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Influence of planting patterns on fluoroquinolone residues in the soil of an intensive vegetable cultivation area in northern China

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HIGHLIGHTS

- The vegetable planting model is the major determinant of FQ difference in soil.
- Planting age could not result in spatial pollution differences.
- Interactions between risk factors should be given more concern.

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ABSTRACT

Recent studies have demonstrated the persistence of antibiotics in soil, especially in areas of vegetable cultivation. However, there are very few studies of the influence of planting regimes on the levels of antibiotic pollution. This work introduces *geographical-detector models* to investigate the relationship between planting patterns (vegetable planting model, manure type and quantity, planting age, greenhouse area, and topographic elevation) and residual fluoroquinolones (*FQs*) in soil in a pilot project in Shouguang County, Shandong Province (the largest vegetable-producing area in China). The results led to the following findings. 1. The vegetable planting model is the major determinant of the spatial stratification of *FQ* in the soil. For example, the "cucumber–cucumber" model (growing cucumbers after cucumbers) has a three-fold power of determinant compared to the "pepper–melon" model (growing melons after peppers). 2. Planting age (years with continuous vegetable cultivation) does not necessarily affect the spatial distribution of *FQ* owing to their relatively short degradation period. 3. Interactions between risk factors were more significant than the individual factors for *FQ* pollution. In particular, the interaction between the vegetable planting model and amount of manure resulted in the highest pollution level. The findings of the present study make it possible to introduce effective and practical measures to alleviate pollution of soils by *FQ* in the study area. Adjustment of the vegetable cultivation models and application of chicken manure (less than 6 kg/m^2 manure annually with a more dry than fresh manure) could be an effective and flexible approach to alleviate *FQ* pollution.

1. Introduction

Antibiotics are probably the most successful family of drugs that has been developed to treat and prevent disease and to promote animal growth (Kemper, 2008). The significant increase in the number of large-scale animal feeding operations for swine, poultry, and cattle has resulted in the addition of considerable amounts of antibiotics to animal feeds (Phillips et al., 2004); about 30–90% of these antibiotics are released into the environment via manure (Bound and Voulvoulis, 2004; Leal et al., 2012; Xie et al., 2012). In organic vegetable production, the application of chemical fertilizers is limited across the entire vegetable cultivation area, and therefore, manure is applied to agricultural land as a nutrient source for plants. After entering the soil, antibiotics can be transported to surface water and ground water, and thereafter, it can be taken up by plants (Kuchta and Cessna, 2009). Although the health implications of antibiotic residues in plant-based products are largely unknown, it is anticipated that potentially adverse impacts include allergic/toxic reactions, chronic toxic effects due to prolonged low-level exposure, development and spread of antibiotic-resistant bacteria, disruption of digestive functions, and binding to human serum albumin (Chen et al.,

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2012; Ferber, 2003; Kumar et al., 2005; Martinez, 2008; Xiao et al., 2008; Zou et al., 2011). There is also evidence of ecotoxic antibiotic effects on plants (Aristilde et al., 2010; Jin et al., 2009; Wilson et al., 2003) and soil microorganisms (Fernandez et al., 2004). Thus, addressing the problem of antibiotic contamination of soil in vegetable cultivation areas has become an urgent issue (Siderer et al., 2005; Smukler et al., 2008; Xie et al., 2012).

In order to protect human health and to maintain vegetable quality and the sustainable development of the vegetable economy, it is necessary to take measures to alleviate antibiotic pollution in soil. However, previous studies of antibiotic removal have focused mainly on water bodies (Dong et al., 2009; Elmolla and Chaudhuri, 2010; Jeong et al., 2010; Liu et al., 2009; Rodriguez et al., 2012; Sun et al., 2009; Verlicchi et al., 2012). As of yet, there are no effective methods for the remediation of antibiotic pollution in soil. Excluding antibiotics from animal feed would be the most effective means of preventing antibiotic pollution, but such policies are unacceptable to the animal husbandry sector because of their significant negative influence on the profitability of livestock and poultry breeding. Moreover, in the Chinese context, the restriction of antibiotic residues in manure is not feasible, owing to the lack of relevant regulatory criteria in China.

Therefore, it is vital to identify the factors that determine antibiotic soil pollution and the potential solutions to this problem. Although previous studies have investigated antibiotic species and their residual concentrations in soil, few have investigated the determinants. In the present study, we investigate the relationship between planting patterns and soil pollution levels using a novel geographical detector model (Wang et al., 2010).

FQs (fluoroquinolones) are a group of widely prescribed antibiotics. They are used in both human and veterinary medicine owing to their broad-spectrum antibacterial characteristics. *FQs* were selected as the target antibiotics in the present study because of their long residence time in soil environments (>6 months following application) (Boxall et al., 2006).

2. Materials and methods

2.1. Reagents and sampling

Ciprofloxacin (CFX), enrofloxacin (EFX), and norfloxacin (NFX) were purchased from Sigma (St. Louis, MO, USA). Methanol and acetonitrile (HPLC grade) were obtained from Fisher (New Jersey, USA). Oxalic acid, phosphorous acid, disodium hydrogen phosphate (Na_2HPO_4), and ethylene diaminetetraacetic acid disodium salt (Na_2EDTA) were used as analytical reagents (the Beijing Reagent Company). Water was purified using a Milli-Q system (Millipore, Bedford, MA, USA). Stock solutions of the standards were prepared in methanol at a concentration of 100 mg/L and stored at −20 °C. Working standard solutions at a concentration of 10 mg/L were diluted from the stock solutions before use. Working mixed standard solutions were prepared by mixing the above solutions and diluting with methanol. These solutions can be stored under refrigeration (4 °C) for up to one week.

Shouguang County (Shandong Province) is the largest vegetable-producing area in China, with several hundred years of history of vegetable cultivation. The planting models in this area are intensive, regional, and consistent, thus providing a natural and representative experimental site for the present pilot study. The town of Daotian (approximate area 160 km^2) in Shouguang County was selected for its diverse and representative planting types. The study selected 100 vegetable greenhouses, separated by about 1 km (Fig. 1). Using a small shovel, soil samples were collected 0–15 cm below the soil surface during November 2010. The soil samples were all collected on soil ridges that were about 15–20 cm higher than the surface soil and contained almost no roots. This was done in order to avoid the effect of plant uptake in later analysis (it is difficult to calculate levels of antibiotics in the roots, leaves, fruits, and other biomass of different vegetables in field research). Samples were collected at five sites chosen within each greenhouse following an S-shape, and were combined and fully homogenized to form one sample. These samples were immediately transported to the laboratory under cooled conditions for analysis. The soil type in the target area is mainly cinnamon soil (Alfisol). The physical and chemical properties of one sample are described as follows: soil organic matter was about 32.7 g/kg; pH, 6.7; CEC, 17.4 cmol/kg; and the texture was approximately 45.2% for clay (<0.001 mm), 34.6% for silt (0.001–0.01 mm), and 20.2% for sand (>0.01 mm).

2.2. FQ analysis

The concentrations of *FQ* (CFX, EFX, and NFX) in soil were analyzed using high-performance liquid chromatography and fluorescence detection (HPLC–FD) using the method developed by Turiel et al. (2006). Lyophilized soil (1.0 g) was placed in a 10-mL centrifuge tube and the *FQs* were extracted using 5 mL of potassium phosphate buffer and acetonitrile (v/v = 1:1). The centrifuge tubes were vortexed for 10 min, sonicated for 15 min, and then centrifuged at 3000 ×*g* under air-cooled conditions. The extraction procedure was performed 3 times, and the resultant supernatants were combined.

Oasis HLB (200 mg, 6 mL) and SAX cartridge (200 mg, 3 mL) were obtained from Waters Corporation (Milford, MA, USA) and Bonna-Agela Technologies (Tianjin, China). Each cartridge was sequentially preconditioned with 6.0 mL methanol and 6.0 mL Milli-Q water. The mixture extraction solution with organic reagent under 5% was loaded onto the cartridge. The cartridge was then rinsed with 6 mL of Milli-Q and lyophilized for 40 min. Elution of the antibiotic compounds from the cartridges was done with 6 mL of methanol. Methanol eluate was evaporated to near-dryness under a gentle stream of nitrogen, followed by redissolution with 1.0 mL of methanol.

The mobile phase consisted of acetonitrile and 0.01 mol/L oxalic acid (80:20 v/v) at 1 mL/min flow rate.

2.3. Quality assurance and quality control

Procedural blanks consisting of ultrapure water were analyzed as a control of procedural contamination. After each sample containing potentially high levels of the target contaminants, solvent was injected to prevent potential cross-contamination. A mid-range calibration standard solution (100 μg/L) was run at the beginning, the middle, and at the end of each sequence to monitor instrumental sensitivity and reproducibility.

Recovery tests were performed by spiking a mixture of standards in soil using two different concentrations of 50 and 200 μg/L. The recoveries of CFX, EFX, and NFX were all greater than 65% in soil samples. The limit of detection (LOD) based on a signal-to-noise ratio greater than 3 was less than 1.0 μg/L in all cases. The limit of quantification (LOQ) based on a signal-to-noise ratio greater than 10 was less than 3.5 μg/L in all cases (Table S1).

External calibration curves were constructed by preparing standard solutions at six known concentrations (0.5–500 μg/L). The standard addition method was used to ensure identification of the target antibiotics in soil samples (Li et al., 2011).

2.4. Factors related to planting patterns

Questionnaires were completed during the process of soil sample collection; they included both quantitative and nominal data. Candidate risk factors were investigated (planting model, manure type and quantity, vegetable cultivation category, greenhouse area, and topographic elevation). The quantity of manure applied per square meter was calculated by dividing the total annual manure application by the greenhouse area (Fig. 2). Topographic elevation in the region (obtained using the digital elevation model) ranged from 8 to 47 m, increasing gradually from north to south.

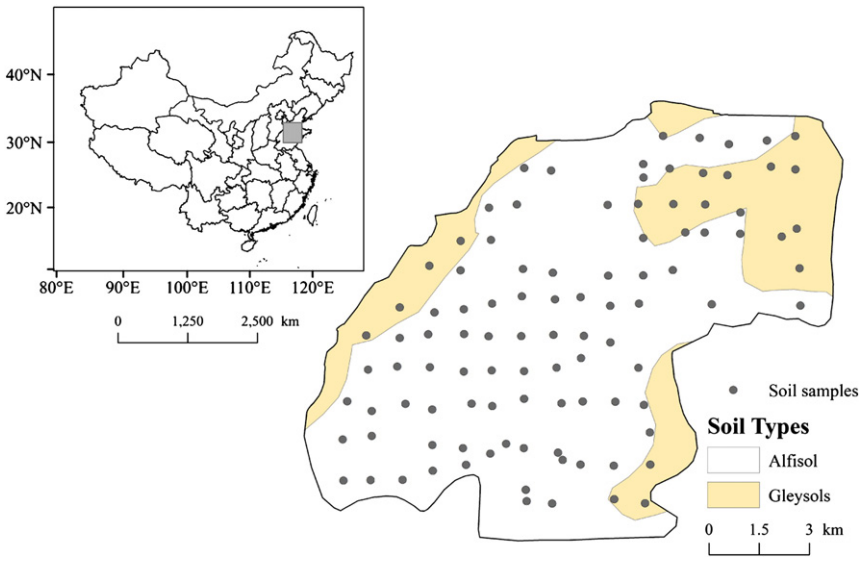

Fig. 1. Distribution of vegetable greenhouses and soil types in the target region.

The area of each greenhouse was within the range of 312–2335 m^2 (most of them were less than 1000 m^2 each). Planting ages ranged from 1 to 20 years, exhibiting no uniform spatial variation over the entire region. Manures (pig, chicken and cow dung) were the only source of veterinary antibiotic pollution. Chicken dung was the main manure source (>95%; Fig. S2), and the application quantity ranged from 1.3 to 17.1 kg/m^2 annually. As shown in Fig. 2d, most greenhouses received applications of manure in the ranges of 3–6 kg/m^2 and 6–9 kg/m^2.

Nominal factors included chicken dung categories and vegetable planting categories. Chicken dung was predominantly fresh (untreated before application), but also included dry dung (with the fresh dung fermented) and mixed dung (fresh dung and dry dung) with a dominant fraction of fresh dung (Fig. 2e).

According to the questionnaire results, two or three stubble vegetables were cultivated each year. Five main regions were examined corresponding to different vegetable growing models for one cultivation period. These regions were labeled by the vegetable names, as follows: "tomato–tomato," "pepper–melon," "pepper–eggplant," "cucumber–cucumber," and "melon–leaf vegetables" (Fig. 2f) (the most common model for a region was assigned to all greenhouses in that region). Of these, the "tomato–tomato" model occupied the largest area. Leaf vegetables, including lettuce, rape, spinach, and *Chrysanthemum coronarium* (edible) were mainly located in the southeastern part of the study region.

2.5. Statistical analysis

There are many risk assessment methods, such as the multivariable logistic regression model and univariate/multivariate analysis (Ellidokuz et al., 2005; Hill et al., 2008; Wu et al., 2010). However,

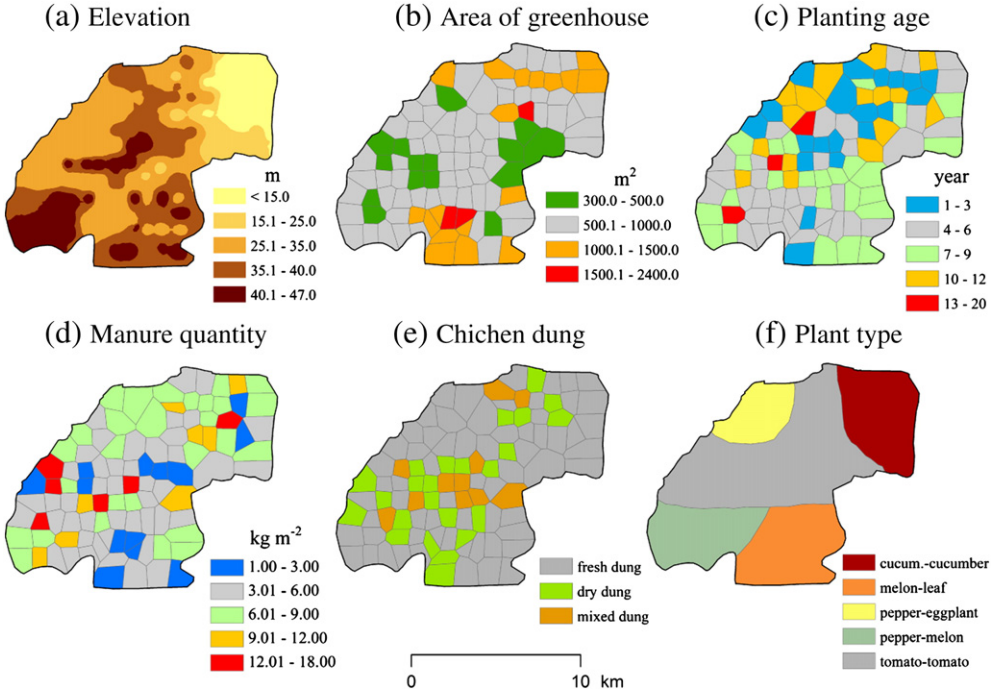

Fig. 2. Spatial distribution of suspect factors.

these methods usually assume homoscedasticity of the stochastic field and numerical data (Christakos, 2010), and are less suitable for interpreting interactions between factors. For example, interactions in the regression model are limited to two-variable multiplication, and these variables have to be numerical. The above limitations do not exist in the case of geographical detectors (Wang and Hu, 2012; Wang et al., 2010). The basic idea of geographical detectors is to measure the correspondence of the spatial distribution of response variables (e.g., residual *FQ*) to that of suspected determinants (e.g., planting age, vegetable types). Geographical detectors can handle both quantitative information and nominal data, and thus offer a novel approach for detecting interactions between the risk factors for antibiotic pollution.

FQ residues will exhibit a spatial variation in correlation with their risk factors, if the risk factors accurately determine the levels of pollution (Wang et al., 2010). Let the total *FQ*s in the region Ω be measured at grid points $h_1, h_2, \ldots, h_n$ (Fig. 3). Assume that *C* and *D* are two determining factors, or determinants, of *FQ* residues. Measurements of *C* and *D* can be continuous or categorical variables. Region Ω is stratified into sub-regions defined in terms of *C* and *D*, which are denoted in Fig. 3 as (c_1, c_2, c_3) and (d_1, d_2, d_3), respectively, under the principle of minimizing the dispersion variance of the explanatory variable within the sub-regions and maximizing the dispersion variance of the explanatory variable between the sub-regions.

In GIS, the *FQ* residue layer *H* is overlain with one determinant layer, say *D*. The average *FQ* residues and residue dispersion variances in each sub-region and in the entire region Ω are denoted by $\overline{y}_{d1}, \overline{y}_{d2}, \overline{y}_{d3}$, and $\overline{y}_D$, and VAR_{d1}, VAR_{d2}, VAR_{d3}, and VAR_D, respectively. If the *FQ* residue distribution is completely controlled by *D*, the concentrations of *FQ* residues should be constant in each of the subregions (d_1, d_2, d_3), meaning that $\mathrm{VAR}_{di} = 0$ $(i = 1, 2, 3)$. However, if the *FQ* residues are completely independent of *D*, then $\Sigma_i \Re_{di} \mathrm{VAR}_{di} = \mathrm{VAR}_D$. The above "*FQ* residue–risk factor" relationship is measured by means of the following Power of Determinant (*PD*):

$$PD = 1 - \frac{(\Re_{d1} VAR_{d1} + \Re_{d2} VAR_{d2} + \Re_{d3} VAR_{d3})}{\Re \times VAR_D}, \quad (1)$$

where $\Re$ and $\Re_{di}$ $(i = 1, 2, 3)$ denote the areas of region Ω and subregion d_i, respectively. Therefore, if the risk factor *D* completely controls the total *FQ*, then $PD = 1$. If, however, the factor *D* is totally unrelated to the ΣFQ, then $PD = 0$. Usually, the *PD* value lies somewhere in the interval [0, 1]. The impact of *D* on the *FQ* residue is directly proportional to the value of *PD*. Here, the geographical detectors used are risk detector, factor detector, ecological detector, and interaction detector (for a detailed discussion, see Appendix A). The relevant calculations were made using the GeogDetector software (Wang and Hu, 2012) (www.sssampling.org/geogdetector).

3. Results

3.1. Total fluoroquinolone residues in surface soil

The *FQ* residues were widely detected in farmland soil, and the distribution of different *FQ* species showed spatial variation. For example, ofloxacin, CFX, and pefloxacin have been detected in the soil of a vegetable cultivation area in northern China (Hu et al., 2010). NFX, CFX, EFX, and lomefloxacin were the main *FQ* species detected in the soil of the Pearl River Delta in southern China (Li et al., 2011). In the present study, NFX, CFX, and EFX were all detected in the greenhouse soils. This variation may be due to the different antibiotics used in animal breeding (Zhao et al., 2010). The vegetable planting areas studied here had concentrations of CFX, EFX, and NFX residues within the ranges 2.4–651.6 μg/kg, 0.1–166.9 μg/kg, and 0.4–288.3 μg/kg, respectively (Table S2). CFX was present at higher concentrations than EFX, although the latter is usually present in chicken- and turkey-based samples (Gans et al., 2007). This could be due to the metabolic conversion of EFX into CFX that occurs in many species (Lewis et al., 2011).

The total *FQ* (represented as ΣFQ) was calculated by adding the concentrations of NFX, CFX, and EFX. According to the technical guidance specified in the European registration procedure for human and veterinary medicines, no risk assessment is required for substances with an exposure level below a certain trigger value for soil (100 μg/kg) (Montforts, 2005). The minimum ΣFQ value with at least one substance that exceeded

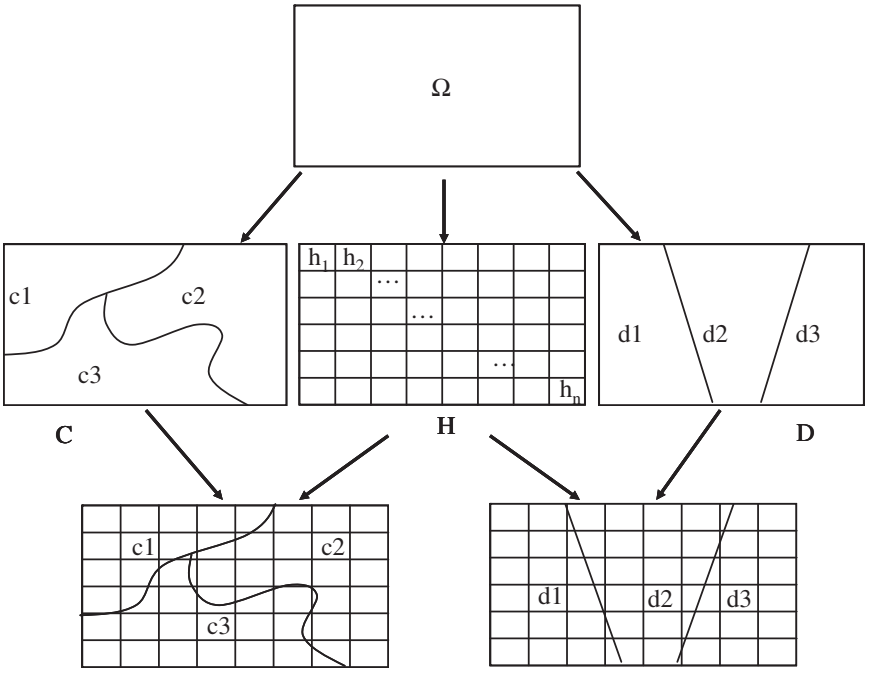

Fig. 3. FQ residues layer *H* and spatial patterns of suspect factors *C* and *D* in the study region Ω.

the trigger value was 165 μg/kg; i.e., Σ*FQ* exceeding 165 μg/kg indicates *FQ* pollution in the soil.

3.2. Influence of factors

The results for the four aforementioned geographical detectors are listed below. The *factor detector* determines the effect of risk factors on the Σ*FQ* pollution, ranked (according to *PD* value) as follows:

Vegetable planting models (0.28) > chicken dung quantity (0.20) > elevation (0.18) > planting age (0.09) > chicken dung categories (0.060) > area of greenhouse (0.02).

The first two factors (with $PD > 0.20$) are considered as the major risk factors.

The *ecological detector* (Table 1) showed that the variations in *PD* values between vegetable planting models, quantity of chicken dung, and elevation were not statistically significant; the differences between the remaining factors were not statistically significant; however, the differences between any one of the first three factors and any one of the remaining factors were statistically significant. According to the factor and ecological detectors, the vegetable planting models, chicken dung quantity, and topographic elevation strongly affect the total pollution (Σ*FQ*), whereas the remaining factors had only weak effects.

The *risk detector* indicated significant differences between all of the five vegetable planting models (Table 2). The order of the corresponding mean values is as follows:

"cucumber–cucumber" (318.5 μg/kg) > "pepper–eggplant" (246.7 μg/kg) > "melon–leaf vegetables" (171.2 μg/kg) ≈ "tomato–tomato" (162.7 μg/kg) > "pepper–melon" (96.2 μg/kg).

The results suggest that the "cucumber–cucumber" model significantly affects *FQ* pollution, with a magnitude approximately three times greater than that of the "pepper–melon" model. There is no significant difference between the "melon–leaf vegetables" and "tomato–tomato" models.

The quantity of chicken dung applied strongly affects *FQ* pollution, but higher *FQ* concentrations are not always associated with increased quantities of dung, as shown by the geographical detector (Table 3). The following order was found for increasing quantities of dung applied:

Level 1 (106 μg/kg) < Level 2 (147 μg/kg) < Level 5 (196 μg/kg) < Level 4(259 μg/kg) < Level 3 (302 μg/kg).

Σ*FQ* first increases and then decreases after reaching its inflection point at 9 kg/m^2; for levels 4 and 5, which had larger quantities of manure, exhibited *FQ* residues less than level 3 (discussed later). The pollution risk in the study area exceeded 165 μg/kg when manure application exceeded 6 kg/m^2.

FQ pollution was also affected strongly by topographic elevation: there was more pollution in low-elevation regions, and less at higher elevations (Table S3). This phenomenon should be investigated further.

Table 1
Differences of total (Σ*FQs*) pollution between factors.

Difference	Models	Dung quantity	Elevation	Dung types	Age	Area
Models						
Dung quantity	N					
Elevation	N	N				
Dung types	Y	N	N			
Age	Y	Y	N	N		
Area	Y	Y	Y	N	N	

Models: vegetable planting categories in one period; dung quantity: quantity of chicken dung per m^2; dung types: types of chicken dung; age: the time of greenhouse planting; area: the area of greenhouses; Y denotes that the difference of influence between the two factors is significant with a 95% CI; and N denotes that it is not.

Table 2
Differences of total (Σ*FQs*) pollution between five vegetable planting models.

Difference	Model 4	Model 2	Model 5	Model 3	Model 1
Model 4					
Model 2	Y				
Model 5	Y	N			
Model 3	Y	Y	Y		
Model 1	Y	Y	Y	Y	

Model 1: "cucumber–cucumber"; Model 2: "tomato–tomato"; Model 3: "pepper–eggplant"; Model 4: "pepper–melon"; Model 5: "melon–leaf vegetables". Y denotes that the difference of influence between the two factors is significant with a 95% CI; and N denotes that it is not. The order is model 4 < model 2 ≈ model 5 < model 3 < model 1.

The *interaction detector* examines the combined impact of two or more risk factors (Table 4). First, the independence of the variables was tested and the results showed that *elevation*, *vegetable planting model*, *dung categories*, and *planting age* could be used in the interaction analysis. The joint impact of vegetable planting model and planting age was greater ($PD = 0.34$) than that of the vegetable planting models alone ($PD = 0.28$). In contrast, the interaction of the vegetable planting model and chicken dung category ($PD = 0.25$) reduced the effect of the vegetable planting model.

4. Discussion

The four detectors discussed above revealed some interesting and unexpected mechanisms of *FQ* pollution. Although manure was the sole source of antibiotic residues in soil in this study area, chicken dung categories did not cause a large spatial variation in *FQ* pollution. According to the questionnaire results, if dry dung was only selected to apply, much more quantity of it than fresh dung was used for its lower fertility. In addition, the fresh dung showed higher levels of antibiotic residues than dry dung (which was fermented) (Dolliver et al., 2008). Consequently, the dung category did not result in spatial variations in *FQ* pollution; therefore, promoting the application of dry dung alone would not significantly reduce pollution.

Another unexpected finding was that the Σ*FQ* concentration did not always increase with dung quantity. This was demonstrated by the questionnaire results (Table S4): there was a higher percentage of fresh chicken dung in level 3 (6.0–9.0 kg/m^2 range) and this gradually decreased in both level 4 (9–10 kg/m^2) and level 5 (>10 kg/m^2). As a result, level 3 contains the highest concentration of *FQ* residues (fresh dung contains more antibiotics than dry dung). This indicates that, below the inflection point of 9 kg/m^2, dung quantity plays a more important role than the dung category; and when application quantity exceeds the inflection point, the ratio of dung categories (fresh/dry dung) is a decisive factor (higher ratios indicate higher risks of *FQ* pollution). This observation illustrates that merely promoting the application of dry dung would be insufficient to significantly reduce *FQ* pollution in practice. Therefore, levels 1 and 2 (i.e., <6 kg/m^2 manure associated with residues levels below 165 μg/kg) would be acceptable in field applications.

Table 3
Differences of total (Σ*FQs*) pollution between five levels of chicken dung quantity.

Difference	Level 1	Level 2	Level 5	Level 4	Level 3
Level 1					
Level 2	Y				
Level 5	Y	Y			
Level 4	Y	Y	Y		
Level 3	Y	Y	Y	N	

Level 1: 1.3–4.0 (kg/m^2); level 2: 4.0–6.0 (kg/m^2); level 3: 6.0–9.0 (kg/m^2); level 4: 9.0–10.0 (kg/m^2); level 5: >10.0 (kg/m^2). Y denotes that the difference of influence between the two factors is significant with a 95% CI; and N denotes that it is not.

Table 4
Interactions (measured by *PD* value) between pairs of factors on total (Σ*FQs*) pollution.

Interaction	Elevation	Model	Dung type	Age
Elevation				
Models	0.32			
Dung types	0.19	0.25		
Age	0.28	0.34	0.16	

Models: vegetable planting categories in one period; age: the time of greenhouse planting.

Another interesting finding is that, although the cumulative Σ*FQ* input via manure application would increase over time, *FQ* residues did not exhibit a systematic spatial variation with planting age. This may be related to their physicochemical properties, e.g., their short half-life (50% degradation time, DT50). The DT50 of most antibiotics is less than six months (Boxall et al., 2006); therefore, planting age does not define *FQ* pollution in the region. This finding has useful practical implications, as it suggests that there is no risk of *FQ* pollution worsening with cultivated age. Furthermore, from a research perspective, it is not possible to estimate *FQ* pollution in soil solely based on planting age. It should be noted that the conclusions here refer only to the original *FQ* substances, and that their degradation products have not yet been taken into account.

The vegetable planting model was found be the most important determinant of spatial variation in *FQ* pollution. Both the "pepper–melon" and "tomato–tomato" models result in concentrations of less than 165 μg/kg, whereas both the "pepper–eggplant" and "cucumber–cucumber" models showed higher risks of soil pollution. Greater attention should be paid to the "cucumber–cucumber" planting region, because the findings suggest that fresh chicken dung (>6 kg/m^2) can lead to more serious *FQ* pollution than in other regions.

The present study suffers from similar limitations to other studies, in that intermediate processes, specifically antibiotic migration, are not quantifiable in field research and are therefore neglected (Hu et al., 2010).

Compared to the effects of individual factors, interactions between factors play a more important role in *FQ* pollution. It was found that the combined effect of the vegetable cultivation model and planting age is greater than their individual effects. In contrast, the combined effect of the vegetable cultivation model and dung categories reduced the effect of the vegetable cultivation model.

The findings of this study facilitate the introduction of effective and feasible measures to alleviate the *FQ* pollution of soil, and these can be summarized as follows.

First, under the current circumstances, dung application should be less than 6 kg/m^2, and a greater proportion of dry dung than fresh dung should be used. Second, changing the vegetable planting model is a valid approach to reducing pollution. For instance, the use of the "pepper–melon" model should be promoted in the north and east regions, which already show serious *FQ* pollution. Third, interactions between risk factors provide important information. Simultaneously adjusting the vegetable cultivation model and chicken dung categories could reduce the effect of changing the vegetable cultivation model alone.

5. Conclusions

The "cucumber–cucumber" cultivation region has the highest *FQ* pollution risk within the study area. The vegetable planting models constitute a major factor in the spatial variation of *FQ* pollution, whereas advanced planting age was not found to cause serious *FQ* pollution. In order to minimize the potential disruption to the expansion of the local vegetable economy, an effective and flexible approach to alleviate *FQ* pollution is provided: adjustment of the vegetable cultivation model and application of chicken manure (less than 6 kg/m^2 manure annually with more dry than fresh manure).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2013.04.002.

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