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Influence of long-term sewage irrigation on the distribution of organochlorine pesticides in soil–groundwater systems

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HIGHLIGHTS

- ▶ HCH, DDT and Endosulfan compounds are main OCPs in the study area.
- ▶ Over-exploitation of groundwater has changed the flow field of groundwater.
- ▶ Positive correlations occurred between OCPs content in soil and in groundwater.
- ▶ Sewage irrigation influenced the distribution of OCPs in soil–groundwater systems.
- ▶ Groundwater flow and current pesticide use were two major influence factors.

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ABSTRACT

Serious shortage of water resources is one of the major factors restricting the sustainable development of cropland and pasture land in northern and northwestern China. Although the reuse of wastewater for agricultural irrigation becomes a well established practice in these regions, many contaminants have been also introduced into the soil–groundwater systems such as persistent organochlorine pesticides (OCPs). To study the influence of long-term sewage irrigation on the distribution of OCPs in soil–groundwater systems, the groundwater flow field was investigated and 31 topsoil samples, 9 boreholes, 11 sewage effluents and 34 groundwater samples were collected in Xiaodian, Taiyuan city, one of the largest sewage irrigation districts, China. During sampling, three representative types of regions were considered including effluent-irrigated area, groundwater-irrigated area served as the control field and no-irrigated area as reference “background”. The results showed over-exploitation of groundwater had changed the flow field of groundwater and wherever in soil or in groundwater, the concentration of OCPs in effluent-irrigation area presented the highest value, which indicated that the sewage irrigation had a strong influence on the distribution of OCPs in soil–groundwater systems. Principal component analysis for OCPs content in groundwater showed that the major influence factors on the occurrence and distribution of OCPs in groundwater systems attribute to the flow field of groundwater and to the current pesticide use.

1. Introduction

With fast economic development and accelerated urbanization, high water demands put a great pressure on water supply and become a major factor restricting the sustainable development of agriculture in arid and semiarid areas such as in northern and northwestern China. Due to its advantages, sewage irrigation has been widely used in many countries as an important supplement to water supply, to reuse nutrients contained in sewage to benefit agriculture production (Yang and Abbaspour, 2007), and to remove pollutants during irrigation (Duan and Fedler, 2010). However, many countries lack regulations to manage sewage irrigation on the agricultural use of such residues (Qadir et al., 2010). As a result of a long term sewage irrigation, soil quality degradation such as change of soil properties (Jalali et al., 2008; Walker and Lin, 2008), increase in nitrate (NO_3^-) concentration (Polglase et al., 1995; Smith and Bond, 1999), accumulation of heavy metals (Liu et al., 2005; Yang et al., 2008), and introduction of organic contaminants (Gibson et al., 2010; Chen et al., 2011a, 2011b). Therefore, the reclamation of wastewater has been recognized as an important source of toxic contaminants to the environment (Li et al., 2008) and some organic components of wastewaters may enhance the solubilization and leaching of recalcitrant compounds in the soils (Gonzalez et al., 2010), which bring potential human health risk through food-chain (Chiou, 2008; Chary et al., 2008).

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Although the accumulation and transport of nitrate, pathogens and heavy metals in the subsurface environment have caused attentions among environmental scientists and managers, little work has been done on the influence of long-term sewage irrigation on the distribution of some trace organic pollutants such as organochlorine pesticides (OCPs) in soil–groundwater systems. Despite of being banned for agriculture in China in 1983, OCPs have been detected in topsoil (Feng et al., 2003; Zhang et al., 2009a, 2009b) and subsoil (Zhu et al., 2005; Zhang et al., 2009a, 2009b), also in wastewater (Chen et al., 2011a, 2011b) and groundwater (Zhao and Pei, 2012). Their presence may be attributed to recent usage of pesticides (Wei et al., 2007; Li et al., 2008) or to their leaching or desorption from the soil as an important reservoir of OCPs under the impact of irrigation or rainfall (Chen et al., 2005; Zhang et al., 2009a, 2009b). But the distribution and transportation mechanisms of OCPs present often as site-specific (Weaver et al., 2012). The aim of this study was to investigate the contamination levels of OCPs in the selected sewage irrigation area; and to evaluate the influence of sewage irrigation on the distribution of OCPs in soil–groundwater systems in the study area.

2. Materials and methods

2.1. Study site

In the Xiaodian irrigation area for this study, one of the largest sewage irrigation districts in China, wastewater has been used as a source for irrigation for more than 30 years. Xiaodian is located in the southeast part of Taiyuan City, which is a representative semi-arid area in northern China. The annual rainfall is 464 mm while annual evaporation is 1045 mm. Groundwater has been the most important source of water supply here. It belongs to an alluvial plain from the Fenhe River to the west, the Xiao River to the South and extended to the East Mountain in the northeast covering 295 km^2 between 37°36′ ~ 37°49′N latitude and 112°24′ ~ 112°43′E longitude. The altitude lowers from the north to the south and from the east to the west. In 1981, the major groundwater flow is from northeast to southwest (Shanxi Geological Survey, 1981). However, according to our investigation about groundwater level in April, 2009 (Du, 2011), the equal value plot of shallow groundwater level is shown in Fig. 1. It is obvious that a long time of artificial extracting of groundwater has changed the flow field of groundwater, where three groundwater funnels and four local flow systems of groundwater had been formed (labeled as I, II, III, and IV in Fig. 1) and over-exploitation of groundwater become one of major causes to contribute local serious land subsidence (Ma et al., 2006).

There are two types of aquifers at and around Xiaodian: Quaternary aquifers and karst and fissure water aquifers of bedrocks (Shanxi Geological Survey, 1981). The Quaternary aquifers can be vertically divided into three major groups of aquifers which are separated by a set of clay layers: the Holocene and upper Pleistocene (No. 1) aquifer at depths of 0–60 m bls (below the land surface), the middle Pleistocene (No. 2) aquifer with depths of 60–141 m bls and the lower Pleistocene confined (No. 3) aquifer with at depths of 141–485 m bls. For the No. 1 aquifer, the sediments consist of interlayer sand and clay, as a result of alternating deposition of alluvial and lacustrine sediments; for the No. 2 aquifer, the sediments consist of medium sand and medium coarse sand with gravel in the northern part, and of fine-medium coarse sand with interlayers of clay in the southern part; for the No. 3 aquifer, the sediments consist of fine-medium coarse sand with sporadically occurring gravels. There are two major soil types in the study area: silty clay and silt, with hydraulic conductivity of 3.23×10^{-3} and 5.43×10^{-4}, respectively. Generally, groundwater receives recharge mainly via vertical seepage of meteoric water in the upper aquifer and via lateral penetration of karst water and fissure water along the mountain front. In addition, leakage from local rivers and irrigation return flow should also be taken into account as groundwater recharge sources. The major ways of discharge include evaporation and artificial abstraction.

Xiaodian has been traditionally associated with intensive agricultural activities. Crops grown in this area include wheat, corn, sorghum, rice and vegetables. Sewage irrigation has been in practice since 1970s. At the beginning, surface water from Fenhe River was used to irrigate the crops, and then replaced gradually by municipal sewage due to the decrease of available surface water. Industrial wastewater was used by mixing with municipal sewage as irrigation source in 1990s. Flood-irrigation method was adopted and the wastewater used as irrigation now comes mainly from municipal sewage of the city and industrial wastewater from nearby industries. In our previous studies, due to years of wastewater irrigation, heavy metals such as Cu, Cr, Pb, Zn, and Ni were detected in the unsaturated zone of our study area with 3–8 times more than that in the soil background field (Du, 2011).

2.2. Sampling

Irrigation water including surface water from the rivers (Xiaohe River and Fenhe River) and the wastewater of three canals (East Main Canal, Beizhang Drainage and Taiyu Drainage) (labeled as SW), groundwater (labeled as GW), topsoil (labeled as SS) and profile subsoil (labeled as GS) samples were collected in August, 2010. The sampling sites are illustrated in Fig. 2, which were selected based on sewage irrigation history and on our previous research work in this area. Three major factors were considered to select sampling sites: effluent-irrigated area around the East Main Canal, Beizhang Drainage and Taiyu Drainage, groundwater-irrigated area served as the control such as the field near GW-18 and no-irrigated area located in the front of the East Mountain as reference "background" sites (near GW-51). Groundwater was sampled mainly from the Nos. 1 and 2 aquifers.

Groundwater samples were collected by suction pumps after pumping 15 min and surface water was directly sampled. Unstable parameters such as the electric conductivity (EC), pH, water temperature (T) and groundwater level were measured in situ. Groundwater samples for OCPs analysis were collected directly in clean, dry, 4.5 L amber glass bottles covered with clean aluminum foil. For surface water, 1 L amber glass bottles were used. Topsoil samples were taken from a depth of 10–15 cm with the grid 2 km × 2 km and 9 boreholes were dug using a shovel to get profile subsoil samples from different depth intervals (0–10, 10–20, 20–30, 30–40, 40–50, 50–70, 70–90, and 90–110 cm). All soil samples were taken in clean aluminum boxes and sealed by airtight strips. Field work was completed in 7 d (August 2010) to rule out any seasonal effects. Samples were kept at ambient temperature in a holding box and transported immediately to the laboratory where they were stored in a refrigerator until processed. Soil samples for OCPs analysis were air dried (about 10 °C) and soxhlet extracted in 1 week.

2.3. Analysis methods

2.3.1. Sample extraction and fractionation

The analytical method used for the extraction and clean-up of OCPs in soil and water was expounded in the previous studies (Chen et al., 2011a, 2011b; Gong et al., 2007). Briefly, for soil samples, 10 g of soils were spiked with 20 ng of 2,4,5,6-tetrachoro-*m*-xylene (TC*m*X) and decachlorobi-phenyl (PCB_{209}) for OCPS as recovery surrogates, and then were Soxhlet-extracted with dichloromethane for 24 h. For water samples, 1 L of groundwater but