Report for 2003IA37B: Fate of Veterinary Antibiotics in Manure Lagoons

- Articles in Refereed Scientific Journals:
    - Kolz, A.C., S.K. Ong, and T.B. Moorman. 2005. Sorption of Tylosin onto Swine Manure, Chemosphere, 60(2):284-289.
    - Kolz, A.C., T.B. Moorman, S.K. Ong, K.D. Scoggin, and E.A. Douglas. 2005. Degradation and metabolite production of tylosin in anaerobic and aerobic swine manure slurries, Water Environment Research, 77(1):49-56.

Report Follows

Fate of Veterinary Antibiotics in Manure Lagoons

Say Kee Ong, Ph.D. and Tom Moorman, Ph.D.

Problem and Research Objectives

Antibiotic residues and increased numbers of antibiotic-resistant bacteria have been reported near confined animal feeding operations (CAFOs) and in agricultural watersheds. A major route for entry of veterinary pharmaceuticals into watersheds is through land application of animal biosolids and spills of animal waste at facilities using these drugs. Swine CAFOs often use antibiotics for therapeutic or growth-promoting purposes. Manure generated at CAFOs, and containing excreted residues, is commonly stored in earthen lagoons for several months before land application. Incomplete degradation of pharmaceuticals in vivo and during manure storage, before biosolids are land-applied, could be a contributing form to the presence of these drugs in waterways.

The fate of these chemicals is of environmental importance as it has been shown that highly resistant pathogens may develop within the manure management facilities and that these chemicals may interfere with the endocrine system of various aquatic species. Currently, research on the fate of these compounds in the manure management system and in the environment is very limited. The objectives of this study are to investigate the fate of two common antibiotics, tylosin and sulfamethazine, used in the swine industry. The focus will be on the sorption and degradation of these antibiotics in manure lagoons under anaerobic and aerobic conditions.

Methodology

The proposed research consists of analytical methods development, batch sorption studies, and batch degradation studies. The antibiotics to be tested are tylosin and sulfamethazine, two major antibiotics used in the swine industry. Manure will be obtained from various manure lagoons and characterized for pH, total organic carbon, total dissolved solids, and ammonia.

A key aspect of studying antibiotics in the environment is the ability to analyze the antibiotics in various media and in low concentrations. Different solvents for extraction of antibiotics from both liquid and sludge from waste manure were tested. The antibiotics were analyzed using liquid chromatograph and liquid chromatograph-mass spectroscopy (LC-MS).

Batch sorption experiments were conducted according to the American Society of Testing and Materials E1195-01 (ASTM, 2002). Sodium azide was added to each vial to inhibit microbial degradation. Anaerobic degradation studies were conducted using a series of 120 mL serum bottles containing sludge from manure lagoons. The serum bottles were spiked with a given amount of antibiotic and the vials were purged with nitrogen to ensure dissolved oxygen was removed. At different times, vials were sacrificed and the concentrations of the antibiotics in both liquid and solid phases

analyzed. The parent compound remaining and metabolites, if any, were determined using LC-MS. Aerobic degradation experiments will be similarly conducted.

Work for tylosin has been completed, while research on sulfamethazine is ongoing.

Principal Findings and Significance

Tylosin disappearance followed a biphasic pattern where rapid initial loss was followed by a slow removal phase. The 90% disappearance times for tylosin, relomycin (tylosin D), and desmycosin (tylosin B) in anaerobically incubated slurries were 30 to 130 hours. Aerating the slurries reduced the 90% disappearance times to between 12 and 26 hours. Biodegradation and abiotic degradation occurred, but strong sorption to slurry solids was probably the primary mechanism of tylosin disappearance. Dihydrodesmycosin and an unknown degradate with molecular mass of m/z 934.5 were detected. Residual tylosin remained in slurry after eight months of incubation, indicating that degradation in lagoons is incomplete and that residues will enter agricultural fields.

Sorption of tylosin was conducted on manure solids (<2 mm) and colloidal materials (<1.2 lm) collected from open (OL) and covered (CL) anaerobic swine manure lagoons. The aqueous concentration of tylosin in the sorption studies bracket the levels expected in lagoons, between 1 mgL^{-1} and 30 mgL^{-1}. Sorption isotherms were found to be slightly non-linear for 2 mm solids, with Freundlich distribution coefficients (K_f) of 39.4 with n = 1.32 for CL slurry and 99.5 with n = 1.02 for OL. These values are comparable to those reported for loam soils, but higher than those reported for sandy or clay soils and lower than those reported for fresh manure. Normalization of K_d to the organic carbon content of the solids gave K_{oc} values of 570 L/kg^{-1} and 818 L/kg^{-1} for CL and OL solids, respectively. The K_d and K_f values were not significantly different between colloids and 2 mm solids in OL slurry but were significantly different in CL due to the non-linearity of the colloid isotherm. Based on the K_d values obtained and comparing the K_d values of other antibiotics, tylosin is strongly sorbed to manure and would be more mobile than tetracyclines but less mobile than sulfonamides, olaquindox, and chloramphenicol. However, tylosin mobility may be facilitated through transport with colloidal manure materials.

We are currently conducting sorption and degradation studies on sulfamethazine in manure and in soils.