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Research article

Effects of solid-liquid separation and storage on monensin attenuation in dairy waste management systems

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ABSTRACT

Environmental release of veterinary pharmaceuticals has been of regulatory concern for more than a decade. Monensin is a feed additive antibiotic that is prevalent throughout the dairy industry and is excreted in dairy waste. This study investigates the potential of dairy waste management practices to alter the amount of monensin available for release into the environment. Analysis of wastewater and groundwater from two dairy farms in California consistently concluded that monensin is most present in lagoon water and groundwater downgradient of lagoons. Since the lagoons represent a direct source of monensin to groundwater, the effect of waste management, by mechanical screen separation and lagoon aeration, on aqueous monensin concentration was investigated through construction of lagoon microcosms. The results indicate that monensin attenuation is not improved by increased solid-liquid separation prior to storage in lagoons, as monensin is rapidly desorbed after dilution with water. Monensin is also shown to be easily degraded in lagoon microcosms receiving aeration, but is relatively stable and available for leaching under typical anaerobic lagoon conditions.

1. Introduction

Monensin is a polyether ionophore antibiotic used exclusively for veterinary applications, specifically in poultry and cattle to treat coccidiosis and enhance growth (Fig. 1) (Russell and Houlihan, 2003). Monensin is also used in lactating dairy cattle to increase milk productivity, making monensin one of the few feed additives approved for lactating cattle in the United States (US FDA, 2004; NMFP, 2015). While standards on antimicrobial use have typically been more stringent in other countries than the US, monensin continues to be used as a growth promoting feed additive worldwide (Maron et al., 2013). Due to its high toxicity profile and the largely unknown fate in the environment relative to other antimicrobials, monensin is 1 of 13 veterinary active ingredients listed as high priority for detailed risk assessment out of 83 investigated (Capleton et al., 2006).

The potential for high monensin use in dairies raises concerns of significant monensin loading into the environment through manure management. Dairy manure, which contains excreted antibiotics and metabolites, is often collected by flushing freestalls. Solids are then separated from diluted manure through a variety of techniques. Solids are commonly separated by settling basin sedimentation, which uses gravity to remove the solids. Mechanical screening of manure is another common technique, often followed by further separation by settling. After solid-liquid separation, the liquid fraction is stored in a lagoon and often applied to forage crop fields as a soil amendment. This practice is concerning in California, where dairies are one of the state's most prevalent concentrated animal feeding operation (CAFO) industries (NASS, 2014). Further, most of California's dairies are located in the San Joaquin Valley (CDFA, 2009), a topographically flat region of predominantly alluvial and fluvial unconsolidated aquifers with some areas of shallow water tables, making them particularly vulnerable to contamination.

There are two potential pathways for monensin to reach groundwater: by direct leaching from the production area and through the waste management system, such as lagoon storage and manure application to fields. Previous research in a California dairy system has shown that contamination of groundwater via waste storage is considerable, while liquid manure application to irrigated

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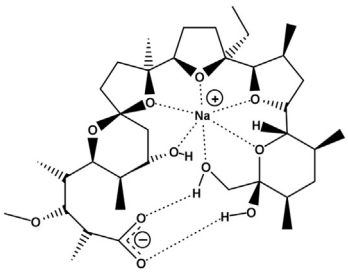

Fig. 1. Structure of monensin, an ionophore antibiotic administered to dairy cattle as a feed additive. Structure created with MarvinSketch, Marvin 15.4.20.0, 2015, ChemAxon (http://www.chemaxon.com).

crops is not a significant source of monensin to groundwater (Watanabe et al., 2008; Hafner et al., 2016). Once released into the soil, processes for reduction of available monensin appear to be rapid and include degradation and sorption to soil (Carlson and Mabury, 2006; Davis et al., 2006; Sassman and Lee, 2007; Song et al., 2010).

Reducing monensin concentrations in the lagoon will reduce a major groundwater contamination pathway. The primary goal of this study is to investigate the effects that waste management practices have on final aqueous monensin concentrations in manure slurries. In order to fully assess monensin distribution in groundwater beneath dairies, additional samples were collected from the two dairy systems monitored in Watanabe et al. (2008). Groundwater, flush lane, and lagoon water samples were collected during four sampling events over two years. Laboratory manure microcosms were then constructed to determine the potential effect of waste management practices on monensin attenuation. Given that monensin is nearly exclusively excreted in the feces (Donoho et al., 1978), the effect of solid-liquid separation by screening (2 mm) on final lagoon monensin concentrations is investigated. Short-term microcosms (<48 h) were constructed to evaluate the effect that settling basin detention time has on final monensin concentration. Long-term microcosms were used to determine these same effects on a time scale more relevant to lagoon storage (weeks), and included a treatment for the effects of aeration on monensin transformation.

2. Materials and methods

2.1. Dairy study sites

Two dairy farms in the San Joaquin Valley of California were studied. Detailed descriptions of the site hydrogeology are given in Harter et al. (2002). Groundwater levels in the study area range from 2 to 5 m, with a well-drained sandy loam as the dominant surface soil texture. Monitoring wells (MWs) are 5 cm in diameter and 7–10 m in depth, with screening below 2 or 3 m. Wells capture recent recharge from irrigated fields, lagoon leachate, or corral recharge. Dairy maps and monitoring well locations are provided in Supporting Figure S1.

On both dairies, concrete-lined freestalls are flushed with lagoon water three to four times per day to collect waste. Flushed manure then undergoes solid-liquid separation before collection of the liquid fraction into a lagoon. Dairy I separates solids by mechanical screen and settling basins, while Dairy II utilizes only settling basins.

Groundwater, flush lane, and lagoon water samples were collected during four sampling campaigns in the fall (Oct. 17–18, 2006), spring (April 26 and May 22, 2007), summer (Sept.4–5, 2007) and winter (Jan. 2–3, 2008) from Dairy I and Dairy II. Due to restricted accessibility, not all wells were sampled at each sampling date. Methods of groundwater, flush lane, and lagoon water sampling are detailed in Watanabe et al. (2008).

2.2. Manure collection

Manure for laboratory experiments was collected from the University of California Dairy Teaching and Research Facility in Davis, CA. Treatment samples were obtained from milking cows consuming monensin at a dose of 6 mg lb^{-1} dry matter. Manure without monensin was also collected from an organic dairy farm in Orland, CA, for use as a negative control. The feed rations for both dairies are described in the Supporting Material. Fresh manure was shoveled into buckets lined with plastic bags and stored at 4 °C. Treatment dairy manure was collected directly from the bedding while the organic control manure was collected at the end of the flush lane. Total solids (TS) was determined by taking three representative samples (~25 g) from both the treatment and control manure and oven drying at 105 °C, cooling in a desiccator, and weighing remaining solids. The process was repeated until samples differed by < 50 mg over one hour of drying (Eaton et al., 1995). The bedding material in the treatment manure (sand) was removed from final samples prior to determining TS. Fecal samples were then diluted with 18.2 MΩ·cm water to reach an initial TS of 3% for use in all experiments. Diluted manure for screened treatments was passed through a 2 mm sieve with gentle stirring and light pressing of solids, and TS was determined for the final screened suspension. Due to the volume of diluted manure required for long-term lagoon microcosms with and without aeration, two separate dilutions were made and distributed throughout the time points.

2.3. Chemical analysis

Chemical preparation for dairy site samples is described in Watanabe et al. (2008). The analysis of monensin and nigericin by liquid-chromatography mass-spectrometry for the microcosm study is detailed in the Supplemental Material.

Quality control for dairy site sample analysis is described in Watanabe et al. (2008). Quality control for microcosm experiments was evaluated by performing spike-recovery experiments on manure extracts. Diluted manure was screened to <2 mm (2.3% TS organic control, 1.2% TS treatment), centrifuged in 40 mL fluorinated ethylene propylene (FEP) Nalgene Oak Ridge tubes (20 min at 20,500 × *g*), and filtered to 0.3 μm using stacked glass fiber filters (1.3 μm, 0.7 μm, 0.3 μm) on glass frit filtration assemblies. Extracts (25 mL) were fortified to 6.6 μg L^{-1} with monensin and 6.4 μg L^{-1} with nigericin. Nigericin is an ionophore not used by the dairy industry, and was therefore appropriate to use as a surrogate to evaluate analyte loss. Samples were mixed for 15 min at 130 rpm on a platform shaker (New Brunswick Scientific, Edison, NJ) before solid-phase extraction (SPE) with Waters Oasis hydrophilic-lipophilic balance (HLB) cartridges (3 mL, 60 mg). Cartridges were conditioned with 3 mL methanol and equilibrated with 6 mL water before sample addition. Cartridges were then washed with 6 mL water before drying and eluting with 2 mL methanol. Recovery samples for each treatment were prepared in triplicate, with triplicate unfortified samples. Monensin recovery from filtered organic manure was 87.6% (4.3% CV), and recovery from filtered treatment manure was 87.1% (9.3% CV). Nigericin recovery from filtered treatment manure averaged 82.2% (4.5% CV).

2.4. Extractable monensin from manure

The effect of the manure quantity on monensin concentration for both screened and unscreened manure was evaluated by three manure dilutions. Wet manure (90, 110, or 130 g) was weighed into