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Estrogenic activity and nutrient losses in surface runoff after winter manure application to small watersheds

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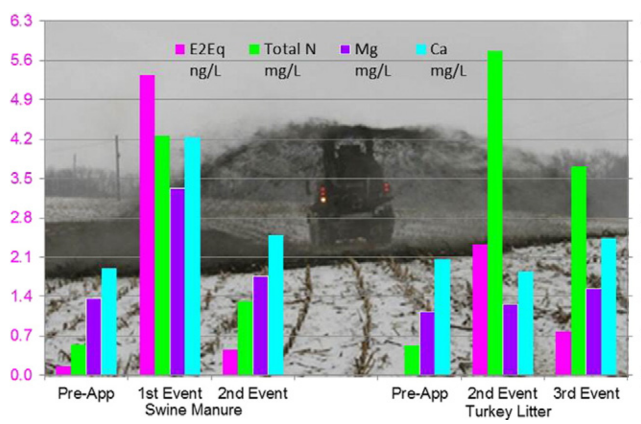
HIGHLIGHTS

- Runoff evaluated post-application of swine or turkey waste to frozen fields.
- Highest estrogenic activity below the Lowest Observable Effect Concentration for estradiol
- Most runoff below Predicted No Effect Concentration
- With high enough nutrient application, a correlation may exist for Ca^{2+}, Mg^{2+}, and K^{+}, and E_2Eq.
- No runoff was obtained from foraged plots post-waste application.

GRAPHICAL ABSTRACT



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ABSTRACT

Confined Animal Feeding Operations generate large amounts of wastes that are land-applied to provide nutrients for crop production and return organic matter to the soil. Production practices and storage limitations often necessitate that wastes be applied to frozen and snow-covered soil. Use of application setbacks have reduced concerns related to nutrient losses in surface runoff from manure, but the estrogenic activity of runoff under these conditions has not been evaluated. Therefore, we measured and sampled surface runoff when manure was applied in the winter at a rate to meet crop N needs and measured estradiol equivalents (E_2Eqs) using E-Screen. In year one, six small watersheds used to produce corn were evaluated, treatments: 2 no-manure controls, 2 liquid swine manure with 30-m setbacks, and 2 turkey litter with 30-m setbacks. In addition, beef manure was applied to six frozen plots of forage. For years 2 and 3, applications were repeated on the swine manure watersheds and one control watershed. E_2Eqs and nutrient concentrations generally peaked in the first runoff event after application. The highest measured E_2Eq (5.6 ng L^{-1}) was in the first event after swine manure application and was less than the 8.9 ng L^{-1} Lowest Observable Effect Concentration (LOEC) for aquatic species and well below the concentrations measured in other studies using ELISAs to measure hormone concentrations. No runoff occurred from plots planted with forage, indicating low risk for environmental impact, and therefore

Abbreviations: E_2, estradiol; E_2Eq, estradiol equivalents; CW, Coshocton Wheel; LC–MS2, liquid-chromatography tandem-mass spectrometry; LOEC, lowest observable effect concentration; MWTP, municipal wastewater treatment plant; NRCS, National Resources Conservation Service; npH_2O, nanopure H_2O; PNEC, predicted no effect concentration; SPE, solid phase extraction; TOC, total organic carbon; Total N, total nitrogen; WS, watershed.

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plots were discontinued from study. In years 2 and 3, estrogenic activity never exceeded the Predicted No Effect Concentrations for E_2 of 2 ng L^{-1}. When post-application runoff contained high estrogenic activity, strong correlations (R^2 0.86 to 0.96) of E_2Eq to Ca^{2+}, Mg^{2+}, and K^+ concentrations were observed, indicating under some condition these cations might be useful surrogates for E_2Eq measurements.

1. Introduction

The impact of row crop production on surface water quality has been investigated for decades, mainly in terms of soil and nutrient losses as affected by crop management practices and climatic factors (Carpenter et al., 1998). Among the practices of concern is the land application of animal wastes. While these biosolids are an important source of nutrients for crop growth and organic matter for maintaining or increasing soil carbon levels, they can also contribute to degradation of surface water quality. In particular, large confined animal feeding operations (CAFOs) produce large quantities of manure, often posing a disposal issue, with potential risks to the environment. Additionally, winter application of manure has come under increased scrutiny due to a heightened risk of nutrient and organic matter losses in surface runoff when applied to frozen or snow-covered soils (Srinivasan et al., 2006). As a result, a number of states in the U.S. have adopted various restrictions and guidelines to minimize the risks associated with winter manure application, but these are often based on anecdotal information and have not yet been extensively tested.

In their investigation of winter time application of turkey and swine manure to small watersheds following Ohio NRCS recommendations, Owens et al. (2011) noted that the highest concentrations of N (nitrogen) and P (phosphorus) in surface runoff generally occurred in the first few events after application (typically predominated by snowmelt with minimal precipitation). In the 3-yr study, losses of these nutrients were elevated compared to no-manure controls, but were within acceptable levels, though P was of more concern than N. Additionally, their data suggested that filter strips (setbacks) reduced nutrient concentrations and losses. Use of grassed buffer strips has been investigated for its efficacy in reducing runoff volume, and nutrient and contaminant concentrations (Nichols et al., 1998; Milan et al., 2013). State regulatory guidelines often mandate vegetative (grassed) buffer strips between crop land or feedlots and waterways, to minimize the nutrient-rich runoff reaching surface waters. While these buffer strips appear to reduce contamination of surface waters, they are not 100% effective, particularly in the case of high rainfall events (Watts and Torbert, 2009 JEQ, review — Dorizo et al., 2006). For this reason, we included evaluation of runoff from manure applied to frozen grassed plots.

Recently, losses of naturally occurring hormones present in manure have become an additional issue of concern. For example, Alvarez et al. (2013) noted that hormone levels and estrogenic activity in stream water sampled from watersheds in 12 US states sometimes rose following manure application. Our ability to measure hormones in surface waters and assess their environmental effects has been hampered by limitations in the available methodology. Radio-immunoassays and enzyme-linked immunoassays (ELISAs) can result in spurious results due to antibody cross-reactivity and susceptibility to non-specific binding, whereas liquid-chromatography tandem-mass spectrometry (LC–MS^2) can be affected by interfering substances (matrix effects). Use of deuterated internal standards, as well as standard addition, can in some cases, overcome matrix effects seen in LC–MS^2. Likewise, our understanding of the environmental impact of hormones, such as estrogens, is incomplete. In their 21-day in vivo study, Jorgenson et al. (2015) noted differential species sensitivity to 30 ng/L estradiol (E_2) with survival of 47% for Rio Grande Silvery minnow, 61% for Fathead minnow, and 100% for Bluegill sunfish. Bioassays such as E-Screen (Soto et al., 1995) provide information about the potential biological response to complex mixtures of chemicals released into the environment from animal waste.

Thus, while hormonal release into surface waters as a result of runoff following manure application is an issue of concern, there is little information on the specific land and manure management factors that can contribute to this problem. Therefore, our specific objectives were to expand on the winter manure application watershed study of Owens et al. (2011) by determining 1) the estrogenic activity of runoff from agricultural lands post-application of various types of animal waste to frozen ground and comparing to the No Observable Effect Concentration (NOEC) or Lowest Observable Effect Concentration (LOEC) of E_2 for aquatic species, 2) the effect of vegetation on estrogenic activity of runoff, and 3) potential correlations between measured estrogenic activity and various, more easily measured nutrients/ions that might be used as an indicator for the presence of estrogenic activity. The E-Screen bioassay (based on proliferation of the non-transfected human mammary epithelial cell line MCF-7 BOS) was used to quantitate the estrogenic activity, as this assay reflects an integrated response to estrogens, not just receptor binding or transcriptional activation. Data are presented on a concentration basis (necessary for determining potential biological impacts), a flow-weighted basis (to compensate for differential runoff volumes), and as a % of mass applied (to assess if losses are proportional to mass applied).

2. Materials and methods

2.1. Watershed treatments

The study was conducted from 2009 to 2011 at the former North Appalachian Experimental Watershed near Coshocton, Ohio (40°22′ N and 81°48′ W) using six gaged small watersheds in 2009 and three watersheds (WS) in 2010 and 2011. The watersheds were of similar size (0.55 to 0.79 ha) with variable slopes, from 2°–12° within one watershed up to 6°–18° in others (Table 1, Fig. 1). The predominant soil types were Coshocton silt loam, Keene silt loam, and Rayne silt loam. Soil descriptions can be found in the footnote of Table 1, with greater detail on WS and soil characteristics available in Kelley et al. (1975). Each WS was planted using no-till corn (Zea mays) each year except on the WS designated as Swine 2, which was disked to 10–15 cm in the spring prior to corn planting and cultivated between the rows twice each year during the growing season. Swine manure consisted of slurry obtained from the manure pit of a wean to market feeding operation. Turkey litter was a combination of wood shavings and excreta from breeder hens that had been stored outside for ≤2 months prior to application. (See SI for details of application and sampling). Manure or litter was applied in one day, in January or February of each year, dependent on weather conditions, leaving a 30 m application setback above the outlet of each WS. Manure application rate was adjusted based on pre-application analysis to achieve a target N level of 180 kg N ha^{-1}, sufficient for a 8.8 Mg ha^{-1} corn grain yield (Vitosh et al., 1995). Urea was applied to the control WSs each year at the time of planting, as well as to the setback areas, at a rate of 180 kg N ha^{-1}. In 2009, two WSs per treatment were used as follows: control (no manure), turkey litter, and swine manure slurry, hereafter referred to as manure (Table 1). In 2010 and 2011, three WSs were sampled, two WSs that had received swine manure in 2009 received additional swine manure and one no-manure control WS (Control 2).

Runoff, which only occurred when there was significant snow melt or precipitation, was sampled on an event basis using H-flumes equipped with water stage recorders for determining surface runoff volumes and timing (Brakensiek et al., 1979). The flow-proportional