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Excretion of manure-borne estrogens and androgens and their potential risk estimation in the Yangtze River Basin

Yanxia Li[1,*], *Shiying Gao*[1], *Shufang Liu*[1,2], *Bei Liu*[1], *Xuelian Zhang*[1], *Min Gao*[1], *Linjie Cheng*[1], *Boyang Hu*[1]

1. *State Key Laboratory of Water Environmental Simulation, School of Environment, Beijing Normal University, Beijing 100875, China*
2. *Yellow River Institute of Hydraulic Research, Yellow River Conservancy Commission, Zhengzhou 450003, China*

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ABSTRACT

The Yangtze River is the longest river in China, and the river basin spans one fifth of the area of the whole country. Based on statistical data, the excretion of manure-borne steroid hormones, including steroid estrogens (SEs) and steroid androgens (SAs), in 10 provinces of China within the region has been estimated. The potential environmental and ecological risk of manure-borne steroid estrogens to the surface water in this region was also assessed. The manure-borne SE and SA excretions in the 10 provinces and municipalities vary in the order: Sichuan > Hunan > Hubei > Yunnan > Jiangsu > Anhui > Jiangxi > Chongqing > Qinghai > Shanghai. The highest increase of manure-borne SEs (1434.3 kg) and SAs (408.5 kg) was found in Hunan and Hubei provinces, respectively, and the total excretion in 2013 was 65% more than 15 years earlier in these two provinces. However, the emissions in Anhui and Shanghai decreased in this 15 year period of time. Swine urine, chicken feces, cattle urine, and laying hen feces were considered the dominant sources of manure-borne E1, βE2, αE2, and SAs, respectively. Although Jiangsu province did not have the largest excretion of manure-borne SEs, it had the highest level of predicted 17β-estradiol equivalency (EEQ_s) value of 16.65 ng/L in surface water because of the limited surface water resources. According to the lowest observable effect level of 10 ng/L for 17β-estradiol, the manure-borne SEs in Jiangsu province might potentially pose ecological risk to its wild aquatic organisms.

Introduction

Natural steroid hormones, excreted from all vertebrates, are a series of typical endocrine-disrupting chemicals. Although the environmental concentrations of these contaminants are always low, it has been demonstrated that even at levels as low as nanograms-per-liter (ng/L), a wide variety of aquatic organisms exposed to these pollutants can be adversely affected (Kang et al., 2008; Leet et al., 2011; Örn et al., 2006; Shappell et al., 2010). Two important sources of hormone contaminants entering into the environment are municipal wastewater treatment plants (WWTP) and concentrated animal feeding operations (CAFOs). Recently, the livestock source of hormones has started drawing more and more attention, especially the natural steroids such as estrogens and androgenic, because of their large excretion from animal metabolism (Li et al., 2012b; Liu et al., 2012a). Some researchers have pointed out that the amount of hormones excreted by livestock was on

* *Corresponding author*. E-mail: liyxbnu@bnu.edu.cn (Yanxia Li).

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the same order of magnitude as that by humans, or in certain cases may be even exceed human contributions (Gadd et al., 2010; Hanselman et al., 2003; Johnson et al., 2006; Kolodziej et al., 2004; Lange et al., 2002; Liu et al., 2012b; Zheng et al., 2008). In the United States, the total daily emissions of 17β-estradiol and estrone from swine and dairy cattle were reported to range from 10 to 30 kg and from 20 to 80 kg, respectively, and the mass was 9 times greater than that from WWTP (Raman et al., 2004). In fact, various manure-borne hormones have been detected at high levels in the soils, streams, wells, and surface waters around animal farms or the places receiving animal wastes (Gall et al., 2011; Liu et al., 2012a). Hence, it is necessary to study the contamination issues associated with livestock wastes.

China is the largest animal farming country in the world. The annual animal feces and urine production were estimated to amount to 4 billion tons (Zhang et al., 2011). However, the Chinese animal farming industry is still at a poor management level compared to that in developed countries. Unlike domestic wastewater, there are no specific treatment requirements in China for livestock farm wastes before discharge (Li et al., 2012b), and only 20% of animal farms have treatment facilities like lagoons or sedimentation tanks (Liu et al., 2012a). Direct discharge into streams and application of livestock wastes on agricultural lands are common practices in China (Li et al., 2012b; Liu et al., 2012a). Once the wastes enter water or soil without thorough treatment, manure-borne estrogens will expand their contamination range, posing a serious threat to surrounding groundwater or surface waters (Arnon et al., 2008; Lee et al., 2007). Unfortunately, many previous studies and management policies are focused only on the manure-borne N and P (Khan et al., 2008; Li et al., 2007), and the few studies that investigated the occurrence and fate of steroids in swine or dairy wastes were limited to animal farms and their surrounding environments (Li et al., 2012a, 2012b; Liu et al., 2012a, 2012b). For example, Liu et al. (2011) observed up to 412 ng/g of 17β-estradiol (17β-E2) from dairy fecal matter or 20,700 ng/L of androsterone in farm wastewater; some steroids appeared in the aqueous phase, particle phase and sediment of the streams receiving farm effluents, and the concentrations were much higher than the pollutants in water receiving wastewater from WWTPs (Liu et al., 2011, 2012a, 2012b). Even though these results were from limited studies on point sources, they still help to understand the excretion and risk of animal manure-borne steroid hormones.

The Yangtze River, which is the longest river in China and the third longest in the world, plays a vital role in China's economic development and ecological environmental conservation. The drainage basin of the river accounts for one fifth of Chinese land area and the river flows through 11 provinces and municipalities. In recent years, more and more issues related to animal farming pollution were reported, but few concerning manure-borne hormones. The aim of this study is to estimate the excretion of manure-borne steroid hormones in 10 provinces of the Yangtze River basin (with the exception of the Tibet autonomous region) and assess the potential environmental and ecological risks from manure-borne hormones in the study region using Chinese statistical data and reported data in the literature.

1. Materials and methods

1.1. Study region

The Yangtze River is about 6380 km long and has water resource quantity of 9.5×10^{11} m^3/year, with a watershed of over 1.8 million km^2. The main stream flows through 11 provinces and municipalities. The Tibet autonomous region, Qinghai, Sichuan, Yunnan provinces, and Chongqing municipality belong to the upper reaches of the Yangtze River. The middle reaches contain Hubei, Hunan, and Jiangxi provinces. Anhui, Jiangsu provinces, and Shanghai municipality are in the downstream reaches of the river. As the animal farming in Tibet is too small and statistic data is lacking, Tibet was excluded from this study.

1.2. Mass estimation of manure-borne steroid hormones

1.2.1. Selection of livestock and hormones

Based on the statistics of animal production and manure production in these 10 provinces and municipalities (Table 1), it is clear that pigs, dairy and beef cattle, and chickens are dominant types of livestock and poultry, accounting for 94.1% of animal production and 95.2% of manure production. Since the populations of other species such as sheep, horses and rabbits are extremely small, they are not representative enough to be considered. In addition, since the majority of studies on manure-borne hormones have been focused on pigs, dairy and beef cattle, laying hens and broilers, there has been sufficient and systematic data for the estimation of these 5 species. Therefore, pigs, dairy and beef cattle, laying hens and broilers were selected in this study.

The steroid estrogens (SEs) and the steroid androgenic (SAs) are frequently studied sex hormones, because they are emitted in huge quantities by animals and are the most probable cause of endocrine disruption observed in aquatic organisms (Arnon et al., 2008; Barbosa et al., 2008; Gadd et al., 2010). The manure-borne endogenous SEs mainly include estrone (E1), 17α-estradiol (αE2), βE2 (17β-estradiol), and estriol (E3). While the range of steroid estrogens and their metabolites varies among vertebrates, βE2 and E1 appear to be

Table 1 – Animal[a] and manure[b] production in the Yangtze River basin in 2013.

Livestock category	Animal production (million head/year)	Percentage (%)	Manure production ($\times 10^4$ ton/day)	Percentage (%)
Beef cattle	25.2	2.3	53.1	27.9
Dairy	1.3	0.1	7.2	3.8
Swine	214.3	19.4	111.3	58.4
Chicken	799.3	72.3	9.9	5.2
Other categories	65.6	5.9	9.1	4.8
Total	1105.7	100.0	190.6	100.0

[a] Animal production data are from China Agriculture Yearbook (2013).
[b] Manure production was calculated as the mean excreted coefficients suggested by Ma et al., 2006.