

Occurrence and fate of veterinary pharmaceuticals and antibiotics in soils fertilized with manure and evaluation of their persistence and potential to migrate to groundwater bodies

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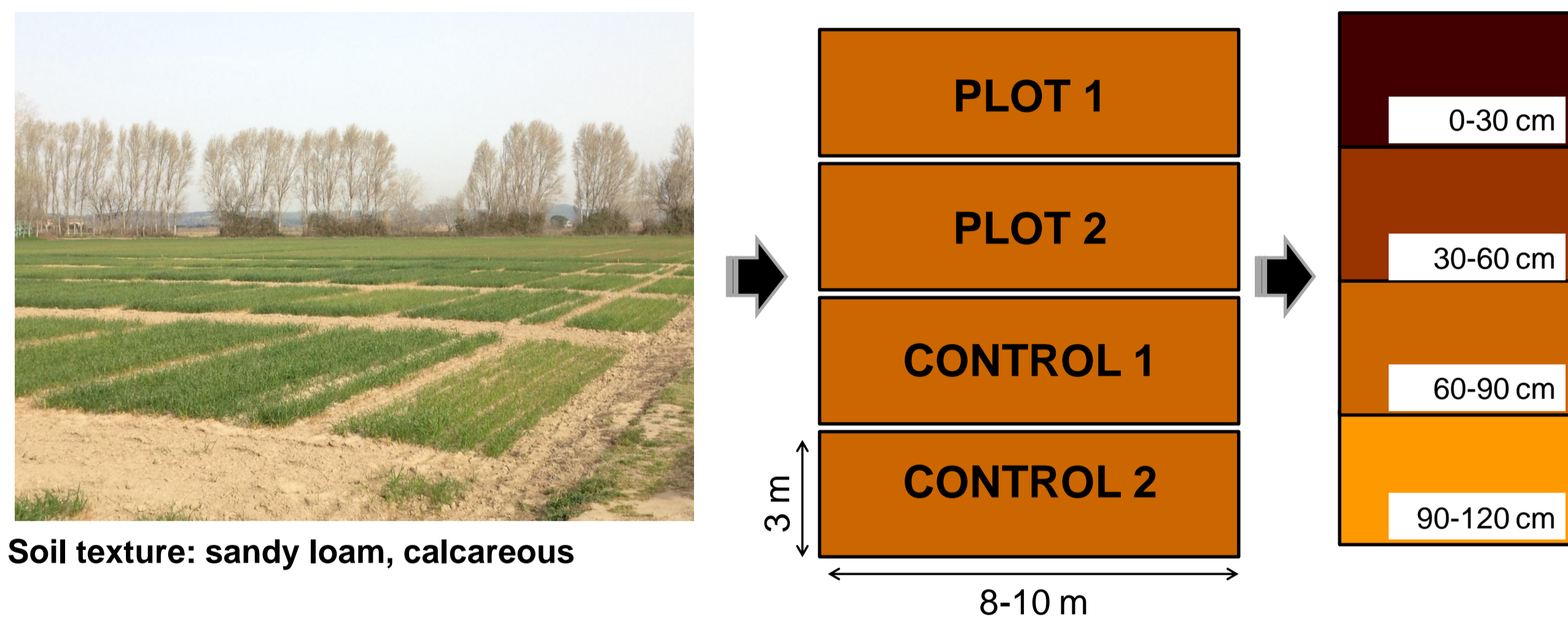


1. INTRODUCTION

Animal manure is extensively used as soil fertilizer in agricultural fields as it is rich in nutrients that are beneficial for crop growth and soil conditioning. Nevertheless, animal manure may be an important reservoir of antibiotics and veterinary medicines used in animal husbandry operations. From manure-amended soils, antibiotics and veterinary drugs could be taken up by crops and, thus, enter the human food chain [1], they could leach to surface and groundwater bodies [2] or recalcitrant pharmaceuticals may persist and accumulate in the soil [3]. In this study, the occurrence, distribution and fate of **40 multiple-class antibiotics and veterinary pharmaceuticals** was investigated in field experiments fertilized with animal manure, including control plots without fertilization. The fields under study were cultivated with wheat and barley, alternatively. Soil sampling was performed in December 2015, prior to fertilization, and in February and July 2016, two and seven months after fertilization, respectively. Moreover, manure used as soil amendment was also analyzed. Soil samples were taken at four different soil depths to identify the most persistent pollutants that accumulate in soil over time and those with the highest mobility and that are more liable to leach to groundwater bodies and deteriorate groundwater quality.

2. FIELD EXPERIMENTS

Plots fertilized with 170 kg N/ha: A) cattle manure (2); B) swine slurry (2); 2 control plots (non-fertilized) per treatment.



3. ANTIBIOTICS AND PHARMACEUTICALS

Antibiotics	Compounds	Antibiotics	Compounds	Pharmaceuticals	Compounds
Tetracyclines	Tetracycline Oxytetracycline Doxycycline Chlorotetracycline	Macrolides	Tilmicosin Tylosin Spiramycin Erythromycin Tylvalosin	Anti-protozoan	Salinomycin Monensin Ivermectin Doramectin Toltrazuril
Sulfonamides	Sulfamethoxazole Sulfamethazine Sulfadimethoxine Sulfamerazine	Lincosamides	Lincomycin	Anti-inflammatories	Flunixin Ketoprofen Diclofenac Salicylic acid
Quinolones	Flumequine Oxolinic acid Cinoxacin	B-lactams	Ceftiofur Amoxicillin		
Others	Tiamulin Trimethoprim	Glucocorticoids	Dexamethasone	Anthelmintics	Albendazole Fenbendazole Flubendazole Levamisole
		Fluoroquinolones	Ciprofloxacin Ofloxacin Enrofloxacin Danofloxacin Marbofloxacin		

4. ANALYTICAL METHODS

SWINE SLURRY (LIQUID)

Sample filtration (0.7 µm)
Addition of IS mixture
Addition Na₂EDTA 0.1M

SOLID PHASE EXTRACTION (SPE)

Volume: 10mL
SAX (500mg, 6cc) &
Oasis HLB (200mg, 6cc)

SOIL & SOLID MANURE

Sieve soil (< 2 mm)
1g manure; 2 g soil
Addition of IS mixture

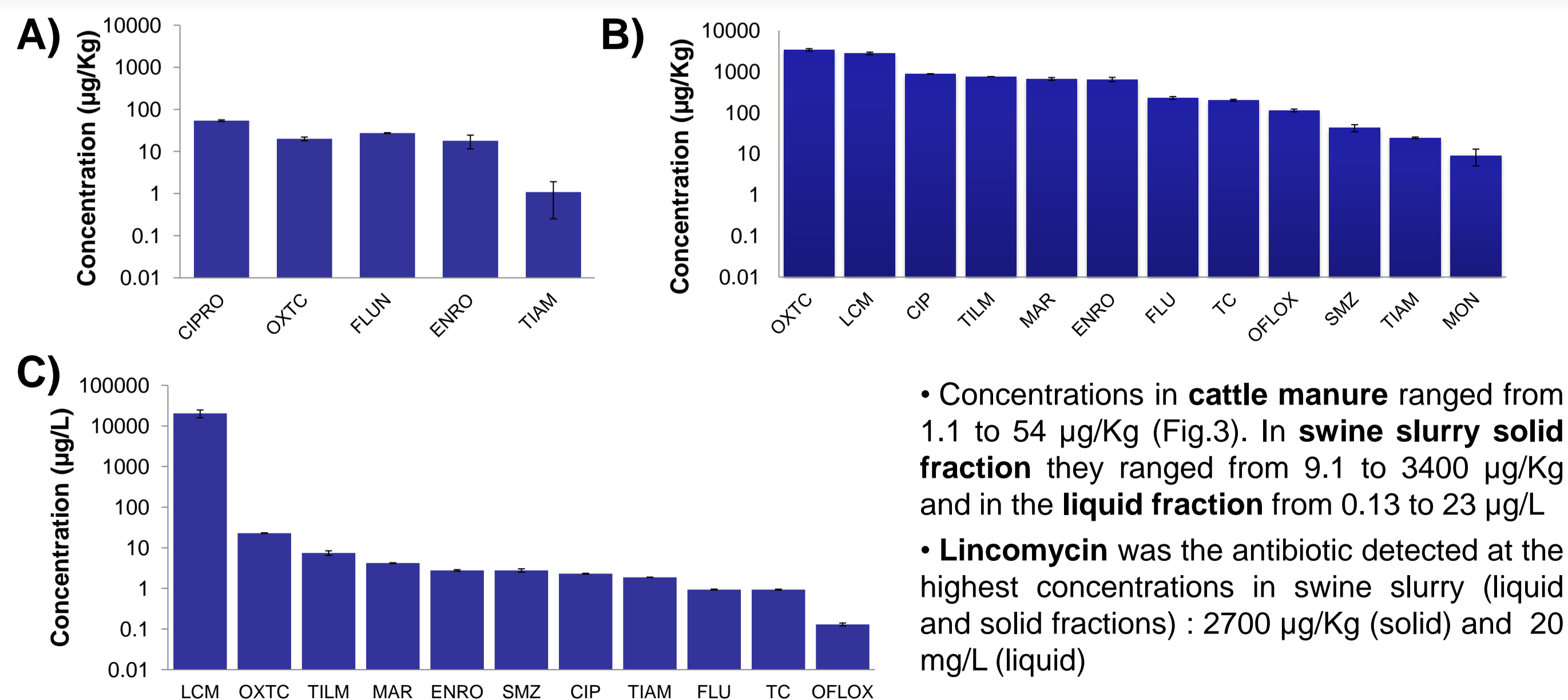
ULTRASONIC EXTRACTION

Clean-up SPE
Citrate buffer/AcN (1:1, v/v)
SAX (500mg, 6cc) &
Oasis HLB (200mg, 6cc)



Solid manure: dairy cattle manure and swine slurry solid fraction

6. RESULTS: PHARMACEUTICALS IN FERTILIZER



7. CONCLUSIONS

Preliminary results show that a wider range of veterinary pharmaceuticals were detected in swine slurry compared to cattle manure amended soils. Following the spread of fertilizers in the fields, most veterinary antibiotic residues were distributed at different soil depths (e.g. enrofloxacin, ofloxacin, flubendazole, salicylic acid and monensin), indicating their liability to leach to groundwater bodies. Other substances, such as oxytetracycline, tiamulin and flunixin, tend to accumulate in the surface layers of soil. These results will be complemented with another field experiment where separate solid and liquid fractions of swine slurry are used as soil amendment.

5. RESULTS: PHARMACEUTICALS IN AMENDED SOILS

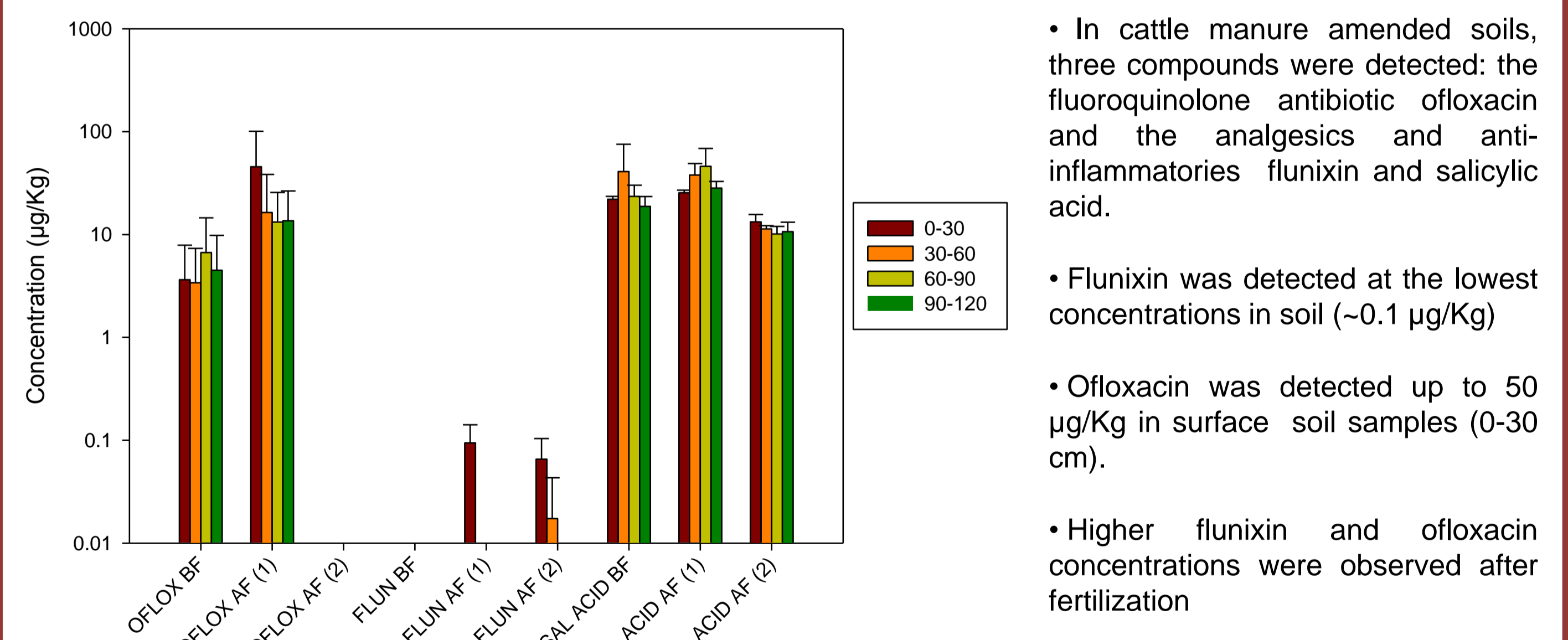


Figure 1. Pharmaceuticals detected in plots fertilized with dairy cattle manure (BF: before fertilization; AF (1): samples collected 2 months after fertilization; AF(2): 7 months after fertilization). Concentrations expressed in logarithmic scale

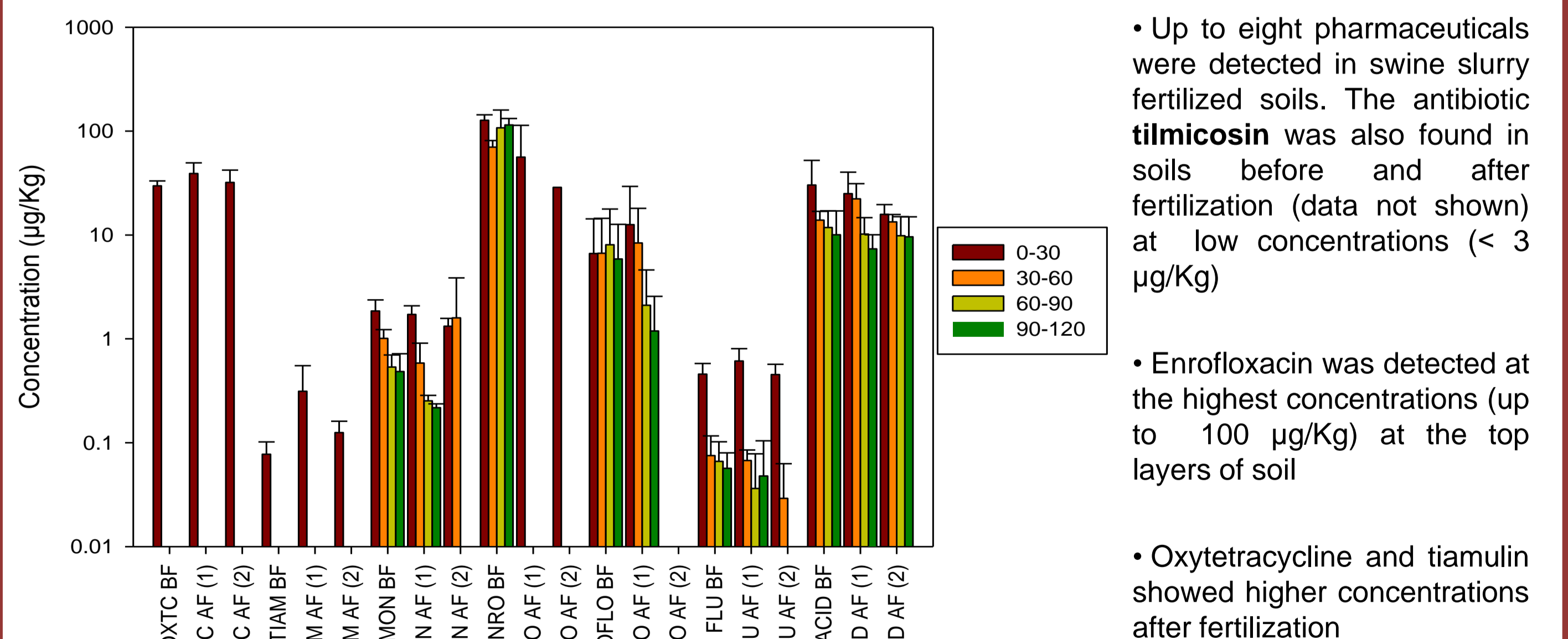


Figure 2. Pharmaceuticals detected in plots fertilized with swine slurry (BF: before fertilization; AF (1): samples collected 2 months after fertilization; AF(2): 7 months after fertilization). Concentrations expressed in logarithmic scale

REFERENCES

- [1] Rahube T.O. et al. 2014. Appl. Environ. Microbiol. 80:6898-6907
- [2] Frey S.K. et al. 2015. Sci. Total Environ. 532:138-153
- [3] Sollicc M. et al. 2016. Sci. Total Environ. 543:524-535

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