



DESALINATION

Desalination 226 (2008) 121–133

www.elsevier.com/locate/desal

# Occurrence of antibiotics and hormones in a major agricultural watershed

Osman A. Arikan<sup>a,b\*</sup>, Clifford Rice<sup>a</sup>, Eaton Codling<sup>a</sup>

<sup>a</sup>USDA-ARS, Environmental Management and Byproduct Utilization Laboratory, Beltsville, MD 20705, USA <sup>b</sup>Department of Environmental Engineering, Istanbul Technical University, Istanbul 34469, Turkey Tel. +90 212 285 37 87; Fax +90 212 285 37 81; email: arikan@itu.edu.tr

Received 18 December 2006; revised accepted 30 January 2007

#### Abstract

Antibiotics and hormones are considered emerging environmental microcontaminants because of their potential adverse effects on ecosystems and human health. Available information on the source of these emerging contaminants in surface waters is very limited. The objectives of this study were to determine the occurrence of antibiotics and hormones in an agricultural watershed and to determine the seasonal variability of these contaminants. Water samples were collected from 15 subwatershed stations and 7 stations on the major receiving river, Choptank, Maryland, USA, over four different seasons (April, June, September and December). Antibiotics (sulfathiazole, sulfamerazine, sulfamethizole, sulfamethazine, sulfachloropyridazine, sulfamethoxazole, sulfadimethoxine, tetracycline, oxytetracycline, chlortetracycline and doxycycline) and hormones (estriol, estradiol,  $17\alpha$ -ethynylestradiol, estrone, testosterone and progesterone) as well as arsenic which is used as feed additive were determined in these water samples. In addition, the same antibiotics were analyzed in one set of sediment samples. This study indicated that agriculture may act as a source of antibiotic residues in the aquatic environment.

Keywords: Antibiotic; Hormone; Arsenic; Watershed; Agriculture

#### 1. Introduction

A 2002 survey conducted by the National Agricultural Statistics Service (NASS) reported that there were 104 million cattle, 8.6 billion chickens, 60 million swine, and 275 million turkeys in the United States (US) [1]. The marked

intensification of animal production, particularly in the last 25 years, has resulted in increased water quality problems associated with the production and disposal of animal waste generated by these operations. Current livestock production involves the use of large amounts of different chemicals including antibiotics, hormones and metals (feed additives). These chemicals are considered emerging environmental microcontaminants because

Presented at the 10<sup>th</sup> IWA International Specialized Conference on Diffuse Pollution and Sustainable Basin Management, Istanbul, Turkey, 18–22 September 2006.

<sup>\*</sup>Corresponding author.

of their potential adverse affects on ecosystems and human health. Manure from animals is either stockpiled or immediately applied to farmland as fertilizer. When applied to a field, chemical residues in the manure can ultimately reach surface and ground water by runoff or leaching [2]. A variety of chemicals used for animal production have been detected in manure [3,4] and in surface waters [5–8].

Two commonly used classes of antibiotics for animal feeding operations are sulfonamides (SAs) and tetracyclines (TCs). While SAs are synthetic, TCs are naturally occurring or semisynthetic. Antibiotic residues in the environment are of considerable concern because of the potential development of antibiotic-resistant bacteria. Hormones can be considered endocrine disrupting chemicals (EDCs), and laboratory studies have shown that environmental levels of EDCs can affect fish [9] and birds [10] under laboratory conditions. Animal feed additives include trace metals and metalloids such as arsenic as well as antibiotics [11]. The arsenicals include roxarsone and arsanilic acid, which improve growth performance as well as improve bird pigmentation. Arsenic has become an important environmental concern due to its potential carcinogenic properties [12], and it is also considered the number-one toxin in the USEPA list of priority pollutants [13,14].

Although these emerging contaminants have been found in surface waters, it is not clear whether the sources are urban wastewater or animal feeding operations. Also, available information on the seasonal variation of these chemicals in surface waters is very limited. The objectives of this study were twofold: to determine the occurrence of animal feed additives in an agricultural watershed and to determine the seasonal variability of these contaminants. For this purpose antibiotics (sulfathiazole, sulfamerazine, sulfamethizole, sulfamethazine, sulfachloropyridazine, sulfamethoxazole, sulfadimethoxine, tetracycline, oxytetracycline, chlortetracycline

and doxycycline) and hormones (estriol, estradiol,  $17\alpha$ -ethynylestradiol, estrone, testosterone and progesterone), as well as feed additive metal of arsenic were determined in 15 subwatershed stations and 7 stations on the major receiving river, Choptank, Maryland, USA over four different seasons (April, June, September and December). In addition, the same antibiotics were analyzed in one set of sediment samples (only in June).

### 2. Materials and methods

#### 2.1. Study area

The Choptank River Watershed is located on the Delmarva Peninsula of the Chesapeake Bay, USA. It contains 62% agricultural land, 33% forest land, and 2% wetlands while 5% of land is developed. The Delmarva Peninsula is an agricultural center for the state of Maryland and it is dominated by the poultry industry. Approximately 35% of Maryland's cash farm income was from broilers in 2002. Maryland ranked 9th among the US states in pounds of broilers produced in 2002 with 1.4 billion pounds, and Maryland ranked 7th among the states in the number of broilers produced (585 Million in 2002). Chicken litter from the poultry houses is routinely recycled as a fertilizer on corn and soybean production fields in this region.

Parts of the Choptank River Watershed are identified as "impaired waters" under the Federal Clean Water Act. The Upper Choptank River may require preparation of Total Maximum Daily Loads (TMDLs) for nutrients and sediment, and the Choptank marine beach is impaired with respect to fecal coliform bacteria. Maryland Department of Natural Resource's results of monitoring of water quality and living resource habitat within the watershed since 1985 reveal increasing nitrate, chlorophyll *a*, total suspended solids, and decreasing Secchi depth values over time. Seasonally low oxygen levels are observed in the deeper estuarine portions of the Little Choptank River and Lower Choptank River.

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