmic units for air, water, soil, sediment, and vegetation compartments, respectively. Model results indicated that most heavy metals were buried in sediment, and that transfers between adjacent compartments were mainly deposition from the water to the sediment compartment (48.59 %) and from the air to the soil compartment (47.74 %) via atmospheric dry/wet deposition. Sediment and soil were the dominant sinks, accounting for 68.80 and 25.26 % of all the heavy metals in the multimedia system, respectively. The potential ecological risks from the five heavy metals in the sediment and soil compartments were assessed by the potential ecological risk index (PERI). The assessment results demonstrate that the heavy metals presented low levels of ecological risk in the sediment compartment, and that Cd was the most significant contributor to the integrated potential ecological risk in the oilfield. The SMA model provided useful simulations of the transport and fate of heavy metals and is a useful tool for ecological risk assessment and contaminated site management.

Keywords Heavy metals · Multimedia aquivalence model · Potential ecological risk · Petroleum exploration · Environmental behavior

Introduction

As global demand for petroleum has increased and as the petroleum industry has developed, environmental contamination and ecological degradation have also become increasingly serious in areas where petroleum is exploited. The petroleum exploitation process results in severe contamination of air, water, soil, sediment, and vegetation from, for example, evaporation of various petroleum products, oil spilled on the soil (Zhang et al. 2011), and discharges and irrigation of oily sewage (Khan et al. 2008; Hu et al. 2013a, 2013b; Kisic et al. 2009; Luo et al. 2015; Das and Kazy 2014; Marques et al. 2015) in the immediate vicinity of the petroleum exploitation and further afield. Petroleum exploitation can result in accumulation, amplification, and cycling of biological contaminants in environmental systems, which may threaten ecological health (Sharpe and Mackay 2000).

The contaminants that are derived from the petroleum exploration and drilling processes have very complex compositions and mainly comprise petroleum hydrocarbons and heavy metals. Heavy metals released from petroleum exploitation enter the environment by many different pathways; they are transported between adjacent compartments and are associated with a variety of physical, chemical, and biological transformation processes (Ötvös et al. 2004). Because of their high toxicity and bioaccumulation potential, we need to have information about the concentrations, reactivity potential, and

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distribution of heavy metals derived from petroleum exploitation in air, water, soil, sediment, and vegetation (Cao et al. 2007). Simulation, analysis, and prediction of the transport and fate of heavy metals will therefore provide a basis for risk management of petroleum exploitation areas.

Intermittent sampling is traditionally used to investigate the distribution of chemicals; however, analysis of different compartments may only provide information about their distribution in multimedia and may not include quantification of transport processes. In contrast, previous studies have shown that the multimedia fugacity model can provide useful simulations of the environmental behaviors of persistent organic pollutants (POPs) in multimedia (Beyer and Biziuk 2009; Hu et al. 2013c, 2013d; Li et al. 2012). The fugacity concept, however, is not suitable for non-volatile chemicals. Consequently, the aquivalence concept has been introduced as a new criterion for equilibrium and has been used to investigate the environmental behaviors of heavy metals in lake ecosystems (Mackay and Diamond 1989; Ling et al. 1993), and to simulate the transport and fate of inorganic ions (Hu et al. 2013a, 2013b). Many studies have reported the environmental effects of POPs in multimedia (Suzuki et al. 2000; Ao et al. 2009) and the potential ecological risks of heavy metals in environmental systems (Hou et al. 2013; Zhang et al. 2014; Yang et al. 2014), but few have simulated the environmental behaviors of heavy metals in multimedia environments. Therefore, we used a steady-state multimedia aquivalence (SMA) model to explore the environmental behaviors of five heavy metals (Cd, Cr, Cu, Pb, and Zn) in an oilfield in China. The modeling results will allow quantitative characterization of the environmental behaviors and ecological risks from the heavy metals in the oilfield that may be used to support decision making for contaminated site management.

Materials and methods

Study area

Daqing City is located in Heilongjiang Province, China (Fig. 1) (Yu et al. 2011) and covers a total area of 21,219 km^2. The monthly mean temperatures range from −18.5 °C in December to 23.2 °C in June, with an annual mean temperature of 4.2 °C. The annual average precipitation and evaporation are approximately 314.1 and 1257.5 mm, respectively. The average wind speed is 2.5 m/s, with a maximum speed of 12.3 m/s (Li et al. 2012). There are nearly 200 marshes in Daqing City that cover a total water area of 208 km^2 and have a total water capacity of 1.38×10^7 m^3. Daqing City comprises 8407 km^2 of vegetation, which accounts for about 40 % of the city's area.

Daqing Oilfield is the biggest oilfield and most important petrochemical industrial base in China (Song and Zheng 2002). Petroleum and natural gas resources are abundant in Daqing City, and more than 40 million tons of petroleum are extracted each year from several thousands of oil wells that are in daily operation. Contaminants from the petroleum exploitation are discharged into environmental systems via different pathways, such as volatilization into air, oil spills on clean soil (Zhang et al. 2011), and surface water contamination from discharges of oily wastewater (Grec and Maior, 2008).

Modeling framework and environmental processes

Previous studies have demonstrated that the fugacity approach can effectively describe the multimedia behavior of organic pollutants, but it cannot be applied to non-volatile chemicals because of mismeasurement of partition coefficients, such as metals, polymers, and ionic compounds, in different compartments. The QWASI model has been modified to incorporate the new equilibrium criterion of aquivalence Q (mol/m^3) (Mackay and Diamond, 1989; Diamond et al. 1990). As outlined by Diamond et al. (1990), the linear relationship between fugacity (f or Q) and concentration (C) is described as follows:

$$C = fZ = QZ \tag{1}$$

In aquivalence, the fugacity capacity (Z) of water compartments (Z_w) is defined as 1, and Q is the actual dissolved concentration (mol/m^3) in the water compartment. The fugacity capacity of the air compartment (Z_A) is negligible for a non-volatile chemical because the air-water partition coefficient (K_{AW}) is essentially zero. In the aquivalence approach, transfer fluxes (N) of chemicals in different environmental processes may be presented as the product of the transfer rate coefficient (D) and Q with identical units and may be added and compared when they apply to processes that originate in the same compartment.

The traditional steady-state mass balance for organic chemicals in multimedia environments can be described with the following equation:

$$E_i + G_{\mathrm{A}i} c_{\mathrm{B}i} + D_{ij} f_j - \left(D_{ji} + D_{\mathrm{R}i} + D_{\mathrm{A}i}\right) f_i = 0 \tag{2}$$

in which the subscript i (i = 1, 2, 3, 4, 5) represents the bulk compartments of air, water, soil, sediment, and vegetation, respectively; f_i is the fugacity of compartment i (Pa); E_i is the source emission rate into compartment i (mol/h); $G_{\mathrm{A}i}$ is the advection inflow rate of compartment i (m^3/h); $c_{\mathrm{B}i}$ is the background inflow concentration of regions adjacent to compartment i (mol/m^3); D_{ij} is the transfer rate coefficient from compartment i to j (mol/Pa/h), and $D_{\mathrm{A}i}$ and $D_{\mathrm{R}i}$ represent the advection outflow rate coefficient and the degradation rate coefficient of compartment i (mol/Pa/h), respectively.

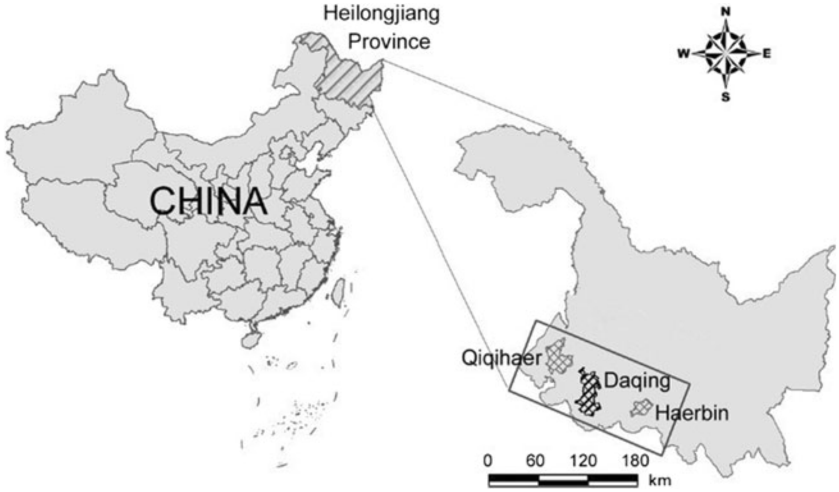


Fig. 1 Overview of the study area: Daqing City, Heilongjiang Province, China

When aquivalence is introduced, Eq. 2 may be rewritten as follows:

$$E_i + G_{\mathrm{A}i}c_{\mathrm{B}i} + D_{ij}Q_j - \left(D_{ji} + D_{\mathrm{R}i} + D_{\mathrm{A}i}\right)Q_i = 0 \qquad (3)$$

in which Q_i is the aquivalence of compartment i (mol/m^3).

A steady-state multimedia aquivalence (SMA) model that incorporated the above modifications was developed to describe the multimedia environmental behaviors of the chemicals of concern. In the SMA model, the environmental system was defined as an enclosed and homogeneous multimedia environment. All the transport and degradation reactions were described as first-order dynamic reactions, and air was assumed as a semi-infinite compartment. The concentrations in the air compartment showed little variation, and a fixed value was provided at the starting point from which to calculate the concentrations and aquivalences in the other compartments. The modeling framework and environmental processes considered in the SMA model are shown in Fig. 2.

Modeling parameters

The input parameters of the developed SMA model were the environmental attributes of Daqing City, physicochemical properties of the heavy metals, environmental transfers, and source emissions. We collected as many parameters as possible from the website of the local government (www.daqing.gov.cn) to represent the environmental characteristics of the study area. The major modeling parameters of the SMA model are listed in Table 1. In the absence of reliable data from the literature, default values were taken from Mackay and Paterson (1991) and Mackay (2001).

The emission rates (mol/h) of heavy metals to the air, water, and soil compartments are not always readily available and so are often estimated from the amount of energy consumed locally, petroleum extraction, and oil wells. Heavy metal emissions to the air compartment are mainly from combustion of biomass, coal, gasoline, diesel, and fuel oil. The emissions from these sources were calculated by multiplying the annual local energy consumption with the corresponding emission factors (EFs) (Tian et al. 2015). Source emissions in the water and soil compartments were estimated by measuring the annual volume of petroleum extracted from the study area, assuming that heavy metals from petroleum extraction were the main source in the petroleum exploitation area. Heavy metal emissions to the water compartment include discharges of drilling wastewater and well-flushing wastewater, and the emission rates of heavy metals to the water compartment were estimated from the amount of wastewater discharged, the heavy metal concentrations, and the removal rate in oily wastewater (Chen et al. 2001). The main input to the soil compartment was thought to be crude oil on the ground surface. The crude oil amount per oil well per year was 0.5–2.0 tons, and the recovery was estimated at 85 % (Zhang et al. 2012). The remainder was assumed to be released into the soils, i.e., emissions of heavy metals to the soil compartment. Thus, the estimated source emissions of the five heavy metals to the air, water, and soil compartments are listed in Table 2.

Results and discussion

Validation of the heavy metal modeling

When validating large-scale models, deviations of less than one logarithmic unit between the modeled and the measured values generally indicate that there is good agreement, and data from adjacent areas may also be used to validate the model reliability (Cao et al. 2007). In this study, data from actual measurements in the study area were used to validate the model reliability in the water and soil compartments (Du et al. 2011). We did not use data for the air, sediment, and vegetation compartments from other published studies of the area (Ye et al. 2007; Tang et al. 2012; Jiang and Zhao 2001;

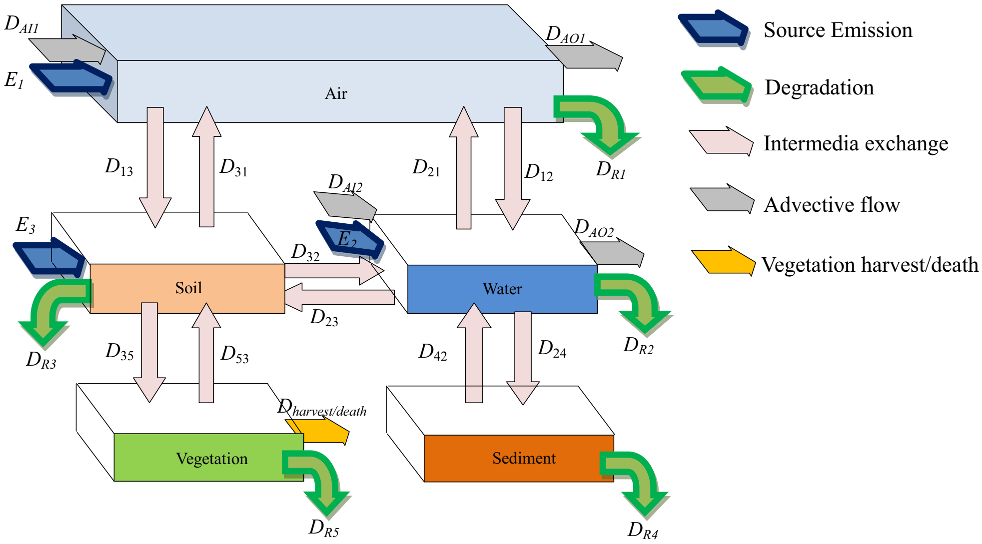


Fig. 2 Modeling framework and environmental processes contained in the SMA model

Zhang et al. 2007). The measured and the modeled concentrations of the five heavy metals are compared in Fig. 3. The calculated logarithmic residual errors of the SMA model were 0.69, 0.83, 0.35, 0.16, and 0.54 for the air, water, soil, sediment, and vegetation compartments, respectively, and indicated good agreement between the modeled and measured values.

The deviations between the modeled and measured values primarily reflect the following two aspects: (1) the modeled

Table 1 Environmental attribute parameters of the study area

The major modeling parameters of the SMA model

Parameters	Unit	Value	Parameters	Unit	Value
Area of air[a]	$\mathrm{m^2}$	2.12×10^{10}	Density of solids in sediment[c]	$\mathrm{kg/m^3}$	2.40×10^{3}
Depth of air[b]	m	2.00×10^{3}	Organic carbon in solids in sediment[b]	g/g	3.10×10^{-3}
Density of air[c]	$\mathrm{kg/m^3}$	1.19	Area of vegetation[a]	$\mathrm{m^2}$	8.41×10^{9}
Density of solids in air[c]	$\mathrm{kg/m^3}$	1.50×10^{3}	Vegetation transpiration rate[b]	$\mathrm{m^3/h \cdot m^2}$	0.02×10^{-3}
Area of water[a]	$\mathrm{m^2}$	1.47×10^{8}	Air side air-water MTC[b]	$\mathrm{m \cdot h^{-1}}$	3.00×10^{0}
Depth of water[a]	m	0.30	Water side air-water MTC[b]	$\mathrm{m \cdot h^{-1}}$	3.00×10^{-2}
Density of water[b]	$\mathrm{kg/m^3}$	1.00×10^{3}	Rain rate[a]	$\mathrm{m \cdot h^{-1}}$	9.70×10^{-5}
Density of solids in water[b]	$\mathrm{kg/m^3}$	2.40×10^{3}	Scavenging rate[b]	$\mathrm{m \cdot h^{-1}}$	2.00×10^{5}
Organic carbon in solids in water[b]	g/g	4.00×10^{-2}	Dry deposition rate[b]	$\mathrm{m \cdot h^{-1}}$	1.10×10
Area of soil[a]	$\mathrm{m^2}$	2.11×10^{10}	Air side air-soil MTC[b]	$\mathrm{m \cdot h^{-1}}$	1.00×10^{0}
Depth of soil[b]	m	0.5	Diffusion path length in the soil[b]	m	5.00×10^{-2}
Density of water[b]	$\mathrm{kg/m^3}$	1.21	Molecular diffusion coefficient in the air[b]	$\mathrm{m^2 \cdot h^{-1}}$	4.00×10^{-2}
Volume fraction of water in soil[b]	%	30	Molecular diffusion coefficient in the water[b]	$\mathrm{m^2 \cdot h^{-1}}$	4.00×10^{-6}
Density of solids in soil[c]	$\mathrm{kg/m^3}$	2.40×10^{3}	Runoff rate[b]	$\mathrm{m \cdot h^{-1}}$	1.14×10^{-6}
Organic carbon in solids in soil[b]	g/g	1.70×10^{-2}	Draining rate[b]	$\mathrm{m \cdot h^{-1}}$	2.30×10^{-8}
Volume fraction of air in soil[b]	%	20	Water side water-sediment MTC[b]	$\mathrm{m \cdot h^{-1}}$	1.00×10^{-2}
Area of sediment [a]	$\mathrm{m^2}$	1.47×10^{8}	Diffusion path length in the sediment[b]	m	5.60×10^{-3}
Depth of sediment[b]	m	0.5	Sediment deposition rate[b]	$\mathrm{m \cdot h^{-1}}$	4.60×10^{-8}
Density of sediment[c]	$\mathrm{kg/m^3}$	1	Sediment resuspension rate[b]	$\mathrm{m \cdot h^{-1}}$	1.10×10^{-8}
Volume fraction of water in sediment[b]	%	70	Sediment buried rate[b]	$\mathrm{m \cdot h^{-1}}$	3.40×10^{-8}

MTC mass transfer coefficient

[a] The values were derived from the website of the local government

[b] The values were derived from Mackay (2001)

[c] The values were derived from Mackay and Paterson (1991)

Table 2 The estimated source emissions of heavy metals to the air, water, and soil compartments (unit: mol/h)

Chemical	Source emissions to the air	Source emissions to the water	Source emissions to the soil
Cd	0.01	9.24×10^{-6}	151.64
Cr	1.62	3.38×10^{-5}	246.53
Cu	9.74	8.26×10^{-5}	444.32
Pb	3.22	1.96×10^{-5}	350.41
Zn	87.54	9.23×10^{-5}	312.55

values represented the average heavy metal levels in the study area, while the measured ones were collected from specific sites; (2) some parameters were obtained from published literature, and the lack of actual measured values may have introduced considerable uncertainty.

Transport and fate of heavy metals in the multimedia environments

The transport and fate of the five heavy metals in the multimedia environments were simulated by the SMA model. The relevance of the transport processes through the multimedia environments were evaluated by calculating the environmental transfer rates. The simulation results showed that, out of all the pathways, buried sediment (64.86 %) was the main output. Heavy metal accumulation in vegetation that was either harvested or died annually (18.96 %) was also an important transport pathway. The remaining heavy metals (16.18 %) were discharged from the multimedia environmental system by advective outflow with the wind.

The results showed that mass transport between water and sediment, which accounted for 49.25 % of all the transport processes, was the most significant process in the multimedia environments. Meanwhile, mass wet and dry atmospheric

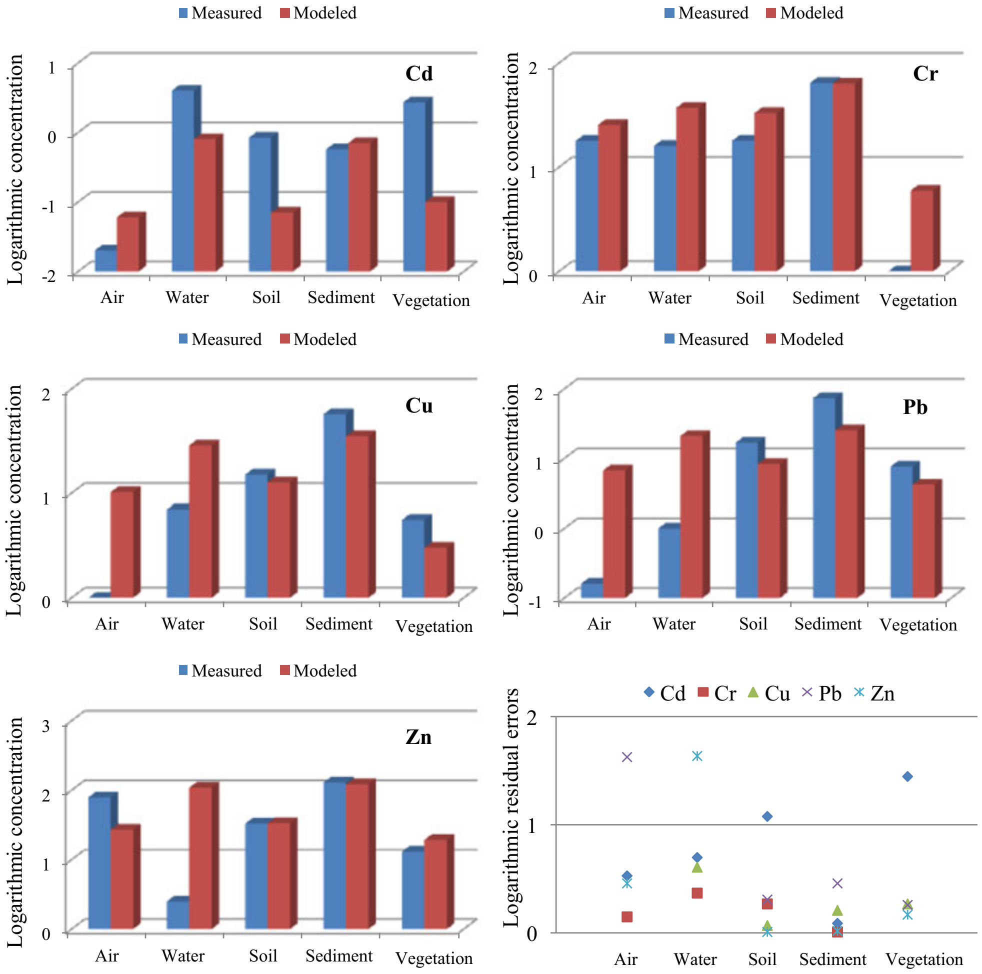


Fig. 3 Comparison of the measured and modeled heavy metal concentrations

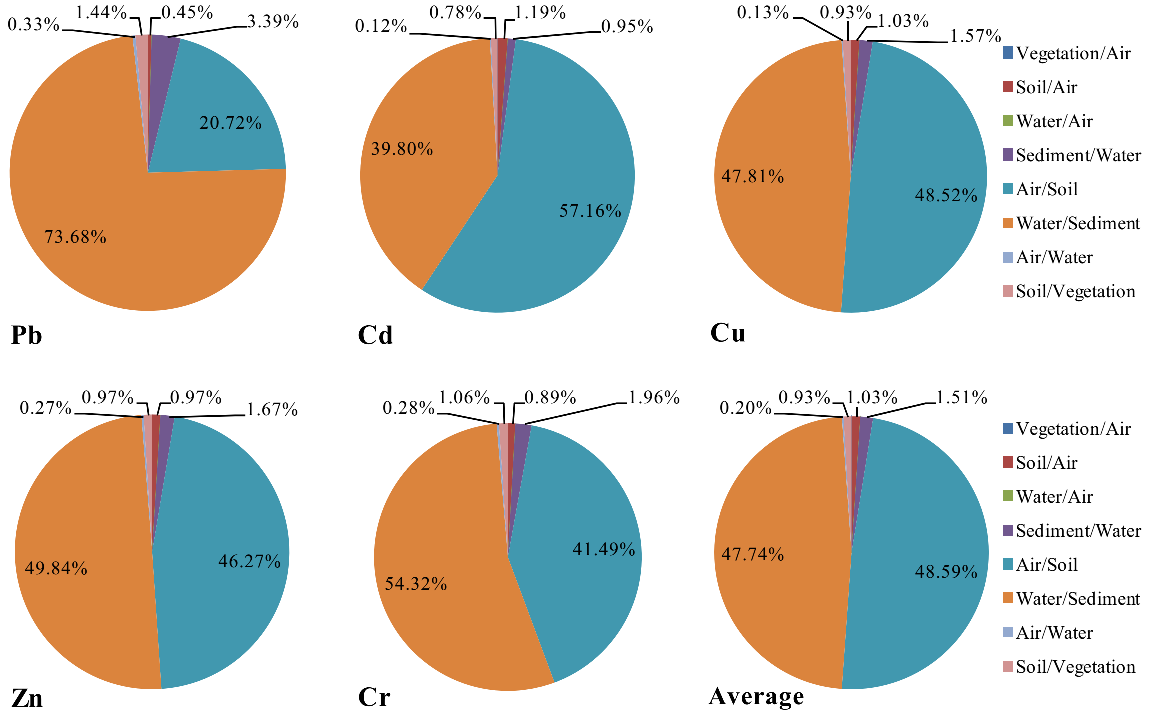


Fig. 4 The proportions of heavy metals exchanged between the multimedia environments

deposition from the air to the soil compartment was also an important pathway for heavy metals in the multimedia environments and accounted for 48.59 %. The mass transport proportions for the five heavy metals are shown in Fig. 4.

The total amounts of heavy metals in the multimedia environments were calculated from the modeled heavy metal concentrations and the volumes of the five bulk compartments, shown in Fig. 5. The sediment compartment was the main sink for the heavy metals, and accounted for 68.80 % of all the heavy metals in the multimedia environmental system. The heavy metal fractions in the soil and vegetation compartments accounted for 25.26 and 5.87 %, respectively. The amounts of heavy metals in the water compartment were relatively low, while the heavy metals in the air compartment were negligible and were therefore ignored. Thus, sediment and soil were the main sinks for heavy metals in the study area.

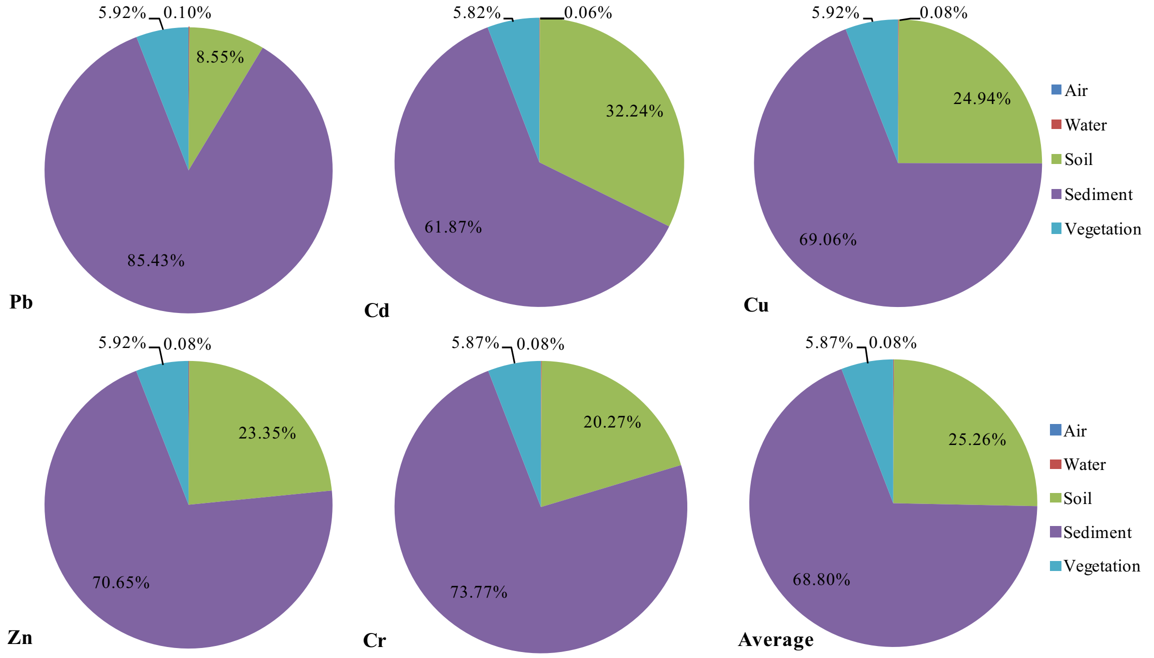


Fig. 5 Distributions of heavy metals in the multimedia environments

Sensitivity analysis of the SMA model

The main purpose of sensitivity analysis is to identify the major uncertainties and variabilities of the input parameters. The sensitivity coefficient (SC) is defined as the ratio of the relative variation of the estimated concentration to that of the input parameter.

$$\mathrm{SC}_i = \frac{\Delta Y_i / Y_i}{\Delta X_i / X_i} \tag{4}$$

where SC_i represents the sensitivity coefficient of input parameter i, and X_i and Y_i represent input parameter i and the corresponding estimated concentration (Ao et al. 2009). The calculation of SC could provide the influence significance for each input parameter.

The SC_i of input parameters to the modeled concentrations of five heavy metals in the five bulk compartments (steady-state) are shown in Fig. 6. The result indicates that the environmental attribute parameters (such as the areas and depths of the different compartments) and sources emissions in different compartments are the most sensitive input parameters for predicting heavy metal concentrations. Besides, the parameters related to rainfall and runoff rates in the study area are also contributed to the modeling results of the contaminant environmental behaviors.

Potential ecological risks from heavy metals in sediment and soil

The potential ecological risk index (PERI) presented by Håkanson (1980) was used to quantitatively assess the potential ecological risk level from the five heavy metals in the sediment and soil compartments. The calculations of the PERI are described in Eqs. 5 and 6.

$$E_{\mathrm{r}}^i = T_{\mathrm{r}}^i \times C_{\mathrm{f}}^i \tag{5}$$

Table 3 Potential ecological risks of heavy metals in the soil and sediment compartments

	C_{n}	T_{r}	E_{r}		Single PERL	
			Soil	Sediment	Soil	Sediment
Pb	35	5	1.19	3.60	Slight	Slight
Cd	0.2	30.00	10.58	105.70	Slight	Medium
Cu	35	5	1.81	5.02	Slight	Slight
Zn	100	1	0.34	1.22	Slight	Slight
Cr	90	2.00	0.73	1.41	Slight	Slight
IPERI			14.65	116.95		
Integrated PERL			Slight	Low		

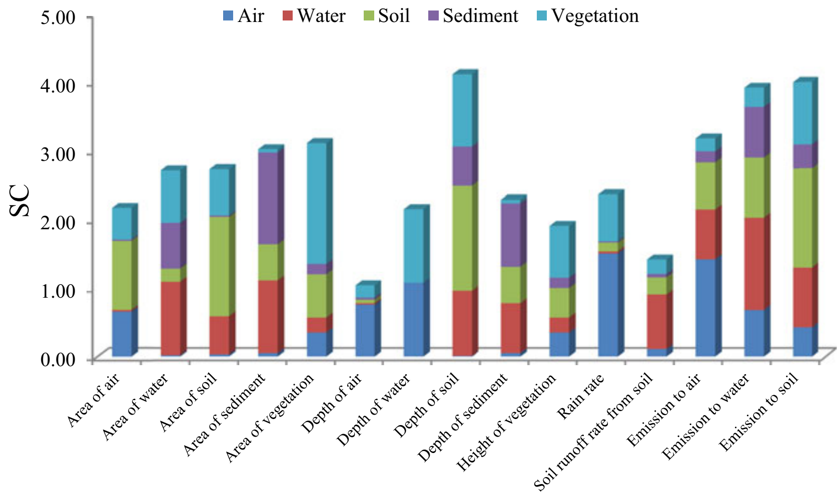
$$\mathrm{IPERI} = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \times C_f^i = \sum_{i=1}^{n} T_r^i \times C_s^i / C_n^i \tag{6}$$

in which E^i_{r} is the single PERI for the ith heavy metal; T^i_{r} is the toxic coefficient for the ith heavy metal; C^i_{f} is the concentration coefficient for the ith heavy metal; C^i_{s} is the actual/modeled concentration for the ith heavy metal in milligram per kilogram; C^i_{n} is the reference concentration for the ith heavy metal in milligram per kilogram, and IPERI is the integrated PERI for all the heavy metals.

The ecological risk calculated by PERI is divided into five levels, namely slight ($E^i_{\mathrm{r}} < 40$, PERI < 90), low ($40 \le E^i_{\mathrm{r}} < 80$, $90 \le$ PERI < 180), medium ($80 \le E^i_{\mathrm{r}} < 160$, $180 \le$ PERI < 360), high ($160 \le E^i_{\mathrm{r}} < 320$, $360 \le$ PERI < 720), and very high ($320 \le E^i_{\mathrm{r}}$, $720 \le$ PERI). The E^i_{r}, PERI, and the corresponding potential ecological risk levels (PERL) of the five heavy metals in the study area are shown in Table 3.

Out of the five heavy metals, the single PERL for Cd was the highest and reached medium in the sediment (Table 3). The single PERLs for the other four heavy metals (Cr, Cu, Pb, and Zn) were slight. The integrated PERLs for Cd in the soil and sediment compartments were slight and medium, respectively. The quantitative contributions of the five heavy metals to the

Fig. 6 Relative sensitivity of input parameters in various compartments



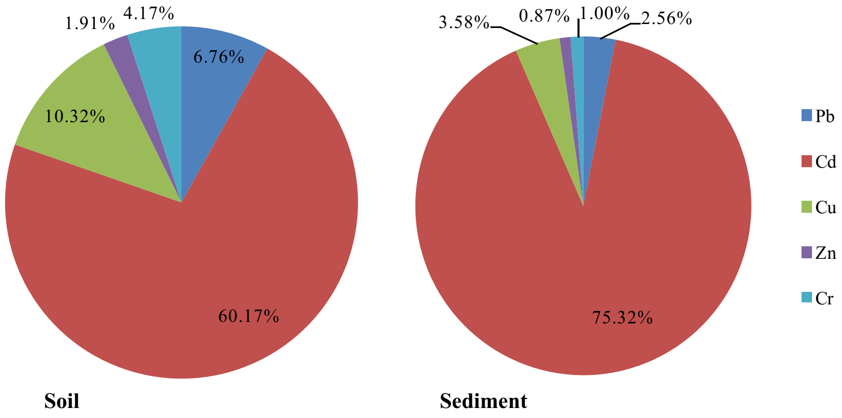


Fig. 7 Quantitative contributions of the five heavy metals to the integrated ecological risk

potential ecological risk are shown in Fig. 7. The highest risk contributor is Cd, which accounts for 60.17 % in the soil and 75.32 % in the sediment, followed by Cu and Pb; the contributions of Cr and Zn are relatively low.

The heavy metals of Cr and Cd have the strongest transfer ability between soil-vegetation environmental systems, and the most typical vegetations in the study area are reed and seepweed. Reed plays an important role in the sewage treatment and has well resistance to oil contamination because of the leaf, stem, and roots of reed are aerenchyma. Besides, herbaceous vegetations (such as seepweed, green bristlegrass) in the study are the most sensitive to the oil spilled on the soil. The ground part of the vegetations will die when the oil contamination is not serious, and all the vegetations will die out if the oil contamination is serious. Thus, the contamination of heavy metals in the oilfield must be effectively controlled to avoid the ecological risk to surrounding environmental system and human health. Further researches into potential risks of heavy metals derived from the petroleum exploration for soil including soil organisms as well as for vegetation are still needed. More information on transformation of various forms of heavy metals, such as metal ion and metallic compound should be considered in the further development and application of multimedia aquivalence model, which will provide scientific guidance to for environmental behaviors simulation and risk management of contaminant in the petroleum exploitation area of China.

Conclusions

Multimedia environmental behaviors and potential ecological risks of five heavy metals in air, water, soil, sediment, and vegetation compartments in a Chinese oilfield were investigated using a steady-state multimedia aquivalence model. The leading input (source emission in soil), output (buried in sediment), and major transfer process (from water to sediment and from air to soil) were verified based on transfer flux calculation. Sediment was the dominant sink for heavy metals. Sensitivity analysis revealed that the environmental attribute parameters and source emissions in different compartments were the most sensitive input parameters for adjusting uncertainties of heavy metal concentrations in multimedia environments. Cd was the most significant contributor to the integrated potential ecological risk in the study area.

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