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Effect of the chlortetracycline addition method on methane production from the anaerobic digestion of swine wastewater

Lu Huang[1], *Xin Wen*[1], *Yan Wang*[1,2,3], *Yongde Zou*[4], *Baohua Ma*[4], *Xindi Liao*[1,2,3], *Juanboo Liang*[5], *Yinbao Wu*[1,2,3,*]

1. *College of Animal Science, National Engineering Research Center for Breeding Swine Industry, South China Agricultural University, Guangzhou 510642, China. E-mail: lu.huang@merial.com*
2. *Ministry of Agriculture, Key Laboratory of Tropical Agricultural Environment, South China Agricultural University, Guangzhou 510642, China*
3. *Guangdong Enterprise Lab of Healthy Animal Husbandry and Environment Control, Xinxing 527400, China*
4. *Nanhai Entry-Exit Inspection and Quarantine Bureau, Foshan 528200, China*
5. *Institute of Tropical Agriculture, Universiti Putra Malaysia, 43400 Serdang, Malaysia*

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ABSTRACT

Effects of antibiotic residues on methane production in anaerobic digestion are commonly studied using the following two antibiotic addition methods: (1) adding manure from animals that consume a diet containing antibiotics, and (2) adding antibiotic-free animal manure spiked with antibiotics. This study used chlortetracycline (CTC) as a model antibiotic to examine the effects of the antibiotic addition method on methane production in anaerobic digestion under two different swine wastewater concentrations (0.55 and 0.22 mg CTC/g dry manure). The results showed that CTC degradation rate in which manure was directly added at 0.55 mg CTC/g (H_{SPIKE} treatment) was lower than the control values and the rest of the treatment groups. Methane production from the H_{SPIKE} treatment was reduced ($p < 0.05$) by 12% during the whole experimental period and 15% during the first 7 days. The treatments had no significant effect on the pH and chemical oxygen demand value of the digesters, and the total nitrogen of the 0.55 mg CTC/kg manure collected from mediated swine was significantly higher than the other values. Therefore, different methane production under different antibiotic addition methods might be explained by the microbial activity and the concentrations of antibiotic intermediate products and metabolites. Because the primary entry route of veterinary antibiotics into an anaerobic digester is by contaminated animal manure, the most appropriate method for studying antibiotic residue effects on methane production may be using manure from animals that are given a particular antibiotic, rather than adding the antibiotic directly to the anaerobic digester.

Introduction

Veterinary antibiotics are widely used in animal husbandry to enhance growth and to improve feed efficiency and animal health at sub-therapeutic dosages. According to the IFAH (International Federation for Animal Health, Belgium) annual report, the global gross sales of veterinary drugs including veterinary medicine and feed additives increased from $8.65

* *Corresponding author*. E-mail: wuyinbao@scau.edu.cn (Yinbao Wu).

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trillion in 1999 to $18.60 billion in 2009, representing an average annual increase of 5.2% (IFAH, 2009). Although the use of veterinary antibiotics as growth promoters has been progressively restricted by the EU and some industrialized countries, these drugs are still used in large quantities in animal husbandry for prophylaxis or treatment (Marco et al., 2003). After treatment, veterinary antibiotics are excreted via feces and urine in their original form or as active metabolites that have a direct or indirect effect on environmental organisms. Previous research demonstrated that the excretion level of the parent material and its metabolites accounted for 40% to 90% of the total dosage of administered veterinary antibiotics (Halling-Sørensen et al., 1998; Jemba, 2002; Phillips et al., 2004). A study on the ecosystem effects of veterinary antibiotics has been the recent focus of international ecotoxicological assessments.

Chlortetracycline (CTC) is a broad-spectrum antibiotic in the tetracycline (TC) family, and it inhibits protein synthesis in many common gram-positive and gram-negative bacteria (Fang et al., 2014). It is widely administered to farm animals to control intestinal and respiratory infections. Zhang et al. (2008) determined the primary chemical components of swine and poultry manure that were collected from typical intensive animal farms in seven provinces or municipalities in China. The authors reported that the average contents of oxytetracycline (OTC), TC, and CTC in manure were 9.09, 5.22, and 3.57 mg/kg, respectively, which suggested the presence of significant levels of antibiotic residues in the environment.

The inhibitory effects by antibiotics on methane production are inconsistent with prior reports (Lallai et al., 2002; Poels et al., 1984; Sanz et al., 1996; Shi et al., 2011). The discrepancy between reports could be caused by differences in the type and concentration of antibiotics, the reaction temperature, reactor type, and ratio of manure to water. Anaerobic digestion is an important component of the three-stage wastewater treatment system that is widely used for livestock wastewater treatment in large and medium-scale swine farms in China. The efficiency of anaerobic digestion is affected by various factors, including veterinary antibiotic residues. This issue has generated considerable concern and research. The two most common methods where antibiotics are added to an anaerobic digestion system to study their effect on methane production are by (1) adding manure from animals that consumed a diet containing antibiotics and (2) supplementing antibiotic-free manure with veterinary antibiotic before adding the manure to the anaerobic digestion system. Stone et al. (2009) reported that manure and urine from swine receiving a diet with 22 mg CTC/kg body weight exhibited a 27.8% inhibition in methane generation during anaerobic digestion. Loftin et al. (2005) directly added eight common veterinary antibiotics to an anaerobic digester and found that the methane production had been inhibited by 20% to 45%. Although many investigations have addressed the antibiotic effects on methane yields during anaerobic digestion, few studies have distinguished between two common antibiotic addition methods. It is not clear whether the two different antibiotic addition methods would actually lead to differences in the methane production from an anaerobic digestion system. This study was designed to examine the effects of two different initial CTC concentrations on methane production by an anaerobic digestion system in laboratory-scale semibatch reactors under two antibiotic addition methods.

1. Materials and methods

1.1. Swine manure collection

Twenty growing swine (± 30 kg) consumed an antibiotic-free diet for 28 days and were used to supply the manure for this study. The swine were divided into two groups of equal number; one group (control) continued to receive an antibiotic-free diet and the other received a similar diet containing 100 g of CTC/ton feed (Allen et al., 2011) for 7 days. Approximately 2 kg of pooled manure excreted by the swine in each group was collected daily and stored at −20°C for subsequent use. The samples from the CTC treatment (n = 7) were analyzed for CTC concentrations. The maximum (0.55 mg/g dry manure, DM) and average (0.22 mg/g DM) CTC concentrations detected among the manure samples from the CTC treatment group were used as treatment levels for the anaerobic digestion study.

1.2. Anaerobic digesters

The experiment was conducted in laboratory model anaerobic reactors. Each model was a three-neck glass reactor with a working volume of 1.3 L. The three openings of the digester were sealed with rubber stoppers. Swine slurry was added to the reactor through the left opening, and the displaced content was collected from the right opening. The biogas generated from the system flowed into a 500 mL absorbent flask filled with 0.5 mol/L NaOH solution via a flexible tube; the volume of NaOH solution replaced by incoming methane was pushed out of the absorbent flask into a receiver flask, and the volume of methane was obtained by measuring the volume of NaOH solution collected in the receiver flask. The methane concentration in the biogas absorbed by NaOH solution was more than 99.8% (You et al., 2003; Yang and Li, 2004).

The reactor was filled with 100 g of dry swine manure from swine that consumed an antibiotic-free diet. One hundred milliliters of slurry obtained from an anaerobic wastewater treatment pond from the same farm was added to the digester as inoculum, followed by 800 mL of water. The headspaces of the reactor were then flushed with nitrogen gas to maintain an anaerobic condition. At 08:00 am, the reactor was swirled gently to allow its content to mix evenly before 100 mL of the slurry was poured out of the reactor through the right-hand opening. The content was immediately replaced with an equal volume of antibiotic-free manure (10 g of dry manure/100 mL water) via the left opening. The reactors were used for subsequent anaerobic digestion experiments only when the biogas production volume in all of them had reached equilibrium (approximately 15 days).

1.3. Experimental design

The experiment was set up in a completely randomized design consisting of a control (antibiotic-free manure) and four treatments. The treatment groups were designed to have a combination of two antibiotic sources, i.e., the addition of CTC via manure containing 0.55 and 0.22 mg CTC/g manure from swine receiving a diet with CTC (H_{MED} and L_{MED}, respectively) and the direct spiking of the above two CTC levels into the system (H_{SPIKE} and L_{SPIKE}, respectively).

Each treatment was performed in triplicate by using 3 reactors, which were placed in a biochemical incubator at 20 ± 0.5°C. The experimental period was 28 days. In the control, 100 mL of antibiotic-free manure slurry (prepared as described in Section 1.2) was added daily throughout the 28 days; for the treatment groups, the antibiotic-containing manure slurry with the indicated treatments was added for the initial 7 days followed by antibiotic-free manure slurry for the remaining 21 days.

1.4. Sample collection and analysis

The biogas production of each reactor was measured at 08:00 am and 06:00 pm as described in Section 1.2. Fifty milliliters of the collected slurry was sampled from each reactor on days 0, 1, 2, 3, 4, 5, 6, 7, 8, 10, 12, 14, 17, 21 and 28, and their respective pH, chemical oxygen demand (COD), total nitrogen (TN) and CTC values were determined. The initial pH, COD, TN and ammonia nitrogen values were 6.02 ± 0.01, 50.72 ± 1.30 mg/mL, 2.26 ± 0.04 mg/mL and 1527.53 ± 35.22 mg/L, respectively.

1.5. Detecting CTC in the slurry

Five milliliters of slurry samples collected from each reactor was extracted twice with 5 mL of 0.01 mol/L Na_2EDTA in Mcl-Vaine buffer by vortexing for 30 sec and then vibrating for 10 min at 3000 r/min. After each extraction, the extracts were centrifuged at 12,000 r/min for 10 min at 5°C. The supernatants were pooled in a new centrifuge tube, centrifuged again and filtered through 2.5 mm filter paper using a Buchner funnel, and they were passed through prewashed Phenomenex Strata-X-CW C-18 cartridges. The cartridges were prewashed with 1 mL of methanol followed by 1 mL of distilled water. After the extracts were loaded, the cartridges were flushed with 1 mL of distilled water, followed by sample elution by using 1 mL of methanol and 1 mL of 2% formic acid-methanol solution. Eluents were filtered with a 0.22 μm filter prior to HPLC analysis (Waters 600 Controller, Waters 717 plus Autosampler and Waters 600E-2487 Dual λ Absorbance Detector, USA) with a Kromasil C18 column (250 mm × 4.6 mm, 5 μm, 100A) at 30°C. The detection wavelength was 275 nm and the injection volume was 10 μL. The mobile phase was acetonitrile-5% acetic acid (10:90, V/V) and the flow rate was set to 1.0 mL/min. Under the above conditions, CTC could be detached from the other components in the swine slurry. The CTC retention was 20.51 min with a detection limit of 0.01 μg/mL. The average CTC recovery of blank slurry samples spiked with 0.01 to 5 μg/mL CTC was 71.47%.

1.6. Statistical analysis

The data were statistically analyzed with SPSS 13.0. Analysis of variance and Turkey's test were used to compare the means. Significance was accepted at $p < 0.05$.

2. Results

2.1. CTC concentrations in the slurry

The CTC concentrations of the different treatment groups during anaerobic digestion are shown in Fig. 1. After adding CTC to the reactors, the CTC concentrations of the four treatment groups increased rapidly and plateaued 2 to 6 days later, and no CTC was detected in the control. The average CTC concentrations were 19.68, 52.73, 24.85 and 24.93 mg/L for the H_{MED}, H_{SPIKE}, L_{MED} and L_{SPIKE}, respectively, during the initial 7 days. The CTC concentration in the treatment groups decreased gradually after withdrawing CTC and could not be detected for the 0.22 mg CTC groups (L_{MED} and L_{SPIKE}) on the ninth day or for the 0.55 mg CTC groups (H_{MED} and H_{SPIKE}) on the eleventh day.

The statistical analysis showed that the CTC concentration of the H_{SPIKE} group was higher ($p < 0.05$) than that of the control and the other treatment groups, which were not significantly different during the initial 7 days when CTC was added, indicating that the CTC in H_{MED} group degraded faster than that in H_{SPIKE}.

2.2. Effect of CTC on methane production

The CTC effect on methane production is shown in Fig. 2. During the experimental period, the values for mean daily methane production within each treatment group ranged from 60 to 120 mL. The average methane production for the control, H_{MED}, H_{SPIKE}, L_{MED} and L_{SPIKE} was 103.40 ± 1.49, 91.37 ± 2.44, 90.95 ± 2.71, 95.24 ± 2.16, and 100.56 ± 1.74 mL/day, respectively, for the whole experimental period, and 97.29 ± 3.83, 93.00 ± 5.22, 82.24 ± 3.46, 98.67 ± 2.91, and 94.33 ± 3.24 mL/day, respectively during the 7-day period when CTC was added. A variance analysis demonstrated that methane production from the H_{SPIKE} was 12% lower ($p < 0.05$) than that of control for the whole experimental period and approximately 15% during the 7-day CTC addition period. Methane production from the H_{MED} was also significantly lower ($p < 0.05$) than that of control for the whole experimental period. Methane production from the other treatment groups was not significantly different from the control.

2.3. Effect CTC of on pH, COD and TN

The effect of CTC on the pH value of the anaerobic digestion slurry is shown in Fig. 3a. The pH value of the control group exhibited a similar trend to that of the treatment groups during the whole experimental period. The result indicated

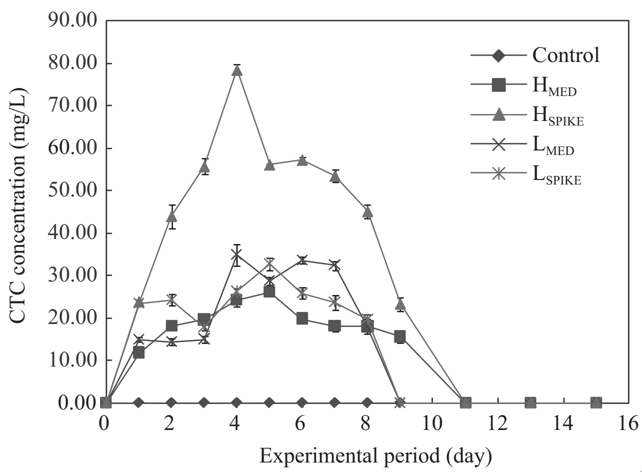

Fig. 1 – Chlortetracycline (CTC) concentration in the digester slurry.

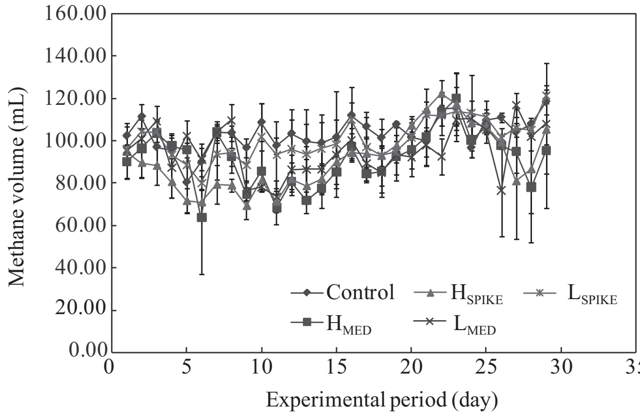

Fig. 2 – Methane production by different treatment groups.

that adding CTC within the concentrations and inclusion methods in this study did not significantly affect the pH value in the anaerobic digesters. Because the pH value was an important factor affecting the methane produced by anaerobic digestion, the pH value change was not responsible for methane production inhibition in H_{SPIKE}.

COD changes in the digesters during the experimental period are shown in Fig. 3b. No significant COD differences were detected, both for the different CTC concentrations and the antibiotic addition methods ($p > 0.05$) during the initial 7 days and over the entire experimental period.

The CTC effect on the TN of the slurry during the experimental period is shown in Fig. 3c. The average TN concentrations were 1.96, 2.24, 1.92, 2.04 and 2.03 mg/mL for the control, H_{MED}, H_{SPIKE}, L_{MED} and L_{SPIKE}, respectively. The statistical analysis showed that the TN concentration of the H_{MED} was significantly higher ($p < 0.05$) than that of the other groups (with no difference among them) during the 7 days of the CTC addition period.

The results suggested that CTC had no negative effects on the pH value and COD of anaerobic slurry because the digester content was mixed daily by adding manure slurry. The above results were consistent with those reported by Arikan et al. (2006) and Fernandez et al. (2009) showing that although an antibiotic (OTC) inhibited cumulative biogas production, it did not negatively affect the stability of the process because there were no significant differences in the reductions in volatile solids, soluble organic carbon and physical properties of the biomass. However, the TN in the H_{MED} treatment was significantly inhibited when compared with that in the control and the other treatments. The above result suggested that the microbial population and/or activities influencing the N concentration during the anaerobic fermentation process may be affected by the presence of CTC or its metabolites.

3. Discussion

3.1. CTC degradation in anaerobic digestion systems

The results showed that the CTC was below a detectable level on the third day in the 0.22 mg/g CTC initial concentration treatments (L_{MED} and L_{SPIKE}), but this result occurred on the fifth day for the 0.55 mg CTC/g treatments (H_{MED} and H_{SPIKE}). This timing might have occurred because of the different initial CTC concentrations. Shi et al. (2011) directly added 25 and 50 mg/L TC to anaerobic digesters and found that TC degraded rapidly during the initial stage, with more than 50% degradation within the first 12 hr, and the degradation rate slowed down thereafter with no detectable TC on day 20. However, Stone et al. (2009) reported that the CTC concentrations decreased from 27.0 to 11.6 mg/L in an anaerobic batch digestion of swine manure over 216 days with the greatest degradation rate occurring between 0 and 82 days, suggesting that the CTC concentrations remained relatively consistent. Similarly, Arikan (2008) collected manure samples from beef calves that were medicated with OTC to study the OTC effect on anaerobic digestion at 35°C, and they reported that the OTC levels in the manure slurry decreased from 9.8 to 4.1 mg/L in 64 days with a half-life of 56 days. In comparison with the above studies, the CTC retention time in anaerobic reactors in this study was relatively short. The shorter retention time recorded here could be explained by the lower initial CTC concentration in comparison with the above studies and the adaptation of the dynamic anaerobic digestion model in which the CTC content of the digester was continuously diluted by adding CTC-free swine manure during the last 20 days. It must be emphasized that the two CTC levels used for this study were based on the maximum and mean CTC values in the feces of swine receiving a diet with 100 g CTC/ton feed (Allen et al., 2011) for 7 days, and thus they were more relevant under practical conditions.

Interestingly, the result of this study indicated that the CTC degradation rate was faster in the H_{MED} than in the H_{SPIKE} treatment at similar initial CTC concentrations. The reasons for this difference could be (1) the adaptation of microorganisms in the H_{MED} treatment, because they were pre-exposed to CTC earlier and had more resistance to CTC than those in the H_{SPIKE} treatment, and (2) the effect of the chemical reaction and the concentration of intermediate products during CTC degradation. Our previous study (Chen et al., 2014) compared the OTC degradation in soil under different antibiotic addition methods. The OTC degradation half-lives exhibited the following significant differences ($p < 0.05$): manure from swine receiving

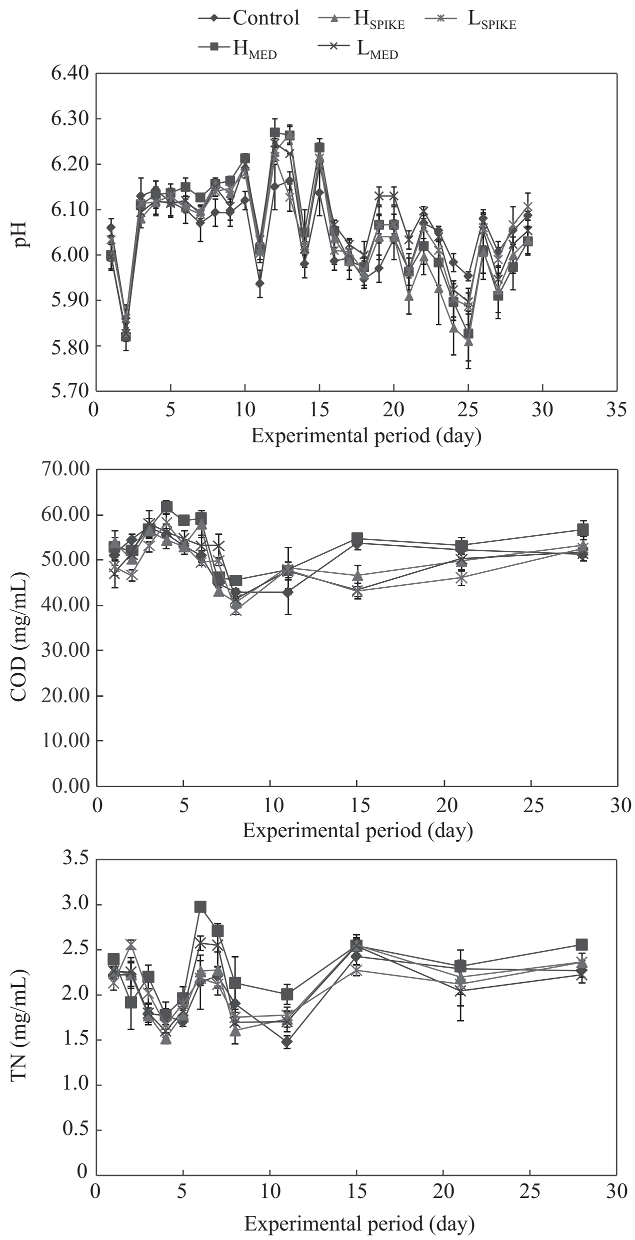

Fig. 3 – Variation of digester pH (a), chemical oxygen demand (COD) (b), and total nitrogen (TN) (c) during anaerobic digestion.

OTC < antibiotic-free manure + OTC < OTC. The differences could have been caused by distinct chemical reaction equilibria from dissimilar concentrations of 4-epi-OTC and α-apo-OTC. Soeborg et al. (2004) reported low CTC biodegradation in the environment, and its stability was usually dependent on abiotic factors such as temperature, organic matter and the pH value of environmental media. In this study, all the above mentioned abiotic factors were similar among treatments; therefore, they could not be the factors that influenced the CTC biodegradation rates in the different treatments. However, we only determined the concentration of CTC parent material without considering the presence and role of its metabolites. A literature review (Sankvist et al., 1984; Halling-Sørensen et al., 2005; Arikan et al., 2006) suggested that the effect of veterinary antibiotic metabolites is inconsistent and thus worthy of further studies.

3.2. Effects of CTC addition methods on methane production by anaerobic digestion

Anaerobic digestion is widely used for swine wastewater treatment because of its ability to remove organic substances and produce biogas. The results of this study showed that methane production in the H_{SPIKE} (0.55 mg/kg initial CTC) was significantly inhibited by approximately 15% during the first 7 days in comparison with the control, and no differences were detected between the control and the other experimental groups. This finding might be explained by (1) the lower degradation rate of CTC in the H_{SPIKE} treatment leading to a higher remaining concentration of CTC in the digester to inhibit microbial activities. The inhibition of related microbial activities might reduce methane production by the H_{SPIKE} treatment group and (2) the concentration of antibiotic metabolites and the degradation of intermediate products. Results by Fedler and Day (1985) and Sankvist et al. (1984) suggested that veterinary antibiotic metabolites inhibited anaerobic bacterial activity more than the veterinary antibiotic itself. However, Halling-Sørensen et al. (2005) reported that the OTC degradation products have less biological activity on sludge bacteria than OTC itself. We have not found any studies on the effects of CTC metabolites and intermediate products on methane production by anaerobic fermentation and thus must conduct further study.

4. Conclusions

Inconsistencies exist among various studies regarding the antibiotic effects on methane production by the anaerobic fermentation of swine manure slurry. This finding might be explained by the different antibiotic addition methods in the anaerobic digestion system. The results showed that direct CTC addition to anaerobic fermentation (H_{SPIKE}) resulted in a lower CTC degradation rate than the manure from swine receiving CTC (H_{MED}). However, the methane production was inhibited in the H_{SPIKE} treatment. This inhibition might be explained by (1) a slower CTC degradation rate in the H_{SPIKE} treatment leading to a higher CTC concentration in the digester, which inhibited microbial activity and methane production; and (2) the effect of the chemical reaction and the intermediate product concentration during CTC degradation. Because the primary entry route of veterinary antibiotics into an anaerobic digester is via contaminated animal manure, our findings suggest that it is more appropriate to use manure from animals that received antibiotics, which is more adopted to assess the CTC effect on methane production by anaerobic digestion.

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