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Residues and risks of veterinary antibiotics in protected vegetable soils following application of different manures

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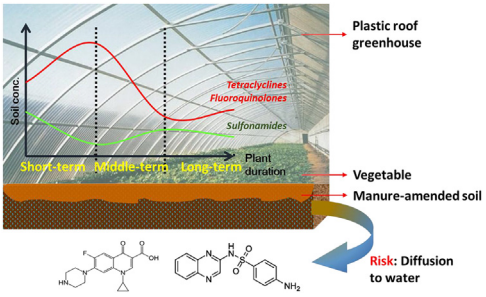
HIGHLIGHTS

- Intensive land manure application elevated antibiotics contamination in soil.
- Short-term planting affected tetracyclines and fluoroquinolones accumulation mostly.
- Manure sources impact levels and types of residual antibiotics in the farmland soils.
- Organic farming has less antibiotics residue than conventional greenhouse farming.
- Ciprofloxacin and sulfachinoxalin have higher migration risk than other antibiotics.

GRAPHICAL ABSTRACT



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ABSTRACT

The protected vegetable farming is a style of high frequent rotation farming which requires a huge amount of fertilizers to maintain soil fertility. A total of 125 surface soils covering from east to west of China were sampled for the analysis of 17 antibiotics in order to identify antibiotics contamination caused by long-term manures application. The results indicate that the agricultural land has accumulated a statistically significantly higher antibiotics concentration than conventional open croplands. The maximum oxytetracycline concentration was 8400 μg kg^{-1}, the highest level that has ever been reported for oxytetracycline in soils. The residual concentration is decided by both plant duration and manure type. Short-term (<5 years) planting shows the highest residues of tetracyclines and fluoroquinolones in the soils. The organic farming characteristic of applying commercial compost as a single fertilizer in planting shows the lowest antibiotics residue in the soils on the whole. Principal component analysis suggests that the various combinations of antibiotic compounds in the soil may be used to trace the manure source. The antibiotics in soil may threaten water quality through contamination by diffusion.

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Ciprofloxacin and sulfachinoxalin are calculated to be a higher migration risk to surface waters, hence their environmental fate requires further study.

1. Introduction

Manures are commonly applied to agricultural land in China to recycle their plant nutrients. The annual loading of manures is estimated to be up to 150 t ha^{-1} in protected vegetable farming with characteristically high frequency of vegetable rotation even in low temperature areas and this represents ten times the amount applied to crops in open fields (Qin et al., 2002). Therefore, the residue of veterinary antibiotics in soil might occur after long-term application of manure in farmland since most of the manure has been observed to be contaminated with antibiotics. We have found that all the 17 veterinary antibiotics analyzed were detected in 50 manure and compost samples from 8 Chinese provinces, and the oxytetracycline concentration was up to 417 mg kg^{-1} in a chicken manure sample (Zhang et al., 2015). Ubiquitous residues of fluoroquinolones, sulfonamides and tetracyclines in animal faeces have also been reported in several other studies (Zhao et al., 2010; Pan et al., 2011; Chen et al., 2012; Li et al., 2013b).

The veterinary antibiotics may enter soils through land-application of manures in the farmland and further release into water by runoff or leaching. The veterinary antibiotics contamination in farmland soil has been concerned of worldwide (Kumar et al., 2005; Hu et al., 2010; Li et al., 2011; Hou et al., 2015; Wei et al., 2016). However, the fate of antibiotics in soil varied with compounds. Sulfonamide antibiotics do not sorb strongly to soil and thus have been detected frequently in surface water, ground water, soil pore water (Wegst-Uhrich et al., 2014). While another antibiotics such as tetracyclines or fluoroquinolones, may persist for several months to years in soil (Jechalke et al., 2014). Meanwhile, antibiotics may accumulate in soil over time when the input rates exceed dissipation rates. A study on the sulfamethoxazole and ciprofloxacin contamination in Mexican soils demonstrated that these two chemicals could accumulate in the soils as a sequestered form over a period of 20 years during long-term irrigation with untreated wastewater (Dalkmann et al., 2012). The veterinary antibiotics may also transfer from soil to crops and posed a potential health hazardous to human being (Kumar et al., 2005; Hu et al., 2010). Soil contamination of antibiotics has also contributed to the spread of antibiotic resistant genes (ARGs) in the environment, which might result to an even more serious risk to human health (Martinez, 2008; Pruden et al., 2013; Zhu et al., 2013). A close relationship between the antibiotic use and ARGs abundance has been found for sulfonamide and tetracycline of the pig farms and cattle waste lagoons both in China (Zhu et al., 2013) and the United States (McKinney et al., 2010). An increase abundance of tetracycline resistance has been found in the arable soil with long-term application of fresh manure and compost, meanwhile, the dominant tetG genotypes shared strong homology with pathogenic bacteria (Peng et al., 2015).

Therefore, it was assumed that soil contamination by veterinary antibiotics in the protection vegetable farms became more and more seriously with the development of this intensive farming type. It has been estimated that the total cropping area of protected vegetables in China had reached 4.67 million ha by the end of 2010, double the area in 2004 (Yu, 2011). Hence, antibiotics contamination in such high frequency vegetable rotation farming systems has increasingly been of concern in recent years (Li et al., 2013a; Fang et al., 2014; Hou et al., 2015; Ur Rehman et al., 2015; Zhang et al., 2015; Wei et al., 2016), and many of these studies have focused on soil contamination by the antibiotics. In this study, soil samples with varied manure applications and vegetable planting durations were collected to investigate the veterinary antibiotics residue in the soils of intensive vegetable land, and to evaluate the potential environmental risk based on the current residual level. The data could provide a new insight on the antibiotics contamination in the Chinese intensive vegetable farming system and its relationship with the management of manure application.

2. Materials and methods

2.1. Soil sampling

A total of 125 soil surface (0–20 cm) samples were collected from the protected vegetable farm lands situated at 7 areas in Jiangsu province, Shanghai and Yunnan province, China in 2012 (Fig. s1). In addition, a total of other 39 surface soil samples (0–20 cm) were collected from the open farmland as a comparison in these areas, which included 13 samples from cereal crops lands and 26 samples from vegetable lands. The detail information of the sampling area could be found in the references (Wang et al., 2013; Yang et al., 2014). Briefly, the protected vegetable farmlands were selected based on the application of organic fertilizer, planting duration, management, and soil properties.

As shown in Table 1, the highest amount of manure application was in the protected vegetable farmland of Tongshan, Xuzhou (TS) in Jiangsu province. It has reached up to 150 t/ha yr^{-1} and the type of the applied organic fertilizer was dominated by livestock manure. The application amount in the other six sampling areas were all below 100 t/ha yr^{-1}, and the lowest amount was nearly 20 t/ha yr^{-1} in Pulangke (PLK) and Suoshi (SS), Nanjing. The planting duration also varied among the regions, spanning four to 30 years (Table 1). The vegetable farms in Tongshan (TS), Xuzhou have the longest duration of planting while the farms in Hushu (HS), Nanjing, have the shortest duration. Differences in field management correspond largely to the application of organic fertilizers or manures. Two types of management have been identified based on field investigations. One is conventional management such as TS and GL (Guli in Nanjing), HS, SS, and JN (Jinning in Kunming) and the other is an organic farming system such as PLK and QP (Qingpu in Shanghai). The former is typical of the combined use of manures and inorganic compound fertilizers during vegetable planting, and farmyard manures are the primary organic nutrient sources. The latter is characteristic of single using well manufactured organic compost (commercial compost) in planting (Zhang et al., 2015).

The soils are mainly comprised of two soil types based on FAO soil classification: Fluvic Cambisols and Stagnic Anthrosols (IUSS Working Group WRB, 2015). The soils of TS in Xuzhou, QP in Shanghai and JN in Kunming are all comprised of Fluvic Cambisols but developed from different parent materials. The TS soil and QP soil are developed from alluvial material of the Yellow River and the Yangtze River, respectively (Gong, 2003). The soil texture is characteristic of sandy loam to silt loam. While the JN soil is developed from local loamy alluvium. Soils of the four sampling areas in