UNIVERSITI PUTRA MALAYSIA

TOXICITY AND DEGRADATION OF ANIMAL ANTIBIOTIC RESIDUE IN SOIL

WANG YAN

IB 2010 6

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By

WANG YAN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

October 2010

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

TOXICITY AND DEGRADATION OF ANIMAL ANTIBIOTIC RESIDUE IN SOIL

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October 2010

Chairman: Associate Professor Liang Juan Boo, PhD

Institute: Bioscience

Antibiotics are widely used in farm animals especially pig and poultry, however, they are poorly absorbed by the animals and entered the environment mainly via the application of manure for crops production. This thesis aimed to i) investigate the status of the use of antibiotic by pig and poultry farmers in Malaysia, ii) evaluate the environmental risks when manures contaminated by antibiotic are used as fertilizer in soils and iii) propose a possible method to accelerate the degradation of antibiotic in the environment.

To achieve the first objective, a survey on the use of the antibiotics by Malaysian pig and poultry farmers was conducted. The results indicated that tetracyclines (TCs) and sulfanimines (SAs) were the most widely used antibiotics in the Malaysian pig and poultry farms. Among the highest concentrations detected in the diets were

51,948.5 and 28,791.1 ng/g of sulfamethazine in diets of grower pigs and broilers, respectively, while in the fecal samples collected, 130.0 ng/g sulfamethazine and 129.2 ng/g chlortetracycline were detected in the finisher pigs and 380.4 ng/g sulfamethazine in grower broilers.

To evaluate the environmental risks of antibiotic residues in the soil-plant system, three experiments were carried out. The first experiment examined the effect of different concentrations of TCs and SAs on the environmentally beneficial bacteria, and *Shewanella decolorationis* S12 was used as the model bacteria for the study. The results showed that the antibiotics significantly inhibited Fe (III) reduction and dye decoloration activities of *S. decolorationis* S12 in the reduction system. The above inhibition was suggested to be due to a decrease in live *S. decolorationis* S12 population and/or modifications of their cell structures in the presence of antibiotics. The second experiment studied the effects of sulfadiazine (SD) residues on soil nitrogen mineralization and soil microbial community. The results showed that SD significantly inhibited the mineralization of N and microbial parameters including ammonifying and nitrifying bacteria population, microbial biomass N, urease and nitrate reductase and soil microbial communities activities, however, the activities of soil microbial communities can be used as the sole indicator to monitor changes of soil N mineralized under SD pollution. The third experiment investigated the toxicity effects of SD residues on vegetable lettuce. The results clearly showed that SD were absorbed and accumulated in the different organs of the lettuce and adversely

affected their growth. It is suggested that high concentrations of SD caused physiological damages to roots of the lettuce, allowing easy translocation of SD from the roots to the leaves and caused damages to the leaf cells and thus prohibited photosynthesis.

A goethite (α-FeOOH)-oxalate Fonten-like system was evaluated to accelerate the photodegradation of antibitotics under UV irradiation. The results indicated the optimal pH value and initial concentration of oxalic acid in the system were 3.5 and 4.0 mM, respectively. The eco-toxicity of the intermediate products was found to be lower than the mother SD compound, thus suggesting the positive effect of photodegradation on the impact of SD on the environment. These results can help to better understand the natural degradation of SD (possibly other antibiotics as well) and offer the possibility of treating effluents, including livestock wastewater, contaminated with antibiotics at low cost since goethite, oxalate and light exist in natural environment.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

KERACUNAN DAN DEGRADASI RESIDU ANTIBIOTIK HAIWAN DALAM TANAH

Oleh

WANG YAN

October 2010

Pengerusi: Profesor Madya Dr. Liang Juan Boo, PhD

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Antibiotik digunakan dengan meluas dalam ternakan, terutama babi dan ayam Namun begitu, antibiotik tidak diserap dengan baik oleh ternakan dan memasuki alam sekitar terutamanya melalui pengunaan najis ternakan sebagai baja untuk pengeluaran tanaman. Tesis ini bertujuan untuk (i) menyiasat status penggunaan antibiotik oleh penternak babi dan ayam di Malaysia, ii) menilai risiko alam sekitar apabila najis ternakan yang dicemari oleh antibiotik digunakan dalam tanah dan iii) mencadangkan satu kaedah untuk mempercepatkan degradasi antibiotik dalam alam sekitar.

Untuk mencapai matlamat pertama, satu survei mengenai penggunaan antibiotik oleh penternak-penternak babi dan ayam di Malaysia telah dijalankan. Keputusan

menunjukkan tetracyclines (TCs) dan sulfanimines (SAs) ialah antibiotik yang paling banyak digunakan dalam penternakan babi dan ayam di Malaysia. Antara kepekatan yang tertinggi yang dikesan dalam diet babi dan ayam pedaging, masing-masing, ialah 51,948.5 dan 28,791.1 ng/g sulfamethazine, manakala dalam sampel najis yang dikumpul, 130.0 ng/g sulfamethazine dan 129.2 ng/g chlortetracycline telah dikesan pada babi dan 380.4 ng/g sulfamethazine pada ayam pedaging.

Untuk menilai risiko alam sekitar pada residu antibiotik dalam sistem tanah-tumbuhan, tiga eksperimen telah dijalankan. Eksperimen pertama menilai kesan kepekatan TCs dan SAs yang berbeza ke atas bakteria persekitaran yang bermanfaat dengan menggunakan *Shewanella decolorationis* S12 sebagai model bakteria bagi kajian tersebut. Keputusan menunjukkan antibiotik menyekat secara signifikan pengurangan Fe (III) dan aktiviti pemudaran warna *S. decolorationis* S12 dalam sistem pengurangan. Penyekatan di atas telah dicadangkan adalah disebabkan oleh pengurangan populasi *S. decolorationis* S12 hidup dan/atau pengubahsuaian struktur sel mereka dengan kehadiran antibiotik. Eksperimen kedua mengkaji kesan residu sulfadiazine (SD) pada pemineralan nitrogen tanah dan komuniti mikrob tanah. Keputusan menunjukkan SD menyekat secara signifikan pemineralan N, pengammoniaan dan penitritan populasi bakteria, biojisim mikrob N, urease dan reduktase nitrat dan aktiviti komuniti mikrob, namun begitu aktiviti komuniti mikrob boleh digunakan sebagai penanda tunggal untuk mengawasi perubahan N tanah yang

dimineralisasikan oleh pencemaran SD. Eksperimen ketiga mengkaji kesan toksik residu SD pada sayur selada. Keputusan jelas menunjukkan SD diserap dan terkumpul dalam organ-organ selada dan sangat menjejas pertumbuhannya. Adalah dicadangkan bahawa kepekatan SD yang tinggi menyebabkan kerosakan fisiologikal pada akar selada, membenarkan translokasi yang mudah bagi SD dari akar ke daun dan menyebabkan kerosakan sel-sel daun dan dengan demikian, menghalang fotosintesis.

Satu sistem menyamai Fonten goetit (α-FeOOH)-oxalate telah dinilai untuk mempercepatkan fotodegradasi antibiotik di bawah penyinaran UV. Keputusan menunjukkan nilai optima pH dan kepekatan awal asid oksalik dalam sistem masing-masing ialah 3.5 dan 4.0 mM. Ketoksikan eko produk-produk pertengahan didapati adalah lebih rendah daripada sebatian sulfadiazine asal, dengan ini mencadangkan kesan positif fotodegradasi ke atas impak sulfadiazine pada persekitaran. Keputusan ini boleh membantu untuk memahami dengan lebih baik degradasi sulfadiazine secara semulajadi (mungkin juga bagi antibiotik-antibiotik lain) dan menawarkan kemungkinan merawat efluen, termasuk air buangan ternakan, yang dicemari dengan antibiotik pada kos rendah kerana goetit, oksalat dan cahaya wujud dalam alam sekitar semulajadi..

ACKNOWLEDGEMENTS

I would like to express my profound gratitude to my supervisor, Associate Professor Dr. Liang Juan Boo for his dedicated supervision, advice and encouragement throughout the period of my study. My appreciations are also extended to Professor Dr. Li Fang Bai, Associate Professor Dr. Loh Tech Chwen and Professor Dr. Ho Yin Wan, members of my Supervisory Committee, for their advice and suggestion. To Professor Dr. Li Fang Bai, thank you for your support, guidance and patience during my study at Guangdong Institute of Eco-environmental and Soil Science, China.

To Dr. Lai Pui Wah, thank you for translating the abstract into Bahasa Malayu and the help during my study. Appreciation is also extended to Huang Xiaodan, Tan Hui Yin, Qi Xiaojing, Zhang Qin, Saminathan and other coursemates and friends in UPM for their friendship and support.

I would also like to thank the staffs (Liu Chuanping, Liu Chenshuai, Tao Liang, Liu Tongxu) and students (Li Xiaomin, Wang Lusong, Wu Chunyuan, Cao Fang, Liu Liang, Li Xiujuan, Lan Qin, Song Jian, Chen Manjia, Wang Yongkui, Li Huimin, Zhang Wei, Xu Xianghua, Sun Kewen, Wang Xugang, Zhang Lijia, Wu Changan) from Guangdong Institute of Eco-environmental and Soil Science, China for their assistance and kindness. I am grateful for the friendship and team work during my research.

Finally, I am deeply indebted to my parents Wang Shouquan and Xie Yuying, brother Wang Min and his wife Zhang Qi for their understanding and endless support. I also owe my thanks to my beloved husband Tang Qiang for his love, patience and encouragement throughout the duration of my study.

I certify that an Examination Committee met on **28th October 2010** to conduct the final examination of **Wang Yan** on her **PhD** thesis entitled "Veterinary Antibiotic Tetracyclines and Sulfanimines Residues in the Soil-Plant System" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded Doctor of Philosophy.

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This thesis was submitted to the Senate of Universiti Putra Malaysia has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently submitted for any other degree at Universiti Putra Malaysia or other institutions.

WANG YAN

Date: 28 October 2010

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