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RESEARCH ARTICLE

Effects of straw addition on increased greenhouse vegetable yield and reduced antibiotic residue in fluvo-aquic soil

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Abstract

Organic manure application is an important measure for high yield and good quality vegetable production, whereas organic manure is also a main source of residual antibiotic in soils. A 3-yr experiment was conducted on a fluvo-aguic soil in Tianjin of northern China. The objective of this study was to investigate the effects of different fertilization patterns on yield of six-season vegetables with celery and tomato rotation, and dynamic change of tetracyclines residues in the soil during the sixth growing season (tomato season). The field experiment comprised six treatments depending on the proportion of nitrogen of each type of fertilizer: 4/4 CN (CN, nitrogen in chemical fertilizer), 3/4 CN+1/4 MN (MN, nitrogen in pig manure), 2/4 CN+2/4 MN, 1/4 CN+3/4 MN, 2/4 CN+1/4 MN+1/4 SN (SN, nitrogen in corn straw), and CF (conventional fertilization, the amounts of nitrogen application were 943 and 912 kg N ha^{-1} for celery and tomato season, respectively). In addition to CF treatment, the amount of nitrogen application in other treatments was greatly reduced and equal (450 and 450 kg N ha^{-1} for celery and tomato season, respectively). Results showed that the combined application of 3/4 CN+1/4 MN achieved the highest yield and economic benefit in the first four seasons, but addition of straw (2/4 CN+1/4 MN+1/4 SN treatment) performed better in the subsequent two seasons, and the average yields of 2/4 CN+1/4 MN+1/4 SN treatment were respectively higher by 9.9 and 12.8% than those of 4/4 CN treatment, and by 5.6 and 10.5% than those of CF treatment. The residual chlortetracycline (CTC) in manure-amended soil for three consecutive years increased along with the increase of applied amount of pig manure. Under the same amount of pig manure application, content of CTC in straw-amended soil was obviously decreased compared with no straw-amended soil (3/4 CN+1/4 MN treatment), and averagely decreased by 41.9% for four sampling periods in the sixth season. Addition of crop straw facilitated the degradation of CTC in manure-amended soil. As a whole, the conventional fertilization was not the desirable pattern based on yield, economic benefit and environment, the optimal fertilization pattern with the highest yield and profit and the least soil chlortetracycline residue was the treatment of

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2/4 CN+1/4 MN+1/4 SN under this experimental condition.

Keywords: greenhouse vegetable, organic manure, straw, yield, soil chlortetracycline

1. Introduction

Organic manure is necessary for high quality and high yield of vegetables and nutrient cycling, and most of which is originated from animal manure in agriculture. In China, the current production of animal manure reaches 4.5 billion tons (Li *et al.* 2009a), two-thirds of which is applied to the vegetable field accounting for about one-eighth of total arable land (China Agricultural Yearbook Editing Committee 2013). A large amount of manures, such as chicken manure, pig manure and the like have been used in vegetable production with high annual application amount for some farmers ranging from 153.9 to 240.0 t ha^{-1} (Huang *et al.* 2010). As regarding to animal dung originated from intensive livestock farming, 30–90% of antibiotics used during animal breeding period cannot be fully utilized, thus leaving in the dung (Campagnolo *et al.* 2002; Sarmah *et al.* 2006; Arikan *et al.* 2009b). Currently, more than 80% of animal dung employed is directly applied to farm soil without being subjected to harmless treatment (Li *et al.* 2008). Although composting allows for 30–95% reduction of antibiotic residue in manure (Dolliver *et al.* 2008; Mohring *et al.* 2009), antibiotic accumulation in soil occurs due to excessive and continuous input of manure as organic fertilizer and long half-life of antibiotic in soil (Wang and Yates 2008; Hamscher 2009; Selvam *et al.* 2012). Some researches have shown that antibiotic pollution in soil is caused by manure application, the residual antibiotic content commonly ranging from several μg per kg to several hundred μg per kg, and this pollution is mainly focused on three kinds of tetracyclines including tetracycline (TC), oxytetracycline (OTC), and chlortetracycline (CTC) (Hamscher *et al.* 2002; Martinez-Carballo *et al.* 2007; Ramaswamy *et al.* 2010), which may destroy farm product safety and further cause a risk of poisoning animal and human body by means of food chain (Kumar *et al.* 2005; Alexander *et al.* 2009; Hu *et al.* 2010; Xie *et al.* 2012).

Monitoring antibiotic residues in manure and manure-based composts prior to application to agricultural fields is benefit for mitigating environmental pollution by antibiotics (Kwon *et al.* 2011). Harmless treatment related to the reduced antibiotic residues in manure is reported in some cases. Organic materials such as sawdust, husk and oil cake added during the manufacture of manure-based composts would significantly decrease antibiotic residue (Kim *et al.* 2012). He *et al.* (2011) disclosed that rice straw had clear promotion on degradation of tetracyclines in pig manure, and the degradation rate of TC, OTC, and CTC in pig manure compost after adding straw increases by 26, 20 and 34%, respectively. The research on effects of high temperature composting upon the degradation of antibiotic in manure by Zhang *et al.* (2006) shows that wheat straw significantly accelerated the degradation of tetracyclines in pig and chicken manures. By comparison of influences of different composting measures (simple composting of cow dung, mixed composting of cow dung and straw, and mixed composting of cow dung and straw in combination with thermal insulating layer of straw) on the degradation of OTC and CTC, it is found that the degradation rates of the two antibiotics were both higher in the last compositing pattern than those in other composting patterns after 28 days composting (Arikan *et al.* 2009a). In recent years, massive animal manures have been applied in greenhouse vegetable in China (Tai *et al.* 2011; Yin *et al.* 2012). High input and low harmless treatment rate of organic manures result in an increasing accumulation of antibiotics in greenhouse soil. Some researches mentioned above indicated that crop straw could distinctly stimulate degradation of tetracyclines in manure-based composts. It is speculated that annual output of crop straw may reach more than 700 million tons in China, with only 10% of crop straw being returned to field as fertilizer (Dai 2009). However, current studies of the effects of straw or of combined application of different carbon sources including pig manure and straw and chemical fertilizers on the degradation of tetracyclines in soil were seldom reported.

Currently, long-term excessive application of manure with high antibiotic residue has been as a crucial unsafe factor for vegetable production under conventional fertilizer application. Most of researches were simply limited to the feasibility of reduced chemical fertilizer application in terms of yield and economic benefit (Tang *et al.* 2010). However, viewed from high yield production and low antibiotic residue, seeking optimal combined application of organic manure and chemical fertilizers is an urgent issue to be studied. Therefore, a long-term experiment was conducted to investigate the effects of combined application of pig manure, straw, and chemical fertilizers on yield of vegetable, and tetracyclines residues in soil during growth period, and further to provide a sustainable fertilizer application pattern with high-quality vegetable production and low antibiotic residue in soil. This may be used as a scientific basis of high-efficiency and safe application of animal manure as an organic manure.

2. Results

2.1. Yields and economic benefits of tomato and celery

Yields and economic benefits of celery and tomato in different seasons are shown in Tables 1 and 2. The yields and economic benefits of 3/4 CN+1/4 MN treatment (3/4 N from chemical fertilizer, 1/4 N from pig manure) from the first to fourth season were relatively higher. The average yields of the treatment in the first, second, third and fourth seasons were 121.8, 100.6, 151.4 and 114.8 t ha^{-1}, respectively, which were higher by 1.8, 5.4, 5.1 and 6.8% than those of 4/4 CN treatment (complete N from chemical fertilizer), and 1.7, 6.6, 4.4 and 7.7% than those of CF treatment (conventional fertilization). Correspondingly, the average economic benefits of 3/4 CN+1/4 MN treatment in the first to fourth season were in turn 328, 218, 197 and 283 thousand CNY ha^{-1}, which were higher by 4, 10, 7 and 16 thousand CNY ha^{-1} than those of 4/4 CN treatment, and 27, 27, 24 and 36 thousand CNY ha^{-1} than those of CF treatment.

Starting from the fifth season (autumn-season celery), the fertilization pattern of 2/4 CN+1/4 MN+1/4 SN treatment (2/4 N from chemical fertilizer, 1/4 N from pig manure and 1/4 N from straw) achieved relatively higher yield and economic benefit. For the fifth-season and sixth-season vegetables, the average yields of 2/4 CN+1/4 MN+1/4 SN treatment were respectively 136.1 and 103.5 t ha^{-1}, which were higher by 9.9 and 12.8% than those of 4/4 CN treatment, and 5.6 and 10.5% than those of CF treatment. In addition, the average economic benefits of 2/4 CN+1/4 MN+1/4 SN treatment were respectively 340 and 229 thousand CNY ha^{-1}, which were higher by 29 and 21 thousand CNY ha^{-1} than those of 4/4 CN treatment, and 35 and 31 thousand CNY ha^{-1} than those of CF treatment.

2.2. Contents of soil organic matter, NO_3^--N and NH_4^+-N in soil

After the fourth-season and sixth-season harvests, effects of different fertilization patterns on the contents of soil organic matter (SOM) and NO_3^--N were significant (P<0.05), but such effects on the content of NH_4^+-N was not the case (Table 3). Compared to the other four treatments with equal amount of N application, the content of SOM treated with 2/4 CN+1/4 MN+1/4 SN was superior with the increasing ranges of 13.8–32.5 and 12.1–42.3% after the fourth-season and sixth-season harvests, respectively, which was respectively increased by 6.5 and 5.1% compared with those of CF treatment, but the difference was not significant. Also, the content of NO_3^--N in 2/4 CN+1/4 MN+1/4 SN treatment was superior in the four treatments having equal N amount with the increasing ranges of 28.9–55.3 and 28.3–58.8% after the fourth and sixth harvests, respectively, which was respectively increased by 14.9 and 8.5% compared with those of CF treatment.

Table 1 Effect of different N application patterns on the yields of tomato and celery (t ha^{-1}, FW)

Treatments[1)]	Celery season 2009	Tomato season 2010	Celery season 2010	Tomato season 2011	Celery season 2011	Tomato season 2012
4/4 CN	119.6±2.5 a	95.4±2.4 bc	144.0±0.6 b	107.4±1.3 bc	124.1±0.9 cd	91.8±3.4 c
3/4 CN+1/4 MN	121.8±1.4 a	100.6±1.4 a	151.4±2.3 a	114.8±1.1 a	133.1±1.6 ab	100.7±0.9 ab
2/4 CN+2/4 MN	119.7±0.5 a	98.4±1.2 ab	145.0±0.8 b	109.0±1.4 bc	125.5±6.6 cd	97.3±0.9 b
1/4 CN+3/4 MN	119.8±1.8 a	95.8±1.9 bc	146.5±1.0 b	106.9±1.3 c	122.6±3.8 d	99.6±0.7 b
2/4 CN+1/4 MN+1/4 SN	119.6±2.5 a	92.3±2.1 c	144.5±0.9 b	109.8±1.0 b	136.3±2.0 a	103.5±2.7 a
CF	119.7±1.9 a	94.4±2.3 bc	145.0±2.8 b	106.5±1.7 c	129.2±2.3 bc	93.7±1.2 c

[1)] CN, nitrogen in chemical fertilizer; MN, nitrogen in pig manure; SN, nitrogen in straw; CF, conventional fertilization.
Values followed by different letters of each growth season within a column are significant at the 5% probability level.
The same as below.

Table 2 Effect of different N application patterns on the income of tomato and celery (thousand CNY ha^{-1})

Treatments	Celery season 2009	Tomato season 2010	Celery season 2010	Tomato season 2011	Celery season 2011	Tomato season 2012
4/4 CN	324±7 ab	208±6 b	190±1 b	267±3 b	311±2 b	208±8 d
3/4 CN+1/4 MN	328±4 a	218±3 a	197±3 a	283±3 a	332±4 a	226±2 a
2/4 CN+2/4 MN	321±2 ab	210±3 ab	186±1 c	265±4 b	309±17 b	216±2 cd
1/4 CN+3/4 MN	319±5 b	202±4 bc	185±1 c	257±3 c	299±10 b	219±2 bc
2/4 CN+1/4 MN+1/4 SN	322±7 ab	198±5 cd	186±1 bc	268±2 b	340±5 a	229±10 a
CF	301±5 c	191±5 d	173±4 d	247±4 d	305±6 b	198±3 e

In this study, under the identical input of nitrogen in all treatments except CF treatment, the total input amount of carbon in straw treatment was significantly higher than that in other treatments (Tables 4, 5). The content of SOM in treatment of straw addition increased markedly compared with other treatments, the reason of which mainly resulted from high contents of cellulose and lignin in straw and high C/N ratio, thereby promoting soil humus to keep high vitality after being applied into soil and conducive to microbial decomposition and nutrient release. Newly formed organic matter was mainly present in the form of easily-oxidized organic carbon, which increased the activation degree of soil organic matter and the utilization efficiency of soil nitrogen at a certain degree (Dai 2009). Therefore, in the medium and long term, the fertilization pattern of 2/4 CN+1/4 MN+1/4 SN treatment harmonized the balance between soil nutrients and energy in greenhouse soil, thus improving soil quality and further achieving high-efficient and sustainable production of greenhouse vegetable.

2.3. Changing characteristics of chlortetracycline residue during the sixth growing season

In four sampling periods of the sixth season, there were no TC and OTC detected in all treatments, but CTC with different contents in all manure-amended treatments (Table 6).

Before basal fertilizer application (BFA) in sixth season, there was no CTC detected in entire chemical fertilizer treatment (4/4 CN), in contrast, combined application of pig manure and chemical fertilizer significantly increased the content of CTC in soil; and furthermore, the content of residual CTC in soil increased along with the increase of organic manure application, which resulted from different accumulation of antibiotic due to different fertilizer application patterns during the former five seasons. The CTC contents in all manure-amended soils for three consecutive years were still high at 40th day after BFA in the sixth season, and thereafter reduced greatly to a relatively low level at 90th day after that and to even lower level at the115th day after

Table 3 Effect of different N application patterns on soil OM, NO_3^--N and NH_4^+-N content after the fourth-season and sixth-season harvests[1)]

Treatment	Soil OM (g kg^{-1})		Soil NO_3^--N (mg kg^{-1})		Soil NH_4^+-N (mg kg^{-1})	
	After the fourth harvest	After the sixth harvest	After the fourth harvest	After the sixth harvest	After the fourth harvest	After the sixth harvest
4/4 CN	25.1±0.4 d	24.8±0.5 d	36.6±7.7 bc	32.0±1.6 d	15.2±1.2 a	15.4±1.2 a
3/4 CN+1/4 MN	24.9±1.4 d	25.4±1.0 d	34.4±6.9 c	34.6±1.0 cd	16.3±1.4 a	16.7±4.7 a
2/4 CN+2/4 MN	27.6±0.6 c	28.7±1.2 c	41.2±7.2 bc	36.4±3.8 bc	16.1±1.9 a	16.1±3.0 a
1/4 CN+3/4 MN	29.0±1.2 bc	31.5±1.0 b	34.2±1.0 c	39.6±1.7 b	17.6±1.9 a	17.6±2.1 a
2/4 CN+1/4 MN +1/4 SN	33.0±1.7 a	35.3±1.7 a	53.1±8.4 a	50.8±2.8 a	18.1±1.6 a	18.2±1.6 a
CF	31.0±1.3 ab	33.6±1.1 a	46.2±3.8 ab	46.8±2.7 a	18.7±1.7 a	18.4±1.3 a

[1)] OM, organic matter.

Table 4 Inputs of N and C for each treatment in celery season (kg ha^{-1})

Number	Treatment	N input				C input		
		Nitrogen in chemical fertilizer	Nitrogen in pig manure	Nitrogen in corn straw	Total N	Carbon in organic manure	Carbon in corn straw	Total C
1	4/4 CN	450.0	0.0	0.0	450	0	0	0
2	3/4 CN+1/4 MN	337.5	112.5	0.0	450	1130	0	1130
3	2/4 CN+2/4 MN	225.0	225.0	0.0	450	2260	0	2260
4	1/4 CN+3/4 MN	112.5	337.5	0.0	450	3391	0	3391
5	2/4 CN+1/4 MN+1/4 SN	225.0	112.5	112.5	450	1130	4618	5748
6	CF	450.0	493	0.0	943	4743	0	4743

Table 5 Inputs of N and C for each treatment in tomato season (kg ha^{-1})

Number	Treatment	N input				C input		
		Nitrogen in chemical fertilizer	Nitrogen in pig manure	Nitrogen in corn straw	Total N	Carbon in organic manure	Carbon in cron straw	Total C
1	4/4 CN	450.0	0.0	0.0	450.0	0	0	0
2	3/4 CN+1/4 MN	337.5	112.5	0.0	450.0	1130	0	1130
3	2/4 CN+2/4 MN	225.0	225.0	0.0	450.0	2260	0	2260
4	1/4 CN+3/4 MN	112.5	337.5	0.0	450.0	3391	0	3391
5	2/4 CN+1/4 MN+1/4 SN	225.0	112.5	112.5	450.0	1130	4618	5748
6	CF	450.0	462	0.0	912	4992	0	4992

Table 6 Effect of different N application patterns on residual CTC in soil during the sixth growing season (μg kg^{-1})[1)]

Treatment	Before BFA in sixth season	40th day after BFA in sixth season	90th day after BFA in sixth season	115th day after BFA in sixth season
4/4 CN	0.0±0.0 d A	0.0±0.0 e A	0.0±0.0 d A	0.0±0.0 d A
3/4 CN+1/4 MN	43.3±5.1 c B	61.0±5.6 cd A	40.8±5.1 bc B	19.2±5.2 bc C
2/4 CN+2/4 MN	51.0±5.8 c B	70.6±8.1 c A	54.1±8.2 ab B	24.4±2.8 bc C
1/4 CN+3/4 MN	66.2±2.3 b B	102.8±13.4 b A	57.8±16.0 a B	25.0±4.0 b C
2/4 CN+1/4 MN+1/4 SN	0.0±0.0 d D	45.9±4.5 d A	29.4±6.6 c B	16.3±1.7 c C
CF	115.7±12.7 a B	208.9±12.3 a A	63.3±9.0 a C	42.8±6.9 a C

[1)] CTC, chlortetracycline; BFA, basal fertilizer application.
Means within a column followed by the different lowercase letters were significantly different at $P<0.05$ for the different treatments under the same sampling period. Means within a row followed by the different capital letters were significantly different at $P<0.05$ for the different sampling periods under the same treatment.

that. Especially, content of soil CTC in the CF treatment was respectively up to 115.7 μg kg^{-1} (before BFA in the sixth season) and 208.9 μg kg^{-1} (at the 40th day after BFA in the sixth season), and the content of soil CTC in the treatment of 1/4 CN+3/4 MN (1/4 N from chemical fertilizer, 3/4 N from pig manure) was up to 102.8 μg kg^{-1} (at the 40th day after BFA in the sixth season), exceeding ecological security limit (100 μg kg^{-1}) of antibiotic in soil (Karcı and Balcıoğlu 2009). However, the contents of soil CTC in the pattern with low amount of manure (3/4 CN+1/4 MN treatment, 2/4 CN+1/4 MN+1/4 SN treatment) and median amount of pig manure (2/4 CN+2/4 MN treatment, namely 2/4 N from chemical fertilizer, 2/4 N from pig manure) were below 100 μg kg^{-1} during each sampling period.

There was no CTC detected at each sampling period under 4/4 CN treatment. During each sampling period, effects of manure-amended treatments on soil CTC contents were significantly different, which increased along with the increase of pig manure application. Compared to the 4/4 CN treatment, the contents of CTC in all manure-amended soils were significantly increased. Compared to the CF treatment, in the four sampling periods, the 2/4 CN+1/4 MN+1/4 SN treatment took on the biggest decline on CTC content, which respectively reduced by 100% (before BFA in sixth season), 78.0% (at the 40th day after BFA in the sixth season), 53.6% (at the 90th day after BFA in the sixth season) and 61.9% (at the115th day after BFA in the sixth season); whereas 1/4 CN+3/4 MN treatment took on the smallest decline on the CTC content, which reduced by 42.8, 50.8, 8.7 and 41.6%, respectively.

Under the same amount of pig manure application, content of CTC in straw-amended soil was obviously decreased compared with no straw-amended soil (3/4 CN+1/4 MN treatment), respectively by 100% (before BFA in sixth season), 24.8% (at the 40th day after BFA in the sixth season), 27.9% (at the 90th day after BFA in the sixth season) and 14.9% (at the115th day after BFA in the sixth season). So, addition of crop straw facilitated the degradation of CTC in the soil treated with pig manure.

3. Discussion

3.1. Difference of the residual levels of soil tetracycline antibiotics in greenhouse vegetable soil

There are different results on degradation of tetracycline antibiotics (tetracycline, oxytetracycline and chlortetracycline) in manure-amended soil. The residual levels of tetracycline antibiotics in soil was influenced by soil conditions (soil type, temperature, humidity, pH, electrical conductivity (EC), organic matter and so on), initial content of antibiotic in manure, manure application amount, and so on (Jones *et al.* 2005; Wang and Yates 2008; Hu *et al.* 2010). Investigations in Guangzhou and Shenzhen cities by Li *et al.* (2009b) and in Shandong Province by Yin *et al.* (2012) showed that tetracycline antibiotics had relatively high detectable rate, even up to 100%, which was representative of the main types of antibiotics in the vegetable soil. Shen *et al.* (2009) showed that the degradation ratio of the above three antibiotics had a trend of OTC>TC>CTC; and the degradation of them introduced into soil by pig manure was related to the content of antibiotic in manure, the higher the content was, the longer was the half-life of degradation, and then the degradation was more difficult (Wang *et al.* 2009). According to the study by Martínez-Carballo *et al.* (2007) though the contents of TC, OTC and CTC in manure were up to 23, 29 and 46 mg kg^{-1} respectively, there was no OTC and TC detected in the soil applied with the manure and the CTC content was up to 391 μg kg^{-1}. In addition, since CTC was easier to be absorbed by clay mineral and soil than other antibiotics, thus being protected from related biological degradation, so it was relatively difficult to degrade compared with TC and OTC (Shen *et al.* 2009) but liable to accumulate in soil. In this study, the contents of OTC (1.33 mg kg^{-1}) and TC (0.87 mg kg^{-1}) in commercial pig manure were obviously lower than that of CTC (4.51 mg kg^{-1}), thus the contents of them reduced below detection limit due to degradation process and there were no OTC and TC detected at the 40th day after BFA. However, there was CTC detected during the sixth growing season (spring-season tomato), which was substantially consistent with the results in Austria obtained by Martínez-Carballo *et al.* (2007). Hu *et al.* (2010) indicated that the content of OTC, TC, and CTC in soil in winter was in the range of 124–2 683, 21–105 and 33–1 079 μg kg^{-1}, respectively; whereas in summer the content of TC in soil was only 2.5 μg kg^{-1} and the other two could not be detected, suggesting significant difference of soil antibiotic between different seasons. The compositions of tetracycline antibiotics detected in the same soil had different characteristics under different vegetable cultivations (Tai *et al.* 2011).

3.2. Effects of addition of straw on the degradation of chlortetracycline in greenhouse vegetable soil

Indoor simulation experiment illustrated that the change in the content of OTC introduced into soil *via* chicken manure and pig manure took on an "L" shape, that is to say, the content reduced rapidly at early stage of cultivation, and then stabilized at late stage (Wang and Yates 2008). In this study, from the 40th to 115th day after BFA in the sixth season, contents of CTC in manure-amended soil took on almost similar downward trends during the sixth-season vegetable (spring-season tomato). The CTC content was still higher at the 40th day after BFA in the sixth season, and thereafter reduced greatly to a relatively low level at the 90th day after that and to even lower level at the115th day after that. However, treatment with straw addition had the lowest soil CTC residue, and illustrating straw facilitated the degradation of antibiotic.

The decomposition of straw in soil followed the trend: Fast in early stage and slow in late stage, and it mainly took place in the early 8 weeks and terminated in subsequent 16–32 weeks (Li *et al.* 2007). Straw decomposition was capable of supplying a large amount of carbon sources to soil microorganism, enhancing the microbial activity, thus promoting the microbial degradation of tetracyclines (Kühne *et al.* 2000; Verma *et al.* 2007; Rottmann *et al.* 2010). The ratio of C/N suitable for soil microbes was 25–30:1. In our study, under the identical input of nitrogen in all treatments except CF treatment, the total input amount of carbon in straw treatment was significantly higher than that in other treatments, and the ratio of C/N in straw treatment was up to 41:1 (Tables 4 and 5). Addition of straw provided energy to microbial activity at a certain content, whereby promoting CTC degradation. He *et al.* (2011) disclosed that rice straw had clear promotion on degradation of tetracyclines in pig manure, and the degradation rate of TC, OTC, and CTC in pig manure compost after adding straw increased by 26, 20 and 34%, respectively. In this study, under equal amounts of pig manure treatment, content of CTC in straw-amended soil (0–20 cm) was decreased respectively by 24.8, 27.9 and 14.9% at the 40th, 90th and 115th day after BFA, compared with no straw-amended soil. However, in CF treatment, there was the highest soil CTC residue in 0–20 cm soil layer, which respectively up to 115.7 μg kg^{-1} (before BFA in the sixth season) and 208.9 μg kg^{-1} (at the 40th day after BFA in the sixth season), exceeding ecological security limit (100 μg kg^{-1}) of antibiotic in soil (Karcı and Balcıoğlu 2009). Some researches have showed that, tetracycline antibiotics in soil may cause water body pollution by surface runoff and leaching (Blackwell *et al.* 2009; Kim *et al.* 2011), and drug resistance of bacteria, thus threatening human health. Zhang *et al.* (2008) collected 41 soil samples applied with manure and results showed that the mean residues of the CTC, TC and OTC in the surface soils from agricultural field treated with animal manures were 12, 13 and 38 times as high as those from agricultural field without animal manures, suggesting that manure is an important source of tetracycline antibiotics in the agricultural soil. The ratio of the CTC, TC and OTC content in subsurface soils (20–40 cm) to those in the surface soil (0–20 cm) varied from 0.02–1.36, 0.02–1.91 and 0.01–2.13, respectively. These showed that tetracyclines may migrate downward in farm soil. Therefore, there was an urgent need for further investigating the effects of combined application of organic manure and straw on degradation and migration mechanism of tetracycline antibiotics.

3.3. Optimal combined application pattern of organic manure and chemical fertilizers with the highest yield and profit and the least soil antibiotic residue

Effective and widespread use of organic manures has great promise as a source of multiple nutrients and ability to improve soil characteristics and enhance soil fertility (Moller 2009; Suthamathy *et al.* 2013). It can substantially reduce chemical fertilizer use without detrimentally affecting crop yields and can even serve to stabilize global crop prices (Alex 2009). Numerous studies have showed that appropriate proportions of organic manure and chemical fertilizer can promote increases of vegetable production and economic benefit (Masarirambi *et al.* 2012; Isitekhale *et al.* 2013). In this study, the combined application of organic manure and chemical fertilizers with largely reduced amounts of fertilizer was able to guarantee the stable yield of tomato and celery, for example, the yield and economic benefit were highest in the treatment of 3/4 CN+1/4 MN for the early four seasons (from the first to fourth season), and it was the same case in the treatment of 2/4 CN+1/4 MN+1/4 SN for the late two seasons (from the fifth to sixth season). Furthermore, the residual levels of soil antibiotic in the two treatments were relatively lower than all the other treatments. In contrast, due to high application amounts of organic manure and chemical fertilizers in CF treatment, the residual level of antibiotic in soil was the highest, but the yield and economic benefit in this pattern was not the case. Thus, considering the economic benefit and ecological safety, excessive amount of organic manures was not desirable. Some researches indicated that vegetables could absorb, transport, and accumulate antibiotics from manure-amended soils (Boxall *et al.* 2006; Dolliver *et al.* 2007). Antibiotic residues in plants generally tended to increase with increasing antibiotic contents in manure-amended soil (Kumar *et al.* 2005). Ready-to-eat fresh vegetable with larger quantity and quicker transportation of antibiotics among food chains to human body may pose more serious health risks than other crops (Du and Liu 2012). In this study, further analysis on the relationship between the vegetable yield and residual content of CTC in soil showed that there was a significant negative correlation between the vegetable yield and the residual content of CTC as a whole under pig manure application. The correlation coefficients between the total yield of vegetable from the first to sixth seasons and the residual content of CTC at the above four sampling periods of the sixth season were –0.57, –0.60, –0.62 and –0.57 (df=14, $r_{0.05}$=0.50, $r_{0.01}$=0.62), respectively. It can be seen that for the long term large inputs of pig manure, high accumulation of CTC in soil may potentially bring a negative effect on vegetable growth at a certain extent and thus influence the vegetable yield. Therefore, for the four treatments of combined application of organic manure and chemical fertilizers, the treatments with 1/4 N from pig manure had relatively higher yield, economic benefit and lower antibiotic residue. Addition of straw could effectively promote the degradation of antibiotic brought into soil by the applied organic manure, thus it could provide a high-efficiency and environment-friendly approach for treating residual antibiotic in soil. Due to limitation of the scale of location experiment, it is still necessary to investigate the effects of straw on degradation of residual antibiotic in excessive pig manure and other organic manures, and degradation of accumulated antibiotic in soil, so as to provide a basis of removing organic pollutant in soil by means of straw without reducing yield and economic benefit.

4. Conclusion

After the application of pig manure, content of CTC in soil took on almost similar downward trend. At the 40th day after basal fertilizer application in the sixth season, the content of chlortetracycline in manure-amended soil was still relatively high, and declined continuously after that until to the lowest level at the115th day after basal fertilizer application in the sixth season.

The residual level of CTC in soil took on an increasing trend along with the increase of applied amount of pig manure. The treatment of 2/4 CN+1/4 MN+1/4 SN had the lowest residue level of CTC in soil compared with other treatments applied with pig manure. Under the same amount of pig manure application, content of CTC in straw-amended soil was obviously decreased compared with no straw-amended soil (3/4 CN+1/4 MN treatment), and averagely decreased by 41.9% for four sampling periods in sixth season. Conventional fertilization pattern was liable to result in accumulation of CTC and had the largest content in greenhouse soil.

Based on the optimal combination of yield and economic benefit and low residue of antibiotic in soil, the combined application of 3/4 CN+1/4 MN was suitable for use in the previous four seasons (from the first to fourth season), whereas the combined application of 2/4 CN+1/4 MN+1/4 SN was suitable for the fifth and sixth seasons. Also, the fertilization pattern of 2/4 CN+1/4 MN+1/4 SN facilitated to enhance the content of soil organic matter, and maintain constant supply of soil nitrate nitrogen. Therefore, in the case of application of chemical fertilizer and pig manure, straw played a key role in achieving optimal yield and profit and minimal risk of antibiotic residue in greenhouse soil.

5. Material and methods

5.1. Experimental materials

A 3-yr field experiment was conducted in a sunlight greenhouse in Diliubu Village of Xinkou Town in Xiqing District, Tianjin, with annual average temperature of 11.6°C and the total amount of natural rainfall of 586 mm, which belongs to warm temperature sub-humid continental climate. The experiment began in October, 2009 (the greenhouse has been used for cultivation for 7 yr at the beginning of this experiment), with a rotation of autumn-winter season celery (variety: Wentula) and spring season tomato (variety: Jinxin). The soil type was Fluvo-aquic soil (medium loam) with the depth of groundwater of 120 cm. Prior to the experiment, soil collected from the topsoil (0–20 cm) was measured with 25.4 g kg^{-1} organic matter, 136.7 mg kg^{-1} nitrate nitrogen, 144.6 mg kg^{-1} available P, 404.0 mg kg^{-1} available potassium, and pH of 7.9. In addition, there were no tetracycline, oxytetracycline and chlortetracycline detected in 0–20-cm soil layer. As regarding to the experiment, vegetable was cultivated for six growing seasons.

5.2. Experimental design

The field experiment comprised six treatments as follows: 4/4 CN (CN, nitrogen in chemical fertilizer), 3/4 CN+1/4 MN (MN, nitrogen in pig manure), 2/4 CN+2/4 MN, 1/4 CN+3/4 MN, 2/4 CN+1/4 MN+1/4 SN (SN, nitrogen in straw), and

CF (farmer's conventional fertilization). In addition to CF treatment, the other treatments with greatly reduced application amounts of fertilizer and each of the amounts of N, P_2O_5 and K_2O was equal, namely, the total amounts of N, P_2O_5 and K_2O in tomato season were 450.0, 225.0 and 600.0 kg ha^{-1}, respectively; whereas those in celery season were 450.0, 300.0 and 600.0 kg ha^{-1}, respectively. In CF treatment, the amounts of N, P_2O_5 and K_2O in chemical fertilizers in tomato season were 450.0, 300.0 and 450.0 kg ha^{-1} respectively, and the amount of pig manure was 30.0 t ha^{-1}, and those in celery season were 450.0, 600.0 and 300.0 kg ha^{-1}, respectively, and the amount of pig manure was 32.0 t ha^{-1}. The specific inputs of N and C for each treatment in the autumn-winter season celery and the spring-season tomato were given in Tables 4 and 5. The experiments were laid out in randomized complete block design with three replications. Each plot was 14.4 m^2 (width of 2.4 m×length of 6 m), in which each spacing in a row and between rows of tomato was 0.30 and 0.60 m, respectively and the planting density was 25 000 plant ha^{-1}; and each spacing in a row and between rows of celery was 0.20 and 0.15 m, respectively, and the planting density was 330 570 plants ha^{-1}. PVC plates (depth of 105 cm with underground depth 95 cm and aboveground depth 10 cm, and thickness of 4 mm) were embedded for preventing transversal migration of nutrients and water between plots.

All the organic manure was applied as basal fertilizer. In the tomato season of CF treatment, 20% of N fertilizer and 80% of P fertilizer were applied as basal fertilizer, and the rest N, P_2O_5 and K_2O were used as topdressing for four times in equal amount, respectively, namely, each time 20% N, 5% P and 25% K were used. However, In the tomato season of other treatments, 20% N, 70% P, and 20% K of the total amount of chemical fertilizer were used as basal application, and the rest N and K were applied as top dressing for 4 times (each at flowering stage, and the first, second, and third cluster fruit expanding stage of tomato), in which the topdressing ratios of N were 30, 30, 10 and 10%, respectively and those of K were 10, 30, 30 and 10%, respectively, and the rest P_2O_5 were applied in an amount of 15% for the first and second topdressing, respectively. In celery season of CF treatment, 20% of N fertilizer and 100% of P fertilizer were applied as basal application, and the rest N and the total K_2O were used as topdressing for three times in equal amount, respectively. However, in celery season of other treatments, the same proportions of chemical fertilizer as those in tomato seasons of other treatments were used; and the rest N and K were applied as top dressing for three times (each at 5–6-leaf stage, 8–9-leaf stage and 11–12-leaf stage of celery) , in which the top dressing proportions of N were 35, 35 and 10%, respectively, and K, 10, 35 and 35%, respectively, and the rest P was fully applied as the first top dressing. Basal fertilizer was applied by rotary tillage into soil after scattering, and topdressing fertilizer was applied by dissolving in water and washing with water.

Chemical fertilizers used in the experiment included urea (N 46%), calcium superphosphate (P_2O_5 12%), diammonium phosphate (N 18%, P_2O_5 46%), and potassium chloride (K_2O 60%). The contents of N, P_2O_5, K_2O, and C of commercial pig manure were (2.17±0.13)%, (1.39±0.14)%, (1.63±0.19)%, and (218.0±5.0) g kg^{-1} (by dry weight), the water content of the pig manure was (28.9±4.6)%, respectively, and the contents of residual OTC, TC, and CTC in pig manure were 1.33, 0.87 and 4.51 mg kg^{-1}, respectively. The contents of N, P_2O_5, K_2O, and C of corn straw used in the experiment were (1.04±0.10)%, (0.32±0.08)%, (1.69±0.17)%, and (426.9±8.2) g kg^{-1} (by dry weight), respectively, and the water content of which was (64.9±6.4)%.

All treatments (except CF treatment) were subjected to irrigation in terms of field capacity, in which once the field capacity is less than 60%, irrigation was implemented. CF treatment was subjected to irrigation in traditional pattern, that is, broad irrigation was applied regularly. In order to ensure the accuracy of irrigation amount, each plot was equipped with separate PVC inlet pipe and the irrigation amount was recorded by a water meter. The total irrigation amounts of celery and tomato during their growing periods were separately 4 167 and 4 861 m^3 ha^{-1} for CF treatment, and 3 334 and 3 889 m^3 ha^{-1} for the other treatments.

5.3. Measurements and methods

Soil sampling: After the fourth-season and sixth-season harvests, soils were sampled for measuring the contents of soil organic matter, NO_3^--N and NH_4^+-N. As for the sixth-season tomato, soils were sampled for measuring the contents of soil tetracycline antibiotics before basal fertilizer application (BFA), at the 40th day (at flowering period), at the 90th day (at fruit development period) and at the 115th day (after harvest) after BFA, respectively. Ten sites in each plot were selected to sample the soils of surface layer (0–20 cm), which were air-dried and sieved with a sieve having 1 mm aperture for use.

The optimized method for measuring tetracyclines (TC, OTC, and CTC) (Zhang *et al.* 2013): 3.00 g soil was weighed and placed in a 50-mL centrifuge tube. 10 mL Na_2EDTA -mcIlvaine buffer solution containing equal volume of acetone (containing 0.01 mol L^{-1} oxalic acid-methanol), was added in the centrifuge tube, vortexed for 1 min, subjected to ultrasonic extraction for 10 min, and centrifuged at 4 500 r min^{-1} at 4°C for 10 min. Supernatant was collected, and 8 and 6 mL of extracting solutions were added into rest sediment in turn for repeating extraction twice. Supernatants obtained in above three steps were mixed and concentrated by blowing nitrogen to about 12 mL at 40°C to obtain extracted solution. Poly-Sery HLB column was washed succesively with 5 mL of methanol and 5 mL of ultra pure water for activation, and then the extracted solution and 5 mL of 5% aqueous solution of methanol flowed in turn through the Poly-Sery HLB column at the flowing rate of about 1 mL min^{-1} by means of a vacuum pump, and continuously pumped under vacuum for 10 min. After then, HLB column was eluted with 5 mL eluting solution and eluate was collected and concentrated by blowing nitrogen to less than 0.5 mL at 40°C to obtain final extract. The final extract was dissolved with mixed mobile phase to make 1 mL, ultrafiltered with 0.22 µm filter membrane, and transferred to a brown test bottle for measurement by high performance liquid chromatograph (HPLC). The recovery rates of OTC, TC and CTC in tested soil were respectively 80.1, 61.2 and 52.1%. The optimized method recovery was applicable, and greatly reduced the sample analysis time and promoted analysis efficiency.

Soil after harvest of the fourth season and sixth-season crops were sampled for analyzing the contents of soil organic matter, NO_3^--N and NH_4^+-N, in which organic matter was measured by the methods of Potassium dichromate-concentrated sulfuric acid oxidation (outside heating) and ferrous ammonium sulfate titration, and NO_3^--N and NH_4^+-N was extracted by 2 mol L^{-1} KCl solution and measured by double wavelength ultraviolet spectrophotometry method for NO_3^--N and indophenol blue spectrophotometry for NH_4^+-N.

5.4. Data statistics and analysis

Benefit income=Output value–(Fertilizer cost+Irrigation cost+Pesticide cost). Price of N, P_2O_5, K_2O, organic manure, straw and vegetable (the first, third, and fifth harvest is celery, and the second, fourth and sixth harvest is tomato) is separately 4.3, 4.8, 5.3, 0.5, 0.06 and 2.8 CNY kg^{-1} for the first harvest, 4.3, 4.8, 5.3, 0.5, 0.06 and 2.3 CNY kg^{-1} for the second harvest, 4.6, 5.8, 6.8, 0.6, 0.1 and 1.4 CNY kg^{-1} for the third harvest, 5.2, 5.0, 6.7, 0.6, 0.1 and 2.6 CNY kg^{-1} for the fourth harvest, 5.2, 5.0, 6.7, 0.6, 0.1 and 2.6 CNY kg^{-1} for the fifth harvest, 5.2, 5.0, 6.7, 0.6, 0.1 and 2.4 CNY kg^{-1} for the sixth harvest; price of irrigation water is 0.5 CNY m^{-3}; Price of pesticide is separately 3000 and 2250 CNY ha^{-1} in tomato season and celery season.

The amount of pig manure (dry weight)=N input rate from pig manure/N content in pig manure (dry weight). The amount of P_2O_5 or K_2O in pig manure=The amount of pig manure (dry weight)×P_2O_5 or K_2O content in pig manure (dry weight). The amount of C in pig manure=The amount of pig manure (dry weight)×C content in pig manure (dry weight). The amount of commercial pig manure=The amount of pig manure (dry weight)/(1–Water content in commercial pig manure). The calculation of each corresponding item in corn straw is similar to pig manure mentioned above. The amount of P_2O_5 or K_2O in chemical fertilizer=The amount of total P_2O_5 or K_2O–(The amount of P_2O_5 or K_2O in pig manure+The amount of P_2O_5 or K_2O in corn straw).

All the reported data were analyzed by Microsoft Excel 2003 and SPSS 13.0, with significant difference between data detected by means of Duncan new multiple range method.

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