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Q2 Adsorption–desorption behavior of atrazine on agricultural soils in China

Q3 Lin Yue¹, ChengJun Ge^{1,2,*}, Dan Feng¹, Huamei Yu^{1,2}, Hui Deng¹, Bomin Fu¹

1. School of Environment and Plant Protection, Hainan University, Haikou 570228, China

2. Key Laboratory of Protection and Development Utilization of Tropical Crop Germplasm Resources (Hainan University), Ministry of Education, Haikou 570228, Hainan, China

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ABSTRACT

Adsorption and desorption are important processes that affect atrazine transport, transformation, and bioavailability in soils. In this study, the adsorption–desorption characteristics of atrazine in three soils (laterite, paddy soil and alluvial soil) were evaluated using the batch equilibrium method. The results showed that the kinetics of atrazine in soils was completed in two steps: a “fast” adsorption and a “slow” adsorption and could be well described by pseudo-second-order model. In addition, the adsorption equilibrium isotherms were nonlinear and were well fitted by Freundlich and Langmuir models. It was found that the adsorption data on laterite, and paddy soil were better fitted by the Freundlich model; as for alluvial soil, the Langmuir model described it better. The maximum atrazine sorption capacities ranked as follows: paddy soil > alluvial soil > laterite. Results of thermodynamic calculations indicated that atrazine adsorption on three tested soils was spontaneous and endothermic. The desorption data showed that negative hysteresis occurred. Furthermore, lower solution pH value was conducive to the adsorption of atrazine in soils. The atrazine adsorption in these three tested soils was controlled by physical adsorption, including partition and surface adsorption. At lower equilibrium concentration, the atrazine adsorption process in soils was dominated by surface adsorption; while with the increase of equilibrium concentration, partition was predominant.

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Introduction

The herbicide atrazine (2-chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine) is widely used to control certain annual broadleaf and grass weeds, primarily in corn, but also in sugarcane, sorghum, other crops, and landscape vegetation to some extent. Atrazine was developed from research on derivatives of symmetrical triazines begun in 1952 (Knuesli, 1970). Atrazine has been a main herbicide worldwide since 1958, when atrazine was patented in Switzerland and then registered for commercial use in the United States (U.S. Environmental Protection

Agency, 1994). Up to now, atrazine has been registered in more than 70 countries worldwide (Kauffmann et al., 2000; Zhang et al., 2004). In China, current annual atrazine usage is estimated at 10–15 million kg (Deng et al., 2005). However, widespread atrazine use is associated with increasing incidence of contamination in drinking water, with atrazine concentrations above the maximum contaminant level of 0.1 ppb (Guzzella et al., 2006). Atrazine and its degradation products, deethylatrazine and deisopropylatrazine, have been frequently detected in soils (Chung and Alexander, 2002), sediments (Jantunen et al., 2008), surface water (Al-Degs et al., 2009) and

* Corresponding author. E-mail: cjge3007@163.com (ChengJun Ge).

groundwater (Hildebrandt et al., 2008) in many countries. Many recent studies have reported that atrazine and its metabolites are endocrine-disrupting chemicals and may pose potential risks to human health (Geng et al., 2013).

Atrazine accumulates in the soil due to its low chemical reactivity, leading to contamination of groundwater (Frank and Sironi, 1985). The environmental behavior of atrazine in a soil depends upon several factors, including adsorption by soil components, absorption by plants, transportation through runoff and leaching, volatilization, biodegradation, and chemical degradation (Deng et al., 2010). Adsorption and desorption of herbicides by soils play an important role in influencing the fate of herbicides in soil environments (Lesan and Bhandari, 2003; Wu et al., 2011). Close relationship between the adsorption-desorption behaviors of herbicides and the characteristics of soils (Wu et al., 2009), such as clay content and type, ionic strength (Ureña-Amate et al., 2005), soil pH (McGlamery and Slife, 1996), soil organic matter content (Seol and Lee, 2000), soil porous structure (Wang and Keller, 2008) and particles' specific surface area (Tang et al., 1998). Organic matter has been frequently reported to be the most important factor that influences adsorption of atrazine in soil, sediments or in solution (Lesan and Bhandari, 2003). Kulikova and Perminova (2002) reported that atrazine adsorption to soil humic substances was closely correlated to the aromaticity of the soil organic matter. It was showed that the quality of the soil organic matter plays an important role in the atrazine adsorption processes and hysteresis (Kempf and Brusseau, 2009). Most previous studies were conducted to study adsorption of atrazine on humic substances (Sun et al., 2010). Additionally, atrazine adsorption behaviors on biochars (Cornelissen et al., 2005), clays (Laird et al., 1992) and oxyhydroxides (Laird et al., 1994) were also investigated. Desorption and adsorption are of equal importance in determining the fate of atrazine in soils (Moreau and Mouvet, 1997), however, the adsorption and desorption of atrazine in different types of soils are poorly documented. Therefore, adsorption-desorption behavior of atrazine in soils is required to be further explored.

Various physical and chemical mechanisms are involved in herbicides adsorption in adsorbents. They may act separately or together. Hydrogen bonds, Van der Waals forces and hydrophobic bondings are common mechanisms (Moreau and Mouvet, 1998). Senesi (1992) suggested that atrazine can also be adsorbed by ionic bonding on humic acid. Barriuso et al. (1994) showed that smectites absorbed atrazine primarily through relatively weak Van der Waals forces or hydrogen bonds. Piccolo et al. (1992) reported that charge transfer is a specific mechanism for atrazine adsorption to humic acids. However, little is known about the mechanisms involved in atrazine adsorption on soils.

The objective of the present study was to understand the sorption and desorption characteristics of atrazine in three agricultural soils and to reveal the adsorption and desorption mechanisms, so as to provide a scientific basis for environmental risk assessment and ecological restoration of atrazine-contaminated soils and theoretical support for agricultural product quality and safety.

1. Materials and methods

1.1. Chemicals

Standard atrazine with >99.9% purity was purchased from Dr. Ehrenstorfer, Germany. The water solubility of atrazine is 33 mg/L, and the pK_a is 1.7 (Chefetz et al., 2004). Methanol used in this study was high-performance liquid chromatography (HPLC) grade from Sinopharm Chemical Reagent Co. Ltd., China. Deionized water (18.25 M Ω) was obtained from a Spring-S60i + S60i + PALL system. All other reagents were analytically leveled.

1.2. Soil characterization

The laterite and paddy soil used in sorption experiments were collected from Danzhou and Qiongzhong respectively in Hainan Province, China. Alluvial soil came from the Sixth Ring Road, Beijing, China. The soil samples were taken from 0 to 20 cm soil layers and were air-dried. All samples were sieved through a 1-mm mesh and sealed in a container for later use. The basic properties of the soils are summarized in Table 1.

1.3. Adsorption and desorption experiments

An atrazine adsorption experiment was carried out using a batch procedure (Chefetz et al., 2004). An appropriate amount of soil was transferred to 50-mL centrifuge tubes, and 10 mL of atrazine solution (in background solution of 0.01 mol/L CaCl₂ to maintain a constant ionic strength and 0.2 g/L of NaN₃ to inhibit microbial activity, with pH 7) was added at initial atrazine concentrations of 0.5, 1, 5, 10, and 20 mg/L. All tubes were immediately sealed and then mechanically shaken for 24 hr in a thermostatic oscillation incubator at 25°C, except for the thermodynamic experiments, in which temperatures of 15 and 35°C were adjusted and then the suspensions were centrifuged at 5000 r/min for 5 min. A 2-mL supernatant was filtered through the membrane with a pore size of 0.45 μ m and was then analyzed by HPLC. Each processing sets three

Table 1 – Physical and chemical properties of three agricultural soils.

Property	Soil sample		
	Laterite	Paddy soil	Alluvial soil
pH (CaCl ₂)	4.19 ± 0.01	5.21 ± 0.01	7.50 ± 0.01
Organic matter (%)	3.23 ± 0.02	4.19 ± 0.03	6.37 ± 0.03
Cation exchange capacity (cmol/kg)	7.47 ± 0.08	7.16 ± 0.15	7.24 ± 0.10
Sand (%)	34.12 ± 0.40	42.09 ± 0.29	24.50 ± 0.18
Silt (%)	40.19 ± 0.61	23.92 ± 0.72	59.27 ± 0.55
Clay (%)	25.68 ± 0.30	33.99 ± 0.18	16.23 ± 0.21

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