



# Sorption of tylosin on black carbon from different sources



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## ABSTRACT

Sorption of antibiotics on different sources black carbons are poorly understood, thus tylosin (TYL) sorption on different sources (RC, MC, and SC) was determined from aqueous solution, and the sorption characteristics and mechanism were investigated. The pseudo-second-order model provided the best fit for the sorption kinetics of TYL on black carbons and the sorption isotherms were described best with the Slips model. The sorption capacities were RC > MC > SC but the sorption rates were SC > MC > RC and the sorption equilibrium could be attained within 24 h. RC showed relatively higher efficiency for TYL sorption for composition of iron oxide. Solution chemistry such as pH and ionic strength show obvious effect on TYL sorption. The sorption mechanisms of TYL on black carbons were contributed to hydrophobic force, partition process and  $\pi$ - $\pi$  electron donor-acceptor interaction. These observations are useful for reduce the bioavailability of antibiotics and/or predict the fate of antibiotics in black carbon-amended soils.

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## 1. Introduction

Black carbon (BC) refers to the carbonaceous materials generated from incomplete combustion of fossil fuel and biomass and is commonly present in natural soils, characterized by condensed, rigid, and aromatic structures, with high carbon contents and relatively few polar functional groups [1]. Soot is the product of devolatilization reactions after the incomplete combustion of fossil fuel and other carbonaceous materials [2]. Char and charcoal are the carbonaceous solid residues produced after the oxidation and pyrolysis processes of carbonaceous materials in the environment [3]. These substances exhibit a high sorption capacity for organic contaminants in the environment [1,3,4]. Recently, BC received increasing attention because of their potential roles as a possible soil amendment to increase fertility and sequester carbon, as well as a potential low-cost adsorbent to control pollutant migration [5–8]. In the previous studies BC exhibited a high sorption capacity for organic contaminants in the environment and also indicated the importance of BC for the environmental sorption process [9–11]. BCs can be regarded as adsorbents ubiquitous in soil and aquatic environments, consequently influencing the chemical fate in the environment [1,3]. This

implied that the role of BC in modeling the sorption of organic compounds in soils and surface waters must be considered [5,6,9,11].

Black carbon can also be considered structurally similar to activated carbon, consisting primarily of short stacks of graphite sheets with O-containing groups rimmed on the edge to form connected microporous networks [12–14]. Owing to the large specific surface area and high surface hydrophobicity, black carbon often shows extraordinarily strong adsorption affinity for hydrophobic organic contaminants [3]. It is well recognized that carbonaceous geosorbents (black carbon, humin/kerogen, and coal) play a key role in soil/sediment sorption of nonpolar hydrophobic organic compounds such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and chlorinated benzenes, in particular at low solute concentrations [6,7,9,11,12,14]. However, studies on sorption of polar and ionic compounds on environmental black carbon are very limited in literature, and have mainly involved pesticides.

Tylosin (TYL) was produced in large quantities and extensively used in the farming industry as veterinary therapeutics and growth promoters [15]. It given to livestock are often poorly metabolized and absorbed, and a considerable fraction is consequently released into the environment, has received a great deal of attention in recent years [16,17]. Sorption to soils/sediments is a fundamental process controlling the fate, bioavailability, exposure, and reactivity of organic contaminants [18]. Thus, it is of great importance to

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evaluate the relative importance of different soil components to the overall sorption of TYL. Despite the potential importance, unexpectedly no adequate attention has been paid to sorption of pharmaceutical antibiotics on environmental black carbon. Studies on antibiotic sorption by natural geosorbents have focused on soils, iron/aluminum hydroxides, clay minerals, and humic substances [18–20]. But for the properties of their physical, chemical and surface properties, the sorption mechanism of antibiotics on the geosorbents could not suit for black carbon.

In this study, rice straw charcoal (RC), motor vehicles of fly ash (MC), and soot (SC) were selected as representatives of BC particles with different characteristics. Based on the information about surface properties among different BCs, sorption properties and effect of pH and ionic strength on the sorption capacity of TYL on different BC were studied and then the related mechanisms were discussed.

## 2. Materials and methods

### 2.1. Materials and preparation

Tylosin tartrate (purity > 95%) was purchased from Sigma-Aldrich Corporation (St Louis, MO). The molecular structures and physicochemical properties were listed in Fig. 1. TYL is a weak base with a  $pK_a$  of 7.1 and molecular weight of 916.14 g/mol [21]. In acidic condition, there might be formed ionic bonds between protonated TYL and anionic components of soil and manure matrices [22]. Acetonitrile and methanol (HPLC grade, Merck Chemicals Co. AQ5) were used as received. Pure water was prepared by Milli-Q® water machine (Millipore Co., Huainan, China). All the other chemicals were analytical reagent grade and used without further purification. The rice straw charcoal, motor vehicles of fly ash, and soot were collected and prepared as reported by Xia et al. [23].

Primary stock solutions of TYL at 1000 mg/L were prepared with pure water and stored at 4 °C for a maximum of 1 month. The work solutions were prepared by diluting stock solution using 0.01 M  $KNO_3$  solution.

### 2.2. Characteristic of RC, MC, and SC

To eliminate inorganic components, the obtained black carbon products were treated by rinsing them with a mixture of 1 M HCl

and 1 M HF, followed by rinsing with distilled water several times to remove any residual acids. After purification, the samples were dried at 105 °C for 24 h and milled to a 100-mesh sieve. Total C, nitrogen (N) and hydrogen (H) contents in RC, MC and SC were determined with a CHN Elemental analyzer (CarloErba NA-1500). The microscopic features of the hydrochar and MPC were characterized by SEM (XL300, Philips) equipped with an energy-dispersive X-ray (EDX, Link 300) analyzer. X-ray diffraction (XRD) patterns were collected using a PANalytical X' Pert PRO (Almelo, The Netherlands). X-ray diffractometer with a Ni filter, Cu  $K\alpha$  radiation source ( $\lambda = 0.154$  nm), and angular variation of 10–70° operated at a tube voltage of 40 kV and a tube current of 40 mA. The specific surface areas of RC, MC and SC were calculated according to the Brunauer-Emmett-Teller (BET) method. Their functional groups were characterized using Fourier transform infrared spectrometer (FTIR) (Nexus 870, Nicolet, USA) and Raman spectroscopy (Jobin Yvon T64000). Atomic force microscopy (AFM) was carried out with a multimode AFM Autoprobe CP II (Veeco).

### 2.3. Sorption procedure

The sorption experiments were conducted using a batch equilibrium technique at 25 °C. The initial TYL concentrations were set from 0.5 to 50 mg/L. The background solution contained 0.003 M  $NaN_3$  to minimize bioactivity and 0.01 M  $KNO_3$  to adjust ionic strength. A predetermined amount of sorbent was filled with the initial aqueous solution in completely mixed batch reactor (CMBR) systems with teflon gaskets and mixed for sorption equilibrium on a shaker at 150 rpm. After the sorption experiments, the screw cap vials were centrifuged at 4000 rpm for 30 min, and 1 mL of supernatant was transferred into a pre-weighed 1.5 mL amber glass vial for chemical analyses. Each concentration level, including blanks, was run in triplicate. KOH or  $HNO_3$  solutions were used for pH adjustment. Kinetic studies of TYL sorption were carried out from aqueous solutions with a certain concentration 5 mg/L and pH. A mixed volume of the aliquot was withdrawn at designated time points while the reactors were run continuously.

### 2.4. Chemical analysis

The concentrations of TYL in aqueous solution were measured by a reverse-phase high-performance liquid chromatography (Hitachi D-2000 Elite-HPLC) with  $C_{18}$  column (5  $\mu$ m, 4.6  $\times$  250 mm; Agilent) and diode array UV detector (wavelength at 290 nm for TYL). The mobile phase (at a flow rate of 0.5 mL/min) for TYL was a mixture of acetonitrile (35%) and an aqueous solution (65%) containing 0.01 mol/L  $KH_2PO_4$  (pH = 2.0). The injection volume was 20  $\mu$ L. External standards of TYL (0.1–100 mg/L) were employed to establish a linear calibration curve and the sample concentrations were calculated from its integrated peak areas. The solid phase concentrations were calculated based on the mass balance of the solute between the two phases.

## 3. Results and discussion

### 3.1. Characterization of adsorbents

Carbon (C) was the dominant element in RC, MC, and SC (Table 1). The C content composition were SC > MC > BC. Oxygen (O) content in RC was about 3 times that in SC, indicating higher degree of oxidation. Surface properties are important properties which may affect chemical and physical interactions on black carbons surfaces. Compared to that of MC, the BET surface area was slightly higher in SC, but lower in RC (Table 1).

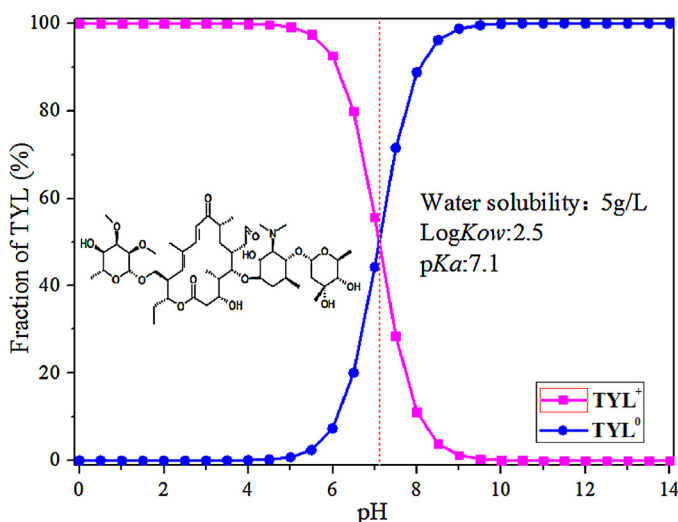


Fig. 1. Percent ionization at different pH, chemical structure, and selected properties of TYL.

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