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Monitoring of selected veterinary antibiotics in environmental compartments near a composting facility in Gangwon Province, Korea

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Abstract Many studies have been recently reported that veterinary antibiotics released into the environment have a detrimental effect on humans such as the occurrence of antibiotic-resistant bacteria. However, only limited information is available regarding to the release of antibiotics in environmental compartments in Korea. Objectives of this study were to evaluate the concentrations of antibiotics in water, sediment, and soil adjacent to a composting facility in Korea and to determine the dilution effects of antibiotics when released into the environment. Seven antibiotics of chlortetracycline, oxytetracycline, tetracycline, sulfamethazine, sulfamethoxazole, sulfathiazole, and tylosin were evaluated by high-performance liquid chromatography–tandem mass spectrometry following pretreatment using solid-phase extraction to clean the samples. Results showed that the highest concentration of each antibiotic in both aqueous and solid samples was detected from a site adjacent to the composting facility. We also found that the studied water, sediment, and soil samples are contaminated by veterinary antibiotics throughout comparison with studies from other countries. However, relatively lower concentrations of each antibiotic were observed from the rice paddy soil located at the bottom of the water stream. Further research is necessary to continuously monitor the antibiotics release into ecosystems, thereby developing an environmental risk assessment.

Keywords Veterinary antibiotics · Solid-phase extraction · Soil · Sediment · Water

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Introduction

Overuse of antibiotics causes detrimental effects on aquatic and terrestrial ecosystems such as the generation of antibiotic-resistant bacteria in the environment (Kemper 2008; Klavarioti et al. 2008; Kümmerer 2008). Veterinary antibiotics induce a severe environmental risk than antibiotics from human pharmaceuticals because they can be directly released into the environment as animal manure (Park et al. 2007). Due to detrimental toxicity, the release of antibiotic residues should be strictly regulated to reduce potential effects on degradations of groundwater, drinking water, and soil (Heim et al. 2004).

About 200,000 tons year^{-1} of antibiotics has been used over the world, served as not only a growth promoter but also an animal medicine in intensive livestock farming systems (Cars et al. 2008; Wise 2002). Especially, veterinary antibiotic groups of tetracycline (TCs), sulfonamide (SAs), and macrolide (MLs) are the most commonly used (Ha et al. 2003; Kim and Carlson 2007a; Kong et al. 2007). However, only 10–20% of antibiotic use can be actively reacted in the animal's body and the rest of these is excreted via urine and feces as a form of parent compounds with their metabolites (Kim et al. 2010). With/without composting, excreted urine and feces from animals are directly top-dressed to soils as an alternative of chemical fertilizers; therefore, veterinary antibiotics can be readily exposed to the environment (Aga et al. 2003; Boxall et al. 2002; Halling-Sorensen et al. 2002; Montforts et al. 1999; Tolls 2001; Vaclavik et al. 2004; Winckler and Grafe 2001).

In Korea, the annual consumption of veterinary antibiotics is magnitude compared to other countries (Kim et al. 2008). Total amount of antibiotic use in Korea was 15 times greater than that in Sweden in 2003 and two times greater than that in the UK in 2004. In addition, total amount of antibiotic use in Korea was 1.5 and 16 times greater than that in Japan and Denmark in 2005 (Johansson and Mollby 2006; NVRQS 2005; VMD 2005). Among the 20 veterinary antibiotic active ingredients (AIs) which are classified by priority on an environmental risk assessment, five antibiotics of chlortetracycline (CTC), oxytetracycline (OTC), sulfamethazine (SMT), sulfathiazole (STZ), and tylosin (TYL) were selected as the greatest priority group in Korea (Seo et al. 2007).

Many studied have reported severe antibiotics contamination in the aquatic environments (Golet et al. 2002; Gross et al. 2004; Kim et al. 2007; Kolpin et al. 2002; Lin et al. 2006). Bradford et al. (2008) found that the concentrations of antimicrobial agent ranged from <0.01 to 1,340 μg L^{-1} in swine, poultry, dairy, and beef lagoon water samples depending on types of antibiotics and lagoon source. Lin et al. (2008) also showed that the SMX residue was detected with an average concentration of 0.2 μg L^{-1} in $>91\%$ of Taiwanese water samples from 23 sites. In addition, the antibiotics of OTC, TC, SMX, and CTC were detected in water samples collected from the Cache la Poudre River in Northern Colorado, USA, and ranged 0.07–0.15 μg L^{-1}, 0.06–0.16 μg L^{-1}, 0.03–0.16 μg L^{-1}, and 0–0.18 μg L^{-1}, respectively (Carlson et al. 2004). In Korea, the concentrations of TCs and SAs ranged 0.11–0.97 μg L^{-1} and 0.45–10.57 μg L^{-1}, respectively, in 12 sewage treatment plant (STP) influents and wastewater sampling sites (Choi et al. 2007). Choi et al. (2008) also found that the highest concentration of SMX (average 0.52 μg L^{-1}) was detected in STP influents, whereas, the SMX concentration in surface water samples was lower than in STP influents (average 0.03 μg L^{-1}). In addition, the significant concentrations (>1 μg L^{-1}) of SMT have been detected in samples which are collected from the four major river basins in Korea, including the Han, Nakdong, Kum, and Youngsan Rivers (Park et al. 2007).

Contaminations of antibiotics in soils and sediments have also received great concern. Hamscher et al. (2002) found that the concentrations of TCs and CTCs were 86.2 μg kg^{-1} and >7.3 μg kg^{-1}, respectively, in top soil after numerous applications of liquid manure from fish farms in Germany. Another study investigated the concentrations of TC, CTC, OTC, SMX, and STZ in sediments which are collected from five sites located at the Cache la Poudre River in Northern Colorado, USA, ranged 4.5–32.8, 9.6–30.8, 5.6–8.3, 1.2–1.7, and 1.7–3.4 μg kg^{-1}, respectively (Kim and Carlson 2007a). They also reported that the concentrations of antibiotics in sediments depend on different physicochemical properties of antibiotic groups (Kim and Carlson 2007b).

Despite detrimental effects of antibiotics which are released into the environment, available information is very limited. The objectives of this study were to determine the levels of antibiotics in water, sediment, and soil adjacent to a manure composting facility in Korea and to compare the contaminations from different antibiotics groups through a monitoring of antibiotic residues in the environment. In addition, this study might be contributed to development of effective management strategies for minimizing the release of antibiotics into the environment in Korea.

Materials and methods

Chemicals

Seven antibiotics such as TC, CTC, OTC, SMT, SMX, STZ, and TYL were used in this study (Dr. Ehrenstorfer GmbH, Germany). These antibiotics were selected because they are the most widely used as animal pharmaceuticals in Korea (Kim et al. 2010; Seo et al. 2007). Each stock solution of the seven antibiotics at a concentration of 100 mg L^{-1} was mixed with methanol (CH_3OH) and was instantly diluted every use to prevent deterioration.

Sample preparation

Sampling sites adjacent to a swine manure composting facility (37°34′28″ N, 127°57′25″ W) were selected in Hongcheon, Kangwon Province, Korea as shown in Fig. 1. In April 2008, surface water and sediment samples were collected from five different locations in a rocky and mountainous portion of the Naerincheon River. One liter water was sampled using amber glass bottles that had been pre-rinsed with acetone ($[CH_3]_2CO$) and DI water (Kim and Carlson 2007a). Sediments were sampled using sanitized zipper bags at the same location from which the water samples were collected, except at site 1. Soils were sampled from three locations, a paddy field which is located near the upstream portion of the Naerincheon River (site 1), an upland field cultivated with different vegetables (site 2), and another paddy field which is located near the downstream portion of the Naerincheon River (site 3). Five discrete soil core samples were collected and composited to homogenize.

In laboratory, water samples were filtered through 0.45 μm glass fiber filter papers and were stored in a refrigerator at −4°C to maintain initial properties. Soil and sediment samples were

Fig. 1 Sampling sites along the Hongcheon River. The *white squares* represent the water and sediment sampling sites while the *white circles* represent the soil sampling sites

Table 1 Physical and chemical properties of water, sediment, and soil samples

		pH	EC 10^{-4} S m^{-1}	T-N mg L^{-1}	NTU	EC S m^{-1}	T-N g kg^{-1}	OM %	CEC $cmol_{(+)}$ kg^{-1}	Texture
Water	Site 1	7.57	44.3	2.34	0.66					
	Site 2	7.82	1,543	123.99	18.2					
	Site 3	7.84	810	58.35	2.69					
	Site 4	7.76	85	3.09	0.83					
	Site 5	7.62	241.4	15.63	2.37					
Sediment	Site 2	6.99				1.09	1.90	0.94	5.58	
	Site 3	6.77				1.26	1.79	1.13	5.92	
	Site 4	7.04				0.42	0.94	0.68	4.61	
	Site 5	6.88				0.51	0.95	0.92	5.03	
Soil	Site 1	5.95				0.82	1.41	2.84	9.24	Sandy loam
	Site 2	5.57				0.97	2.53	2.89	10.14	Sandy loam
	Site 3	6.05				0.80	1.02	2.14	8.90	Sandy loam

air dried at room temperature in dark and then were passed through a 2-mm sieve to remove debris. For the water, sediment, and soil samples, physicochemical properties such as pH, electrical conductivity (EC), total nitrogen (T-N), turbidity (NTU; nephelometric turbidity unit), organic matter (OM), and cation exchange capacity (CEC) were characterized using the methods described by Ok et al. (2007), Yoon et al. (2004), and Yang et al. (2009) as shown in Table 1.

Antibiotic extraction and quantification

For analyzing antibiotics, this study used the same procedure which has been described by Kim and Carlson (2007a). Amount of 120-mL aliquot of sampled water was placed on a 250-mL flask and pH was adjusted to approximately 2.5 using 40% (*v*/*v*) sulfuric acid (H_2SO_4). Then, 500 μL of 5% disodium ethylenediamine tetraacetate (Na_2EDTA; *w*/*v*) was added to each water sample and was vigorously mixed for 15 min at 150 rpm. To extract TCs and SAs from the soil and sediment samples, 1 g of each sample was extracted with 20 mL of Mcllvaine buffer solution followed by shaking for 20 min at 400 rpm. The ammonium hydroxide (NH_3) buffer solution associated with 200 μL of 5% Na_2EDTA (1 mmol) was also used for extracting TYL. Extracted samples were centrifuged for 15 min at 4,000 rpm and then were filtered through 0.2-μm glass fiber filters. This extraction procedure was repeated and the final supernatant was stored at 4°C using a 40-mL vial.

Prior to analysis of quantification of the antibiotic residuals, the solid-phase extraction was used to clean up and concentrate. High-performance liquid chromatography–tandem mass-spectrometry analysis was conducted using API 3000. Electrospray ionization was also used to detect targeted antibiotics. Detailed information and programmed mobile phase conditions are summarized in Kim and Carlson (2007a, b). Each sample had three replicates in this study.

Statistics

Pearson correlation coefficient (*r*) was calculated between average concentration values of each antibiotic as dependent variables and independent variables such as the physiochemical properties of soil, sediment, and water using SAS 9.1 (SAS 2004).

Results

Physiochemical properties of water and soil

In Table 1, water sample from site 2 had the highest turbidity of 18.2 NTU, T-N of 123.99 mg L^{-1}, and EC of 1,543 μS cm^{-1}. Throughout the sampling sites, OM in the sediment and soil samples ranged 0.68–1.13% and 2.14–2.89%, respectively, and the pH values ranged 6.77–7.04 and 5.57–6.05, respectively.

Quality assurance

A recovery study and limit of quantification (LOQ) were evaluated in water and sediment samples. In the aqueous matrix, the recovery ranged 100–127%, 76–124%, and 89–114% for TCs, SAs, and MLs, respectively, while they ranged 40–114%, 62–111%, and 53–128%, respectively, for the corresponding antibiotics in the sediment samples (Kim and Carlson 2007a). The LOQ calculated using a statistical method was 0.01–0.02 μg L^{-1} for water samples and 0.6–2.3 μg kg^{-1} for sediment samples. Calibration curves were constructed in range of 0.01–5 μg L^{-1} for water samples and 1–90 μg kg^{-1} for sediment samples. All six calibration curves for three antibiotic groups were closely linear ($r^2 > 0.99$; Kim and Carlson 2007a, b).

Antibiotics in environmental compartments

Measured concentrations of antibiotics were shown in Table 2. For water samples, the highest antibiotic concentrations of each antibiotic were detected in site 2 which is located closest to the composting facility. At site 2, the concentrations of OTC and SMT represent 51.27% and 61.41% of the total estimated amounts of TCs and SAs, respectively. For sediment samples, the concentrations of SAs and TYL were higher than those from the water samples and the highest concentration of SAs at site 3 (120.91 μg kg^{-1}) was observed because of the highest concentration of SMT (70.32 μg kg^{-1}) compared to other sites. For soil samples, site 2 had the highest concentrations of each antibiotic except for CTC, SMX, and STZ. Concentrations of OTC represent 53.78%, 53.70%, and 41.28% of the total estimated TCs in the soils at sites 1, 2, and 3, respectively. Conversely, site 1 had the higher concentrations of CTC, SMX, and STZ than other sites.

Concentrations of each antibiotic in water were significantly positively correlated with an EC (all $P < 0.01$), T-N (all $P < 0.01$), and turbidity (all $P < 0.001$), as shown in Table 3. For sediment, no correlation between physiochemical properties and concentrations of each antibiotic was found except for pH which had a significant correlation with the concentrations of SMT ($r = -0.988$), SAs ($r = -0.932$), and TYL ($r = 0.963$). Additionally, no significant correlation was found between all antibiotic concentrations in soil and soil physiochemical properties such as pH, EC, OM, T-N, and CEC.

Table 2 Average antibiotic levels in water, sediment, and soil samples

		Average antibiotic concentrations, μg L^{-1}								
		CTC	OTC	TC	TCs (sum)	SMT	SMX	STZ	SAs (sum)	TYL
Water	Site 1	0.54	0.39	0.50	1.43	0.31	0.36	0.30	0.97	0.23
	Site 2	0.76	1.41	0.58	2.75	9.12	0.38	5.35	14.85	2.19
	Site 3	0.57	0.40	0.51	1.48	0.39	0.36	0.46	1.21	0.25
	Site 4	0.53	0.39	0.50	1.42	0.31	0.36	0.30	0.97	0.23
	Site 5	0.55	0.39	0.50	1.44	0.41	0.36	0.39	1.16	0.23
Sediment	Site 2	BDL	BDL	BDL	–	39.37	8.91	23.68	71.96	13.38
	Site 3	BDL	BDL	BDL	–	70.32	11.49	39.10	120.91	12.32
	Site 4	BDL	BDL	BDL	–	38.60	11.77	31.12	81.49	13.28
	Site 5	BDL	BDL	BDL	–	54.93	12.20	40.31	107.44	12.66
Soil	Site 1	0.89	1.99	0.82	3.70	22.63	5.43	9.93	37.99	84.47
	Site 2	0.31	3.77	2.94	7.02	28.38	2.94	7.50	38.82	222.84
	Site 3	0.36	1.68	2.03	4.07	20.30	0.77	3.32	24.39	119.52

BDL below detected limit

Table 3 Correlation coefficient values between antibiotic concentrations and the physiochemical properties of water, sediment, and soil samples

Samples	Pearson correlation coefficients, *r*								
	CTC	OTC	TC	TCs (sum)	SMT	SMX	STZ	SAs (sum)	TYL
Water									
pH	0.510	0.460	0.534	0.473	0.453	0.453	0.468	0.460	0.460
EC	0.936***	0.881**	0.928***	0.894**	0.880**	0.877**	0.890**	0.884**	0.881**
T-N	0.951***	0.902**	0.944***	0.914**	0.901**	0.898**	0.910**	0.904**	0.902**
NTU	0.997***	0.993***	0.995***	0.993***	0.989***	0.988***	0.996***	0.995***	0.993***
Sediment									
pH	–	–	–	–	−0.988***	−0.328	−0.746	−0.932*	0.963***
EC	–	–	–	–	0.484	−0.554	−0.139	0.237	−0.321
OM	–	–	–	–	0.829	−0.130	0.379	0.671	−0.739
T-N	–	–	–	–	0.240	−0.752	−0.388	−0.022	−0.067
CEC	–	–	–	–	0.619	−0.438	0.039	0.396	−0.479
Soil									
pH	−0.962	0.186	0.802	0.414	0.041	−0.960	−0.889	−0.622	0.540
EC	0.983	−0.274	−0.853	−0.131	−0.131	0.930	0.843	0.548	−0.615
OM	0.488	0.562	−0.145	0.677	0.677	0.878	0.953	0.993*	0.212
T-N	0.946	−0.134	−0.769	0.012	0.012	0.973	0.912	0.662	−0.495
CEC	0.941	−0.117	−0.758	0.029	0.029	0.977	0.918	0.675	−0.480

* $P < 0.05$; ** $P < 0.01$; *** $P < 0.001$

Discussion

Our findings show that the representative veterinary antibiotics were released from the composting facility into neighboring ecosystems including water, sediment, and soil. As shown in Table 2, the concentrations of each antibiotic in water, sediment, and soil generally decreased as increasing the distance between the sampling locations and composting facility. In this study, various antibiotic concentrations in water samples were higher than those from studies in other countries which had different environmental matrices. Carlson et al. (2004) showed that an average value of OTC, CTC, TC, and SMX concentrations in water from the Catche La Poudre River of Northern Colorado, USA, was 0.16 μg L^{-1}. Lin et al. (2008) and Park et al. (2007) also showed that the concentrations of SMX and SMT in water were 0.2 μg L^{-1} and >1 μg L^{-1}, respectively. Their studies confirmed a fact that veterinary antibiotics are being overused in Korea compared to other countries (Kim et al. 2010; Seo et al. 2007).

For sediments, the measured concentrations of SMT, SMX, STZ, and SAs in this study were much higher than other measurements of Kim and Carlson (2007b). They collected sediment samples from the Catche La Poudre River of Northern Colorado, USA, and showed that the concentrations of each antibiotic in sediments varied with different sampling sites. Antibiotic concentrations of TCs had too low values to detect in this study, whereas, study of Kim and Carlson (2007b) reported the relatively high concentrations of TCs in sediment samples which were collected from the Catche La Poudre River of Northern Colorado, USA. We believe that this difference is resulted from degradation of TCs at the high temperature condition in Hongcheon, Korea. Our measurements were carried out during April (average 10.5°C), whereas, the Catche La Poudre River of Northern Colorado, USA which has the cold water temperatures and low flow conditions during winter (average −2.8°C) may enhance persistence of these substances (Kim and Carlson 2007b).

For soil samples, the concentration of each antibiotic may vary with application levels of animal manure, crop species, and physiochemical properties of soil. At site 2 which had the highest values of each antibiotic concentration, this site had been cultivated with different types of vegetable, whereas, other sites had rice crop only. Physiochemical properties of soil are also one of the most important factors influencing on antibiotic adsorption in soil (Kemper 2008). Indeed, the soil at site 2 had the higher values of EC (0.097 dS m^{-1}), T-N (2.53 g kg^{-1}), organic matter (2.89%), and CEC (10.14 $cmol_{(+)}$ kg^{-1}) compared to the soils at other sites. Each measured antibiotic concentration in this study was generally higher than other studies except for CTC and TC. Hamscher et al. (2002) showed that the levels of CTC and TC in the soil had 91.26% and 97.76% higher values in the fish farm under long-term investigation compared to our results. We speculate that their higher concentrations of CTC and TC are resulted from numerous top-dressing of liquid manure containing 0.1 mg L^{-1} and 4 mg L^{-1} of CTC and TC, respectively.

Each antibiotic concentration had significantly positive correlations with the physiochemical properties of water samples such as EC, T-N, and turbidity as shown in Table 3 (all $P < 0.01$). Our findings agree with a study of Kim and Carlson (2007b) indicating that the concentration of antibiotics in water is strongly affected by flow conditions and the physiochemical properties of water depending on the site condition and a type of antibiotics. We believe that the strong correlation may result from the impact of the swine composting facility wastewater effluent containing high levels of organic compounds, nutrients, and total soluble solids that possibly increasing water turbidity, T-N, and EC. Once the composting facility wastewater is mixed with water at a highly contaminated site, dilution may occur (Bradford et al. 2008; Yoon et al. 2004). Moreover, antibiotics having pharmaceutical molecules with polar and ionized properties are profoundly influenced by water pH (Kümmerer 2008).

For soils and sediments, antibiotic sorption to OM or soil particles in the solid phase may be reduced due to possession of charged state that leading a more complex ionic mechanism. This explains why the concentrations of each antibiotic were not strongly correlated with physiochemical properties of soil and sediment (Kümmerer 2008). Kim and Carlson (2007b) found that TCs had a strong sorption affinity to soil and organic particles, while SAs had the lowest K_{oc} and was least hydrophobic among antibiotics. Turku et al. (2007) also investigated the adsorption of TC on silica particles and found that approximately 9% of the adsorption sites were irreversible due to an increment of pH and temperature. As results, we believe that rice paddy soil may have complicated biochemical processes such as aerobic or oxidative cyclization, and anaerobic or reductive dechlorination, thereby readily inducing changes in physiochemical properties of soil.

Conclusion

Occurrence of veterinary antibiotic residues in aquatic and terrestrial environments has received great attention. Once veterinary antibiotic residues are excreted by animals and released into water, sediment, and soil, these detrimental impacts depend on types of antibiotics, physicochemical properties of soil and water, and location matrices. However, limited information is only available regarding to the release of antibiotics into the environment as well as a risk to human health and environmental safety. Our findings showed that the levels of each antibiotic concentration near a manure composting facility were higher in sediment and soil samples than in water. The highest concentration of each antibiotic was generally observed from a sampling site adjacent to the composting facility and the concentration gradually decreased as the distance increases between the composting facility and a sampling site. We found that the levels of each antibiotic in Korea were much higher than other countries, and antibiotics can be existed in both solid and aqueous ecosystems. However, the fate of antibiotics in the solid phase should be distinct from the aqueous phase and needs to be elucidated in future study. In addition, a screening of antibiotics in water, soil, and sediment is required to ensure agro-ecosystem safety and reduce the adverse effects of antibiotic residuals in the environment.

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