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TOXICITY AND DEGRADATION OF ANIMAL ANTIBIOTIC RESIDUE IN SOIL

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By

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TOXICITY AND DEGRADATION OF ANIMAL ANTIBIOTIC RESIDUE IN SOIL

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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 sulfanamines (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 antibiotics 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

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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 penggunaan 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 sulfaminines (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 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 reuktase 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 menjejaskan pertumbuhannya. Adalah dicadangkan bahawa kepekatannya 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 \((\alpha-\text{FeOOH})-\text{oxalate}\) telah dinilai untuk mempercepatkan fotodegradasi antibiotik di bawah penyinaran UV. Keputusan menunjukkan nilai optima pH dan kepekan 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.
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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 Sulfanilamines 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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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
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABSTRACT</td>
<td>ii</td>
</tr>
<tr>
<td>ABSTRAK</td>
<td>v</td>
</tr>
<tr>
<td>ACKNOWLEDGEMENTS</td>
<td>viii</td>
</tr>
<tr>
<td>APPROVAL</td>
<td>x</td>
</tr>
<tr>
<td>DECLARATION</td>
<td>xii</td>
</tr>
<tr>
<td>LIST OF TABLES</td>
<td>xiii</td>
</tr>
<tr>
<td>LIST OF FIGURES</td>
<td>xvi</td>
</tr>
<tr>
<td>LIST OF PLATES</td>
<td>xvii</td>
</tr>
<tr>
<td>LIST OF ABBREVIATIONS</td>
<td>xxii</td>
</tr>
</tbody>
</table>

## CHAPTER

1 INTRODUCTION                                                               1

2 LITERATURE REVIEW                                                        3

2.1 Antibiotic                                                            3

2.1.1 Summary of antibiotic                                                3

2.1.2 Usage of antibiotics                                                 4

2.2 Veterinary use of antibiotic                                            5

2.2.1 Usage of antibiotics in livestock                                    5

2.2.2 Tetracycline and sulfonamides                                        8

2.2.3 General concerns on the use of antibiotics in livestock production   13

2.2.4 Regulations of the use of antibiotics in livestock production        15

2.3 Occurrence of antibiotic in the environment                           18

2.4 Fate of antibiotic residues in the soil                               19

2.4.1 Sorption                                                            20

2.4.2 Degradation of antibiotics in the soil                              22

2.5 Hazards of antibiotics in the environment                             25

2.5.1 Hazards on Microorganisms                                           27

2.5.2 Hazards on aquatic environment                                      29

2.5.3 Hazard on plant growth                                              30

2.5.4 Antibiotic resistance                                               31

3 A SURVEY ON THE UTILIZATION OF ANTIBIOTICS IN MALAYSIAN PIG AND BROILER FARMS 34

3.1 Introduction                                                          34

3.2 Materials and methods                                                 36

3.2.1 Sample collection                                                  36

3.2.2 Survey Questionnaire                                                37
3.2.3 Laboratory analyses of TCs and SAs concentration 38

3.3 Results and Discussion 38
  3.3.1 Analysis of TCs and SAs 38
  3.3.2 Antibiotic use in Malaysia 39
  3.3.2 Concentration of TCs and SAs used in pig and poultry farms in Malaysia 43

3.4 Conclusion 46

4 EFFECTS OF OXYTETRACYCLINE AND SULFACHLOROPYRIDAZINE RESIDUES ON BENEFICIAL BACTERIA 47

4.1 Introduction 47

4.2 Materials and Methods 49
  4.2.1 Chemicals 49
  4.2.2 Preparation and characterization of goethite 49
  4.2.3 Microbial strain and culture conditions 50
  4.2.4 Bacteria reduction experiments 51
    4.2.4.1 Batch cultures for Fe (III) reduction and dye degradation 51
    4.2.4.2 The analysis of Fe (II) 52
  4.2.5 Cell structure experiment 52
  4.2.6 Statistical Analysis 53

4.3 Results and Discussion 53
  4.3.1 Effects of OTC and SCP on goethite reduction 53
  4.3.2 Effects of SCP on dye degradation 59
  4.3.3 Effect of OTC and SCP on the population of S. decolorationis S12 60
  4.3.4 Effect of OTC on cell structure of S. decolorationis S12 60
  4.3.5 Reasons for the inhibition of S. decolorationis S12 reduction efficiency by antibiotic 64

4.4 Conclusion 65

5 EFFECT OF ANTIBIOTIC SULFADIAZINE RESIDUE ON SOIL NITROGEN MINERALIZATION AND RELATED MICROBIAL PARAMETERS 66

5.1 Introduction 66

5.2 Materials and methods 68
  5.2.1 Preparation of the soil 68
  5.2.2 Sample incubation 68
  5.2.3 Sample analyses 69
  5.2.4 Data analyses 71

5.3 Results and discussion 71
  5.3.1 Effect of sulfadiazine (SD) on soil nitrogen mineralization 71
  5.3.2 Effects of SD on soil microbial N and populations of ammonifying and nitrifying bacteria 73
  5.3.3 Effect of SD on enzyme activity 75
  5.3.4 Effect of SD on soil microbial community diversity 77
5.3.5 The relationship between soil N mineralization and the microbial parameters in SD contaminated soil 78
5.4. Conclusion 81

6 TOXIC EFFECTS OF ANTIBIOTIC SULFADIAZINE ON VEGETABLE LETTUCE 82
6.1 Introduction 82
6.2 Materials and Methods 83
6.2.1 Preparations of Plant Materials and Treatment Solutions 83
6.2.2 Plant Sampling and Measurement 84
6.2.3 Analysis of SD Concentration 85
6.2.4 Analysis of Roots Electrolyte Leakage 86
6.2.5 Preparation and Analysis of Plant Samples for Cell Structure 87
6.2.6 Statistical Analysis 88
6.3 Results and Discussion 89
6.3.1 Effect of SD on Plants Growth 89
6.3.2 Concentration of SD in Lettuces 91
6.3.3 Electrolyte Leakage of the Roots 92
6.3.4 Effect of SD on Cell Structures of Lettuce 93
6.3.5 Process for the toxicity of Lettuces by antibiotic SD 95
6.4 Conclusions 96

7 PHOTODEGRADATION OF SULFADIAZINE BY GOETHITE-OXALATE SUSPENSION UNDER UV LIGHT IRRADIATION 98
7.1. Introduction 98
7.2. Materials and methods 101
7.2.1 Chemicals 101
7.2.2 Preparation and characterization of iron oxide 101
7.2.3 Experimental procedure for photodegradation study 102
7.2.4 Analysis of sulfadiazine and its intermediate compounds 103
7.2.5 Determination of ecological toxicity 105
7.3. Results and discussion 105
7.3.1 Photodegradation of sulfadiazine 105
7.3.1.1 Selection of the iron oxide 105
7.3.1.2 Influence of initial oxalic acid concentration 107
7.3.1.3 Influence of pH 110
7.3.2 Photo-oxidation pathway and mineralization of sulfadiazine 112
7.3.3 Ecological toxicity of sulfadiazine during photo-oxidation 119
7.4 Conclusion 121

8 SUMMARY, GENERAL CONCLUSION AND RECOMMENDATIONS FOR FUTURE RESEARCH 123
8.1 Summary and General Conclusion 123
8.2 Recommendation for Future Research 128