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Veterinary antibiotics and hormones in water from application of pig slurry to soil

Adilson Pinheiro*, Renata M. Rosa Albano, Thiago Caique Alves, Vander Kaufmann, Marcos Rival da Silva

Fundação Universidade Regional de Blumenau, rua São Paulo, 3250, 89030-000 Blumenau, SC, Brazil

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ABSTRACT

Although the use of liquid waste from piggeries as agricultural fertilizer is common practice, problems arise from contamination of water by chemical compounds. This paper reports on the presence of three hormones and six veterinary antibiotics in water of the surface runoff and drainage flow from volumetric lysimeters to which pig slurry was applied, and which had been planted with cereals, horticultural crops and permanent pasture. High-efficiency liquid chromatography was used for chemical analysis of soil water following two applications of liquid pig slurry. Concentrations of the hormones and antibiotics found were between 4.6 and 1350.8 ng L^{-1}, and were detectable up to the ninth day after the second application of manure. The presence of different crops did not influence the occurrence of the analytes.

1. Introduction

Agriculture and animal production are important areas of the Brazilian economy, making substantial contributions to the national income. As with any such activity of economic importance, residual wastes are produced that have the potential to degrade the natural environment. Animal waste is a major source of such residues, and since it is rich in both organic material and plant nutrients, it is frequently applied to soil as fertilizer.

Casey et al. (2003) confirm that, when applied to fields, waste liquids from pig production may contain veterinary antibiotics and hormones that are produced naturally and excreted by animals. These compounds are also used in livestock rearing to promote growth and efficient food conversion, and for treatment and prevention of disease. Many of the compounds are not completely metabolized within the animal and their residues have been detected in animal waste, soil samples, surface runoff and groundwater. Thus the practice of putting slurry on agricultural land can compromise the quality of surface and underground water bodies and have serious repercussions for public health. Indeed, as well as hormones and antibiotics, other organic contaminants such as bacteria, viruses and protozoa can reach ground waters by the same mechanism, i.e. leaching through the soil (Lugoli et al., 2011).

The rate of excretion of veterinary antibiotics depends on the substances administered, the species of animal and the time of treatment, and in some cases 90% of the dose given may be eliminated in non-metabolized form or as active metabolites. Absorption of such substances by the organism is generally incomplete (Kemper, 2008; Bao et al., 2009).

Sorption of antibiotics and hormones in soil is influenced by combinations of the specific characteristics of the substances and environmental factors which include soil organic matter, clay content, ion-exchange capacity and pH (Drillia et al., 2005). Other factors include the photo-stability of the pharmaceutical/hormone, its rate of decay, leaching capacity, and physico-chemical properties (Diaz-Cruz et al., 2003).

Bioaccumulation of antibiotics in plants and organisms results from their dispersion through soil (Pereira et al., 2012). The development of resistant bacteria is another matter of concern (Chander et al., 2005). In the case of hormones, these may damage an endocrinal organ directly; or alter its function; or interact with a hormonal receptor, or change the metabolism of a hormone in an endocrinal organ (Bila and Dezotti, 2007).

In Brazil, the use of pig slurry as soil fertilizer is regulated by a legal framework set up by the Ministry of Agriculture (Guideline SDA/MAPA 25/2009), with standards determined by levels of soil nutrients and micronutrients. There are also standards defined for the disposal of bio-solids from sanitary-waste treatment stations, issued by the Ministry for the Natural Environment (Resolution CONAMA No. 375/2006). Here, levels of application are established in terms of soil nutrients, inorganic substances, microbiological and pathogenic indicators. Brazilian legislation follows international standards in which endocrine disruptors, pharmaceuticals,

* Corresponding author. Tel.: +55 44 32216078; fax: +55 47 32216001.
E-mail addresses: pinheiro@furb.br (A. Pinheiro), re_zinhaaa@hotmail.com (R.M. Rosa Albano), thiago.caique@gmail.com (T.C. Alves), ambitec.amb@gmail.com (V. Kaufmann), rivail@furb.br (M.R. da Silva).

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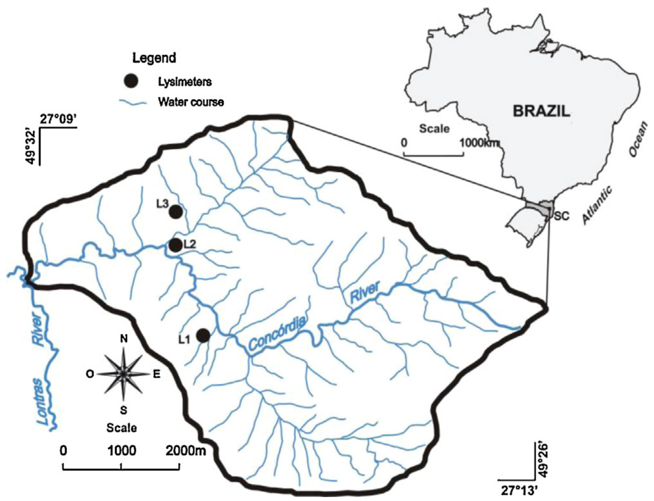


Fig. 1. Map of the south of Brazil showing of the Concórdiariver basin and locations of the lysimeters.

hormones, antibiotics, anti-inflammatory agents and personal care products, present in municipal waste and waste from animal production, are not regulated and monitored by maximum contaminant levels defined in the Safe Drinking Water Act (USEPA, 2004).

Against this background, the objective of the work reported here was to evaluate the occurrence of three hormones (estrone, estradiol and 17α-ethinylestradiol) and six veterinary antibiotics (chlortetracycline, doxicycline, oxytetracycline, tetracycline, sulfadimidine and toltrazuril) in surface runoff and soil water, following the application of pig slurry to farmed soil in the drainage basin of the Concórdia river at Lontras, in the Brazilian State of Santa Catarina.

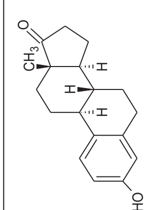
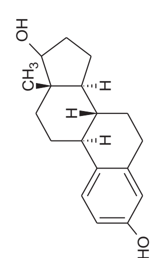
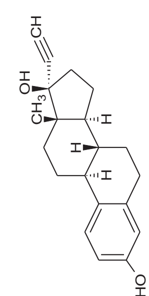
2. Materials and methods

The work was developed using experimental equipment in the form of three drainage lysimeters installed in a river basin near the township of Lontras in Santa Catarina, in the south of Brazil (latitude 27° 11′ 17.0″ S, longitude 49° 29′ 40.1″ W). Land-use in the basin consists of native forest, re-afforestion, pasture, maize (corn) fields, tobacco, onions and manioc. The lysimeters used for the study had the following crops: lysimeter 1 (L1), cereal cultivation; lysimeter 2 (L2), horticultural crops, and Lysimeter 3 (L3), perennial pasture. The soils are classified as Cambisols. Fig. 1 shows the lysimeter locations within the basin of the Concórdiariver.

The lysimeters were constructed from acrylic plates which hold an undisturbed sample with base 1 m^2 and depth 1 m. Each consists of two acrylic boxes, the first containing the soil sample and the second forming the lysimeter base. More details of the lysimeter construction are given by Oliveira et al. (2010). Tubing was installed in the upper part of each lysimeter to allow surface runoff. Within the lysimeter base, more tubing was inserted to collect drainage water percolating through the soil profile.

Rainfall was measured using a Davis® recording raingauge with Novus® datalogger, sited near the lysimeters. Rainfall was recorded at 15 min intervals, and all data were recorded over the period 1 September 2011–29 February 2012. Two pig-slurry applications were given during this period at rates of 50 $m^3\ ha^{-1}$; the first application was on 14 September 2011 when the L1 soil was lying fallow, L2 was under an onion crop and L3 under permanent pasture. The second slurry application was given on 8 November 2011, when L1 was planted with maize, L2 was fallow and L3 again permanent pasture. Samples were collected immediately after rain, on ten

Table 1
Physics-chemical properties of the hormones and antibiotics studied.

Analyte CAS number	MM	W_s (mg/L at 20 °C)	Log Kd	pKa	$t_{1/2}$ (d)	Molecular structure	References
E1 53-16-7	270.4	12.4	2.44–2.72	10.3–10.8	2 to 3		Ying et al. (2002)
E2 50-28-2	272.4	13.3	2.68–2.83	10.5–10.7	0.2 to 9		Chen et al. (2009) Combalbert and Raquet-Hernandez (2010) Ying et al. (2002)
EE2 57-63-6	296.4	4.8	2.65–2.86	10.4–10.7	4 to 6		Pan et al. (2009) Chen et al. (2009) Combalbert and Raquet-Hernandez (2010) Ying et al. (2002)