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Transfer of antibiotics from wastewater or animal manure to soil and edible crops☆

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ABSTRACT

Antibiotics are added to agricultural fields worldwide through wastewater irrigation or manure application, resulting in antibiotic contamination and elevated environmental risks to terrestrial environments and humans. Most studies focused on antibiotic detection in different matrices or were conducted in a hydroponic environment. Little is known about the transfer of antibiotics from antibiotic-contaminated irrigation wastewater and animal manure to agricultural soil and edible crops. In this study, we evaluated the transfer of five different antibiotics (tetracycline, sulfamethazine, norfloxacin, erythromycin, and chloramphenicol) to different crops under two levels of antibiotic-contaminated wastewater irrigation and animal manure fertilization. The final distribution of tetracycline (TC), norfloxacin (NOR) and chloramphenicol (CAP) in the crop tissues under these four treatments were as follows: fruit > leaf/shoot > root, while an opposite order was found for sulfamethazine (SMZ) and erythromycin (ERY): root > leaf/shoot > fruit. The growth of crops could accelerate the dissipation of antibiotics by absorption from contaminated soil. A higher accumulation of antibiotics was observed in crop tissues under the wastewater treatment than under manure treatment, which was due to the continual irrigation that increased adsorption in soil and uptake by crops. The translocation of antibiotics in crops mainly depended on their physicochemical properties (e.g. log K_{ow}), crop species, and the concentrations of antibiotics applied to the soil. The levels of antibiotics ingested through the consumption of edible crops under the different treatments were much lower than the acceptable daily intake (ADI) levels.

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1. Introduction

Wastewater is commonly used to irrigate agricultural land in arid and semi-arid regions because of water shortages resulting from climate change, urbanization, regional drought and pollution (IPCC, 2009; WWAP, 2012). For example, in China, water availability is especially low and unevenly distributed; drought affects an average of 15.3 million hectares of farmland every year, which accounts for 13% of the total farming area (Shi et al., 2012). Wastewater therefore becomes a valuable resource for irrigating arable land. However, studies over the last two decades showed that many toxic inorganic pollutants, organic pollutants (e.g. antibiotics) and pathogens are present in untreated wastewater; these pollutants are generally biologically active and create potential risks when they enter the environment (Chen et al., 2011; Papadopoulos et al., 2009; Yan et al., 2013). Ten million hectares of soil in China were contaminated by organic pollutants, and 3.3 million hectares were contaminated as a result of wastewater irrigation (DAGDP, 2013). Animal manure is frequently used as fertilizer in arable land. However, animal manure may be a source of antibiotic contamination because veterinary antibiotics are widely used (Pan and Chu, 2017b). Most veterinary antibiotics are water soluble and cannot be completely absorbed by animals. Approximately 75–90% of veterinary antibiotics are excreted in animal manure per dose (Halling-Sørensen, 2001). Martínez-Carballo et al. (2007) reported that the concentration of tetracycline in 30 pig manure samples from Austria was up to 23 mg/kg, whereas the concentrations of norfloxacin and enrofloxacin were found to be as high as 225 and 1420 mg/kg, respectively, in chicken manure from China (Zhao et al., 2010). Thus, antibiotics can enter into the environment through the spreading of manure and slurry on agricultural land, direct deposition by grazing livestock, or discharge of untreated

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wastewater (Pan and Chu, 2017a; Wu et al., 2012). Hamscher et al. (2002) detected antibiotics at a maximum concentration of 307 μg/kg in soil. When untreated wastewater or animal manure is used for agricultural irrigation or fertilization, antibiotics have the potential to accumulate in soil and subsequently taken up by crops (Pan and Chu, 2017a). Previous studies also indicated that some pharmaceuticals and antibiotics have the potential to accumulate in different plants (Prosser and Sibley, 2015; Wu et al., 2014, 2015). Humans may be exposed to antibiotics through the consumption of crops grown using wastewater or manure.

Since large quantities of antibiotics are present in untreated wastewater and animal manure as a mixture, further research is clearly needed to better understand the transfer and uptake of antibiotics in crops and their risks to crops and humans. Most studies examined the concentrations of antibiotics in wastewater-irrigated soil or manure-applied soil separately (Li et al., 2015) or were restricted to only one type of antibiotic under artificially high levels (Kang et al., 2013; Watkinsona et al., 2009). Some studies were carried out in hydroponic environments, which did not reflect plant uptake or the accumulation of antibiotics in a realistic soil environment (Goldstein et al., 2014; Wu et al., 2013). Application in the form of wastewater and animal manure will entail different modes, rates and frequencies in application which, when combined with their intrinsic antibiotic and residual concentrations, will result in varying translocation and ecological risks of antibiotics in soil and crops. However, there are no studies comparing the effects of antibiotic-contaminated wastewater irrigation and animal manure application on antibiotic accumulation and translocation in different crops. Accordingly, there is limited understanding on the transfer and accumulation of antibiotics in soil and crops under wastewater irrigation or animal manure fertilization, and their corresponding human exposures. Therefore, different concentrations of antibiotics in wastewater and animal manure treatments were used for cultivating different crops in this study. The objectives were (1) to investigate the distribution of different types of antibiotics from wastewater and animal manure to soil and crops, (2) to comparatively evaluate the uptake and translocation of antibiotics in various crops, (3) to compare the antibiotic accumulation effects for wastewater irrigation and manure fertilization with two different antibiotic concentrations, and (4) to evaluate the ecological risks of antibiotics for different crops and human exposure. To our knowledge, this is the first study to contrast the effects of antibiotic-contaminated wastewater and animal manure on edible crops. Findings from this study would facilitate a better understanding of the transfer of antibiotics from water irrigation and fertilizer application to the soil-plant system and the potential ecological and human risks from exposure to antibiotic contamination.

2. Materials and methods

2.1. Antibiotics and chemicals

Target antibiotic compounds were selected based on their frequent use in the environment and their different physicochemical properties (Jiang et al., 2014; Sarmah et al., 2006; Zhou et al., 2011). Tetracycline (TC), sulfamethazine (SMZ), norfloxacin (NOR), erythromycin (ERY) and chloramphenicol (CAP), which belong to five different antibiotic classes, were examined (Table S1). All antibiotic standards and some internal standards (SMZ-d_4, NOR-d_5 and ERY-$^{13}C_2$) were obtained from Sigma-Aldrich (USA); CAP-d_5 was purchased from Dr. Ehrenstorfer GmbH (Germany); and TC-d^6 was obtained from Toronto Research Chemicals (Canada). Oasis HLB extraction cartridges (6 mL, 500 mg) (Waters Corporation, USA) were used for the extraction and purification of the target compounds. All the organic solvents used were HPLC grade and purchased from Merck Corporation (Germany). Individual stock solutions and internal standards were prepared at 100 mg/L in methanol and stored in an amber glass vial at −20 °C.

2.2. Experimental design

Lettuce (*Lactuca sativa*), carrot (*Daucus carota*) and tomato (*Solanum lycopersicum*) were used as target crops, as they have different edible parts and significant economic and ecological values, and are commonly used in salads and often consumed raw. Their seeds were purchased from a local seed supplier (Choi Hing Lee International Ltd, Hong Kong). The agricultural soil used was collected from the 0–20 cm depth surface layer in an organic farm (Produce Green, Hong Kong) and sieved through a 2 mm sieve after air drying. It was a clay loam with a pH of 6.38 and an organic carbon content of 0.80%. No antibiotics in any form were applied to the soil, and no target compounds were detected in the soil samples. Chicken manure was collected from an organic animal farm in Huizhou (Guangdong, China), accredited by the Department of Agriculture of Guangdong Province as animal-drug free; no target compounds were detected in the manure samples.

Wastewater irrigation and animal manure application were carried out in a greenhouse study. Each plastic pot (15 cm id × 15 cm height) contained 3.8 kg soil and one crop species, with all crops being grown from seed. For the wastewater treatment, each crop species was exposed to two levels of mixed antibiotics spiked into the irrigation wastewater, viz. 2 and 20 μg/L (WW2 and WW20 initial concentrations for each antibiotic). Two hundred milliliters of antibiotic-contaminated wastewater were applied to each pot every day (FAO, 2010). For the animal manure treatment, each crop species was exposed to two levels of mixed antibiotics spiked into the animal manure fertilization, viz. 200 and 2000 μg/kg (AM200 and AM2000 initial concentrations for each antibiotic). Twenty grams of antibiotic-contaminated manure were used to fertilize each pot half-monthly, and each pot was irrigated with deionized water every day. The following two controls were included: a negative control, without antibiotics in the irrigation water and fertilizer, was used to detect any antibiotic contamination emanating from other sources during the experiment, and a positive control, without crops, was used to assess the possible dissipation of antibiotics in the soil. The lower and higher spiked concentrations of antibiotics in wastewater and manure were within the range of commonly detected levels in surface water and animal manure (Hamscher et al., 2002; Yao et al., 2015). For example, TC, SMZ and CAP were detected in the range of 110-29,300, 230-2,000, and 140-14,300 μg/kg, respectively, in animal manure (Hu et al., 2010). Each treatment consisted of four replicates, and all crop tissues (root, leaf/shoot and fruit) were harvested when they reached marketable size (lettuce 60 d; carrot and tomato 120 d) for biomass and antibiotic determination, and the soil in the pot was collected for dissipation analysis. The drainage water after harvest was also collected for further analysis. All pots were placed in a greenhouse at 25 ± 2 °C and 60% relative humidity, and the plants were watered every day to maintain the water-holding capacity at 70%.

2.3. Extraction and analytical methods

The studied antibiotics were extracted and determined according to the procedures described in our previous study (Pan et al., 2014). Briefly, one gram of freeze-dried soil was ground up and spiked with 100 μL of each internal standard (1.0 mg/L). Thirty mL of extraction buffer (acetonitrile and 0.2 M citric acid, v:v = 1:1, pH 4.4) was added to each soil sample. The samples were

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