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Research Paper

Characteristics of antimicrobial residues in manure composts from swine farms: Residual patterns, removal efficiencies, and relation to purchased quantities and composting methods in Japan

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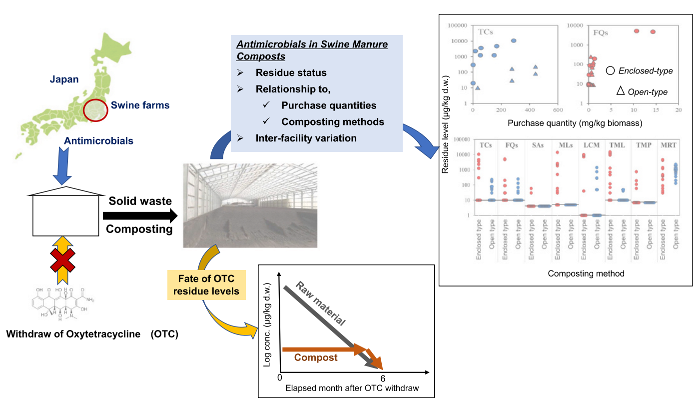
HIGHLIGHTS

- We present the first holistic report on antimicrobial levels in Japanese swine manure compost.
- Tilmicosin and tiamulin concentrations were the highest in composts.
- The removal efficiency depended on antimicrobial types, composting practices, and facility conditions.
- Morantel, a feed additive, was relatively resistant to degradation while composting.
- Once its usage was withdrawn, tetracycline residues in composts dissipated within 6 months.

GRAPHICAL ABSTRACT



Abbreviations: AMs, antimicrobials; AMR, antimicrobial resistance; ABs, antibiotics; ARB, antibiotic-resistant bacteria; TCs, tetracyclines (as AM classes); FQs, fluoroquinolones; SAs, sulfonamides; MLs, macrolides; TC, tetracycline (as an AM compound), OTC, oxytetracycline; CTC, chlortetracycline; DOXY, doxycycline; ERFX, enrofloxacin; CPFX, ciprofloxacin; NFLX, norfloxacin; MBFX, marbofloxacin; DNFX, danofloxacin; OBFX, orbifloxacin; SMX, sulfamethoxazole; SMMX, sulfamonomethoxine; SDMX, sulfadimethoxine; SDZ, sulfadiazine, SMZ, sulfamethazine (sulfadimidine); TS, tylosin; TMS, tilmicosin; LCM, lincomycin; TML, tiamulin; TMP, trimethoprim; MRT, morantel; RMs, raw materials for composts; MCs, matured composts from open-type facilities; ESFs, early stages of composting (nearly RMs) from open-type facilities; ISF, intermediate stages of composting from open-type facilities; SFPs, composts of secondary fermentation in piles (nearly MCs) from the enclosed-type facilities; WWT, wastewater treatment; MDL, method detection limits; MQL, method quantifying limits; EDTA, ethylenediamine-N,N,N′,N′-tetraacetic acid disodium salt dihydrate; PP, polypropylene; MHLW, The Ministry of Health, Labour and Welfare, Japan; MAFF, Ministry of Agriculture, Forestry and Fisheries, Japan; NAVL, National Veterinary Assay Laboratory, MAFF, Japan.

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A B S T R A C T

Present study provides first comprehensive results on the residual levels of 19 antimicrobial (AM) residues in 12 Japanese swine manure composting facilities that use open or enclosed types of treatment methods. Tilmicosin (14000 μg/kg d.w.) and tiamulin (15000 μg/kg d.w.) were present in the highest concentrations in manure composts. Morantel (MRT) had the highest detection frequency (100%) in compost, suggesting its ubiquitous usage and resistance to degradation during composting. Sulfamethoxazole had low detection frequencies and concentrations, likely due to limited partitioning to the solid phase. A positive correlation ($p < 0.05$) between purchasing quantities and residue levels in manure composts was detected for fluoroquinolones (FQs). The removal efficiencies of AMs in enclosed-type facilities were lower and more inconsistent than those in open-type facilities. Tetracyclines (TCs), lincomycin, and trimethoprim were easily removed from open-type facilities, whereas FQs and MRT persisted in both facilities. After discontinuing the usage of oxytetracycline (OTC), TCs concentrations reduced drastically in input materials, remained pseudo-persistent in composts for up to 4 months, suggesting a time lag for composting and were not detected (<10 μg/kg) after 4 months of OTC withdrawal. This study emphasizes on the effectiveness of manure composting methods in reducing AM residues in swine waste.

1. Introduction

Antimicrobials (AMs), including antibiotics (ABs), are among the greatest discoveries of modern medical sciences and are key driving forces for human health as well as veterinary and animal husbandry practices. Despite their usefulness, inappropriate usage of AMs has resulted in the emergence of AM resistance (AMR). In 2019, approximately 4.95 million deaths occurred due to bacterial AMR [29].

The global annual usage of ABs was estimated to be between 100000 and 200000 t [46] and has increased by 65% between 2000 and 2015 ([16]; from 21.1 to 34.8 billion defined daily doses in 76 countries). Van Boeckel et al. [37] estimated that the global AM consumption for livestock would increase by 67% from 2010 (63151 t) to 2030 (105596 t). A similar estimation was reported by Tiseo et al. [36] (increasing by 11.5% from 93309 t in 2017 to 104079 t in 2030).

In 2018, nearly 1761 t of AMs were traded in the Japanese market, almost half of which was used in veterinary drugs and animal feeds (as feed additives) for livestock [27]. Tetracyclines (TCs), penicillins, sulfonamides (SAs), and macrolides (MLs) as veterinary drugs and polyethers as feed additives are frequently sold AMs in Japanese livestock industries [27]. Veterinary ABs are more often used in swine farms (60%) than in bovine or poultry industries [27].

The application of livestock manure and its treated materials is one of the major sources of ABs, antibiotic-resistant bacteria (ARB), and antibiotic-resistant genes in the environment [15,43,44,49,5,9]. Once administered, 30–90% of these AMs are excreted by animals through urine and feces [12]. Thus, continuous application of these AMs in livestock industries may lead to their persistence in the environment. Even at trace levels, AMs can imply evolutionary bottlenecks in microbes to favor ARB [2,4].

In Japan, the annual amount of swine waste in 2017 was estimated to be approximately 22 million tons and accounted for approximately 30% of the total livestock waste [23]. Livestock wastes are commonly treated after seperating liquid and solid fractions. The liquid fraction is usually treated under aerobic conditions in an activated sludge system, whereas the solid fraction is mainly composted [39]. Conventionally, composting is performed in Japan using various methods, such as pile-type composting with manual or mechanical turning systems, windrow-type composting with mechanical turning systems, and enclosed vertical-type composting systems (forced-composting systems; [18]). All these composting methods are practiced by increasing the compost temperature (>60 °C) during aerobic fermentation. The end products (mature composts [MCs]) are preferably used as fertilizers and/or ameliorants in agricultural fields. In contrast to European countries and China, Japan has extremely limited storage or anaerobic treatment of swine (liquid) manure.

A mid-to-long-term strategy for sustainable food systems in Japan has been developed by reducing chemical fertilizer usage by 30% and increasing organic farming in 25% of farmlands [24]. However, if unchecked, such practices may risk the spread of recalcitrant chemicals, including AMs, in the environment and may result in adverse ecotoxicological consequences.

AM residues in swine manure composts have been already reported in Japan [28,48]. For instance, Yoshizawa et al. [48] analyzed and detected TCs at concentration ranges of 10^1–10^4 μg/kg in 29 composts from 11 swine farms, which were higher than those from cow farms. Meanwhile, Motoyama et al. [28] analyzed AM residue levels in compost originated from different raw materials (RMs), including swine manures, and demonstrated that 10 AMs belonging to 5 classes remained in composts. They found that the concentrations of chlortetracycline (CTC), sulfamonomethoxine (SMMX), and sulfamethoxazole (SMX, <method detection limits [MDL] – 280 μg/kg) in swine manure composts were significantly higher than those in composts originated from other organic sources.

However, the above studies did not provide details on facilities and farms, AM usage in farms, and their effects on AM residue levels. In addition, the number of AM compounds analyzed was limited. Therefore, this study aimed to elucidate the current residue levels and patterns in swine manure composts in Japan and to clarify their relationship with the purchase quantities of AMs and compositing methods. To the best of our knowledge, an actual farm study investigating the fate of AMs in composted manure after discontinuing their usage has never been reported. Therefore, for the first time, the long-term fate of oxytetracycline (OTC) in swine composts after its usage termination was assessed. The study results may provide countermeasures for reducing AM release from swine farms into the environment.

2. Materials and methods

2.1. Chemicals and materials

Eight classes of target AMs were selected on the basis of the purchasing records of farms (Table S1). Four TCs (TC, OTC, CTC, and doxycycline [DOXY]), six fluoroquinolones (FQs) (enrofloxacin [ERFX], ciprofloxacin [CPFX], norfloxacin [NFLX], marbofloxacin [MBFX], danofloxacin [DNFX], and orbifloxacin [OBFX]), three SAs (SMX, SMMX, and sulfadimethoxine [SDMX]), two MLs (tylosin [TS] and tilmicosin [TMS]), one lincosamides (lincomycin [LCM]), one pleuromutilin (tiamulin [TML]), one diaminopyrimidine (trimethoprim [TMP]), and one anthelmintic (morantel [MRT]) were selected for the analyses. In Japan, MRT is used as a feed additive for nutrient ingredients to promote the efficient conversion of feeds, whereas other AMs are currently used only for veterinary drugs. The details of these analytical standards were the same as those of Watanabe et al. [44] (Table S2).

A Captiva EMR-Lipid (3 mL, 300 mg, Agilent, CA, USA) cartridge column was used for purification. Other reagents and equipment were reported by Watanabe et al. [44].

2.2. Sample collection

In this study, 12 composting facilities from 10 farrow-to-finisher commercial private farms located in Kanto and Chubu regions of Japan were examined in 2018–2020 (Table 1). These farms were the same as those reported in a previous study [44]. Of these 10 farms, 4 (Farms B, F, G, and H) had separated their breeding sites based on animal growth stages for biological security control purposes. Farms B and H had composting facilities at each site (Facilities B1 and B2 and Facilities H1 and H2, respectively). Due to varying AM classes or quantities used at each farm site, both facilities within each farm were surveyed. Facility 2 at Farm F could not be surveyed as it outsourced swine waste (e.g., feces, spilled feeds, and sludge from wastewater treatment) composting. At Farm G, swine solid waste from both sites was combined and composted at site 2.

Each facility composted swine solid waste only from its own farm or site. The composting methods used at these facilities could be grouped into two categories: (1) open-type composting, such as pile type (Facilities A and H2) and windrow type (Facilities B1, B2, and C), and (2) enclosed-type composting used for vertical forced composting (Facilities D, E, F1, G, H1, I, and J). In open-type composting, the starting materials were fresh swine solid waste mixed with bulking agents, such as MCs, rice husks, and sawdust, for controlling the water content to approximately 60%. The starting materials were piled or placed at the inlet of the windrow. They were turned approximately once a week using bucket loaders at Facility A or 1–2 times a day using mechanical stirring at Facilities B1, B2, C, and H2. The compost temperature reached over 60 °C within a few days and was maintained for at least 1 week. The composting process continued for approximately 1–3 months. In contrast, in enclosed-type composting, swine solid waste was directly put into the enclosed-type facilities (in some cases, solid waste was mixed with bulk agents to control the water content). The composting chambers in these facilities held composts for approximately 10–20 days and maintained a temperature of over 60 °C at the top layer. Thereafter, the composts emitted from the chambers were further matured using the pile-type method in the stockyards of the enclosed-type facilities, except for Facility F1. It is worth noting that Facility I experienced operational failure, resulting in no increase in chamber temperature during the cold season sampling.

Nine facilities in eight farms were surveyed twice in different seasons, i.e., during the cold season (November to February) and the warm season (June to early October) (Table 1). Facility F1 was additionally surveyed two more times in different warm seasons to understand the intrafacility variation in residual AM concentrations. In addition, OTC usage was terminated at two facilities in Farm B (Facilities B1 and B2) to investigate the fate of TCs and other AMs during a 1-year post-OTC period by additionally collecting samples at 1, 2, 4, 6, and 12 months.

As end products, MCs from open-type facilities and composts of secondary fermentation in piles (SFPs) from enclosed-type facilities were collected. These samples were collected after removing approximately 30–50 cm of the surface of composts. After thoroughly mixing the surrounding composts using a polypropylene (PP) scoop, approximately 100-g samples were collected from each sampling point covering an area of 0.25 m^2. We also collected immature composts from the early and intermediate stages of composting (ESF and ISF, respectively) in open-type facilities and RMs from enclosed-type facilities using same sampling methods as those for MCs and SFPs. The ESF sample was almost the input material because it was collected at the starting point of composting (within a day). Notably, ESF, ISF, and RMs could not be

Table 1
Details of swine farms, composting methods, sample type, and sampling date.

Farm ID	Site ID	Facility ID	Numbers of Sows[a]	Production and growth stage	Composting methods	Bulking agents	Sample type[c]		Sampling date[d]	
							Cold season	Warm season	Cold season	Warm season
A	-	A	81	All	Pile-type	Rice husk, sawdust	-	ESF, MC	-	Jul-2020
B	1	B1	610	Sows - weaning, fattening[e]	Rotary-stirring windrow-type	Rice husk, sawdust	ESF, MC	ESF, MC	Jan-2018	Jun-2018
	2	B2		Fattening	Rotary-stirring windrow-type	Rice husk, sawdust	ESF, MC	ESF, MC	Jan-2018	Jun-2018
C	-	C	1867[b]	All	Rotary-stirring windrow-type	Matured compost	ESF, ISF, MC	ISF, MC	Feb-2018	Jul-2018
D	-	D	523	All	Enclosed vertical-type → Pile-type	-	RM, SFP	RM, SFP	Feb-2018	Aug-2018
E	-	E	863	All	Enclosed vertical-type → Pile-type	-	SFP	-	Feb-2018	-
F	1	F1	4934[b]	Sows - lactating	Enclosed vertical-type	-	RM, MC	RM, MC	Dec-2018	Aug-2018
	2	-		Weaning - fattening	Outsourcing	-	-	-	-	-
G	1	G	502	Sows-lactating, fattening[e]	Enclosed vertical-type → Pile-type	-	SFP	RM, SFP	Jan-2019	Sep-2019
	2			Weaning - fattening						
H	1	H1	321	Sows - weaning	Enclosed vertical-type → Pile-type	-	SFP	-	Nov-2019	Jun-2019
	2	H2		Fattening	Screw-stirring pile-type	Matured compost, sawdust	ESF, MC	ESF, MC	Nov-2019	Jun-2019
I	-	I	90	All	Enclosed vertical-type → Pile-type	-	SFP	SFP	Feb-2019	Aug-2019
J	-	J	236	All	Enclosed vertical-type → Pile-type	-	SFP	SFP	Dec-2019	Jun-2019

[a] Average numbers before six months of the first sampling. n.i.: no information
[b] Including numbers in other separate sites of the same farm
[c] RM: raw material, ESF: early stage of fermentation, ISF: intermediate stage of fermentation, SFP: secondary fermentation in pile type, MC: matured compost
[d] Farm B surveyed continuously for one year after the OTC withdrawal in Jul/2018. Samples were collected on Aug/2018 (1), Sep/2018 (2), Oct/2018 (4), Jan/2019 (6), Jul/2019 (12). (the value in parentheses indicates the elapsed months after OTC withdrawal). Farm F were also surveyed and collected the MC samples on June/2019 and Oct/2019 for understanding the validation of AM concentrations in same facility
[e] Most of the fattening pigs were at Site 2, while approximate 1/10 of them were at Site 1

collected from all facilities because of the limited or lack of accessibility to sampling points or absence of materials at the time of sampling. All samples were packed in PP bottles, transported to the laboratory under cool conditions, and stored at − 20 °C in a freezer.

The calculation of AM purchase quantities per population correction unit (mg/kg biomass) in farms has been reported by Watanabe et al. [44].

2.3. Analytical procedure

2.3.1. Extraction and clean-up procedures

Sample extraction and instrumental analysis were conducted separately for TCs and FQs, and for other AMs. The sample extraction and clean-up procedures for TCs/FQs and other AMs were modified following the method reported by Zhao and Lucas [52] using different solvent mixtures for TCs/FQs and other AMs. Three solvent types (A, B, and C), 5% of ethylenediamine-*N*,*N*,*N*′,*N*′-tetraacetic acid disodium salt dihydrate (EDTA) in ultrapure water (solvent A), 2% of formic acid (FA) in acetonitrile (solvent B), and a mixture of 20% of EDTA/water (0.1%) and 80% of FA/acetonitrile (2%) (solvent C) were used for the analyses of TCs and FQs. In contrast, for the analyses of other AMs, 0.1% EDTA/ultrapure water (solvent A), 0.2% FA/acetonitrile (solvent B), and a mixture of 20% of ultrapure water and 80% of FA/acetonitrile (0.2%) (solvent C) were used.

After removing large particles, such as wood chips samples were mixed well. Then 1 g of each of the ISF, MC, and SFP wet samples and 0.5 g of each of the ESF and RM wet samples were taken in clean 50-mL PP centrifuge tubes. The samples were spiked with 2 mL of solvent A and vortexed for 15 s. After addition of 4 mL of solvent B, the mixture was vortexed for 15 s, shaken for 15 min (470 rpm), and centrifuged at 3750 rpm (∼3000 *g*) at 4 °C for 10 min. The supernatant was transferred to a clean 15-mL PP centrifuge tube, and the remaining residue was added in 4 mL of solvent B and resuspended through ultrasonication. Then, all the above steps were repeated. The supernatant was combined with the first extract in the same 15-mL PP centrifuge tube, and the mixture was thoroughly vortexed. The PP tube was stored at − 20 °C overnight and then shaken for 15 min and centrifuged. The supernatant was then transferred to a new 15-mL PP tube as the final extract.

Three milliliters of the final extract was loaded onto a Captiva EMR-Lipid cartridge and purified through gravity flow, and the cartridge was rinsed with 0.7 mL of solvent C. Then, 3.5 mL of ultrapure water was added to the eluate and vortexed. A portion of this mixture was filtered before injection.

2.3.2. Quantification and identification of antimicrobials

We identified and quantified the analytes using ultra-high-performance liquid chromatography–tandem mass spectrometry. The optimal conditions and multiple reaction monitoring settings for the analytes were the same as those reported in a previous study by Watanabe et al. [44] (Table S2 and Table S3). The AM concentrations were calculated using the external calibration method and expressed on a dry-weight basis. For conversion of measured concentrations of AMs from wet-weight to dry weight, the water content in each sample was measured using the gravimetric method and was applied to the samples.

2.4. Quality control and quality assurance

The recovery rates, MDLs, and method quantifying limits (MQLs) of the targeted AMs were determined following the analysis of actual compost samples spiked with native standard mixtures (Table S4). Five compost samples spiked with quintuple levels of the native standard of the target compounds were analyzed. Overall, acceptable average recoveries were obtained, ranging from 59.6% to 120%, and the variation (relative standard deviation) of recoveries was within 23.8%. In contrast, on only a few occasions, the average recoveries of OTC, CTC, CPFX, and NFLX varied from 42.5% to 55.1%. Moreover (only once), CTC showed the lowest recovery rate of 27.7% at spike level 2 (220 μg/kg). In contrast, the average recoveries of OBFX and TMS were higher, ranging from 96.9% to 152%. The MDL and MQL of the target compounds ranged from 1 to 70 μg/kg and 4–200 μg/kg, respectively (Table S4). The recovery rates, MDL, and MQL for MRT were not calculated because of the high residue levels found in the actual samples (the compost matrices were not suitable for the selected spiking range).

2.5. Removal efficiency of AMs

The removal efficiency of AMs in the composting process was calculated according to the following formula (Eq. 1), assuming that there was no change in the material quantities on a dry-weight basis of the input and the end product at each facility.

$$RE = \frac{Ci - Ce}{Ci} \times 100 \quad (1)$$

where RE represents the removal efficiency, C_i represents the AM concentration in the input materials (ESF or RM), and C_e represents the AM concentration in the end products (MC or SFP). If AM was not detected in the end product but was detected in the input, MDL was used for calculating the removal efficiency. The removal efficiency is shown as “zero” if it was calculated in minus.

2.6. Statistical analysis

The median and average concentrations of AMs in manure composts were calculated from the samples (data were not included when concentrations were <MDL). The correlation between the purchase quantities and residual concentrations of AM classes in the end products was determined using Spearman’s rank correlation analysis. If the residual concentration of an AM in the end product was <MDL and the purchase quantity in this farm was 0 mg/kg biomass, the AM was removed from the statistical analysis. If the purchase quantity was > 0 mg/kg biomass and the residual concentration was <MDL, MDL was used as the residual concentration. The differences in the residue concentrations in the end products or purchase quantities between composting methods were determined using the Mann–Whitney U test analysis. Statistical analyses were performed using BellCurve for Excel (SSRI, Tokyo, Japan).

3. Results and discussion

3.1. AM occurrence and relationship to purchase quantity

This study reports the first comprehensive monitoring data for AMs in swine manure composts in Japan. Of the 19 tested AMs, DNFX, SMMX, and SDMX were not detected in any samples (<MDL). Thus, they were not considered for further discussion (Table S5). Meanwhile, the end products (MCs and SFPs) retained AMs at residue levels in wide ranges from <MDL to 15 mg/kg on a dry-weight basis (Table 2). Among the AM classes, the highest concentrations were detected for TML and MLs (15000 and 14000 μg/kg, respectively), followed by TCs (10000 μg/kg), LCM (10000 μg/kg), FQs (5200 μg/kg), and MRT (4700 μg/kg). The maximum residue levels for TMP (750 μg/kg) and SAs (60 μg/kg) were remarkably lower than those for other AMs. In each AM class, CTC for TCs (9500 μg/kg), NFLX for FQs (3600 μg/kg), SMX for SAs (60 μg/kg), and TMS for MLs (14000 μg/kg) had the highest residue concentrations (Table S5).

In the input materials (ESFs and RMs), the maximum concentration was 130000 μg/kg for MLs, which was close to the maximum levels for dosing to pigs (200 mg/kg = 200000 μg/kg in feeds), followed by that for TML (26000 μg/kg), TCs (24000 μg/kg), LCM (9600 μg/kg), FQs (7400 μg/kg), and MRT (3500 μg/kg) (Table S6). Notably, high AM residue levels detected in the input materials might have been influenced by less mixing compared with the MCs and SFPs, and samples

Table 2
Statistics of AM concentrations in end products from all, open-type, and enclosed-type composting facilities. in Japanese swine farms.

	TCs[a]	FQs	SAs[a]	MLs[a]	LCM	TML[a]	TMP[a]	MRT
All facilities (n = 33)								
average conc. (μg kg^{-1} d.w.)	1500	910	45	2400	2800	4200	280	1200
median conc. (μg kg^{-1} d.w.)	200	90	45	1100	290	590	150	940
maximum conc. (μg kg^{-1} d.w.)	10000	5200	60	14000	10000	15000	750	4700
minimum conc. (μg kg^{-1} d.w.)	10	30	30	10	20	30	60	30
detection frequency (%)	52	36	6.1	30	42	36	12	100
Open-type facilities (n = 19)								
average conc. (μg kg^{-1} d.w.)	100	100	-	-	320	45	-	1100
median conc. (μg kg^{-1} d.w.)	80	70	-	-	50	45	-	1000
maximum conc. (μg kg^{-1} d.w.)	230	250	-	-	1400	50	-	2300
minimum conc. (μg kg^{-1} d.w.)	10	30	-	-	20	40	-	130
detection frequency (%)	47	26	0	0	47	11	0	100
Enclosed-type facilities (n = 14)								
average conc. (μg kg^{-1} d.w.)	3000	1500	45	2400	7100	5100	280	1300
median conc. (μg kg^{-1} d.w.)	1800	90	45	1100	9200	1000	150	430
maximum conc. (μg kg^{-1} d.w.)	10000	5200	60	14000	10000	15000	750	4700
minimum conc. (μg kg^{-1} d.w.)	20	30	30	10	40	30	60	30
detection frequency (%)	57	50	14	71	36	71	29	100

[a] Significant difference between Open- and enclosed-type facilities ($p < 0.05$). Statistical analysis: see Section 2.6.

could not be collected in some sampling events or facilities (e.g., the RM sample was not available in Facility G's cold season; SFPs in Facility G retained the highest concentrations of some AMs in all samples of end products).

Across all AM classes, the detection frequencies for MRT in the end products (MCs and SFPs; 100%) and input materials (ESFs and RMs; 96%) were unexpectedly very high (Table 2; Table S6). Though their concentrations were moderately lower (up to 4700 μg/kg), than other AM classes. MRT is administered to piglets (approximately ≤70 kg b.w.) and growing sows (approximately 60–120 kg b.w.) as a feed additive at a maximum level of 30 mg/kg in Japan. However, we could not find a similar report in the literature (e.g., Table 3), that indicated the unique and indigenous usage of MRT in Japanese swine farms [44]. Furthermore, the detection frequencies for TCs in the end products (MCs and SFPs) were relatively high (52%), followed by those for LCM (42%), TML (36%), FQs (36%), MLs (30%), TMP (12%), and SAs (6.0%) (Table 2). Considering the abovementioned residue levels and detection frequencies, manure composts from Japanese swine farms contain a variety of veterinary drugs, occasionally reaching high concentrations of up to 10 mg/kg.

Interestingly, the detection frequencies and residue levels of SAs (mainly SMX) were lower than those of TMP in both end products and input materials (Table 2; Table S6), whereas SAs and TMP were used in combination as veterinary drugs with a concentration ratio of 5:1. In contrast, most samples of raw wastewater collected from the same farms retained SMX at higher levels than TMP [44]. These results imply that SMX is more distributable to the liquid phase in the primary treatment (solid–liquid separation) of swine waste. Information on the physicochemical parameters of SMX and TMP is limited. However, the water solubility ratio of TMP to SMX was 1:4 on the mol fraction scale (1:3.49 on a weight basis; [31]).

Table 3 shows the maximum AM concentrations reported in swine feces, manure, and their treatment products. Interfarm comparisons of AM concentrations from various countries and treatment facilities of swine wastes are mostly cumbersome because of the limited number of studies, differences in the type and usage of purchased AMs, geographic variations in usage patterns, administered animal head rates in farms, differences in manure treatment methods, and sample types. Moreover, several countries have implemented strict controls or restrictions on the use of AMs (especially growth promoters) in livestock; however, recent data are limited. Thus, the data in this study were compared to predominantly available data from the 2010 s. A previous study reported that the most common AM residues were studied in swine waste and that their products were TCs and SAs (Table 3, [32]).

Among the TC classes, the TC concentrations in our study were extremely lower than most of those reported from European countries and China. This is because commercial TC is not registered as a veterinary drug and a feed additive in Japan, and it may have originated as an impurity from other commercial TCs [44]. Meanwhile, the concentrations of OTC, CTC, and DOXY from Germany [45] and China [10,13] were reported to be as high as several hundred mg/kg (Table 3). These concentrations were at least one order higher than the detected maximum concentrations in the present study. Except for the abovementioned reports, our results for OTC, CTC, and DOXY were within the range of residue concentrations reported in other countries.

The maximum residue levels of SAs in this study were likely lower than most of those reported values in Table 3. This discrepancy likely stems from differences in the treatment processes for swine waste and specific compounds used in Japan compared to other countries. In Japan, SMX was found to be present in liquid swine waste (wastewater) due to primary treatment, resulting in its extremely low concentrations and detection frequency in compost. In contrast, the swine waste from other countries listed in Table 3 consisted of a combination of solid and liquid components (manure) or feces. On the other hand, sulfadiazine (SDZ) and sulfamethazine (SMZ; sulfadimidine) were the major SAs found in pig manures in Europe and China (Table 3). However, SDZ is not purchased as an antibiotic for livestock in Japan (only small quantities are used for pet animals; [30]). Meanwhile, the purchase quantity for SMZ in 2019 in Japan was 257.2 kg (as an activated substance), which was approximately two orders of magnitude lower than that for SMX (44389.0 kg) and SMMX (34061.2 kg; [30]). In addition, SDZ and SMZ were not used in the swine farms surveyed in this study [44].

Among FQs, the maximum residue levels for ERFX and CPFX (a metabolite of ERFX) in this study were relatively lower than those previously reported [19,45,51], but the NFLX levels recorded herein were somewhat comparable to those reported by Zhao et al. [51] and Li et al. [19]. There is very limited information available about the other AMs, including MLs, measured in this study (Table 3). Among MLs, the TS levels were as high as 7700 and 1900 μg/kg in Europe and China and were at least 10-fold higher than those in this study, whereas the TMS concentrations reported in the present study (up to 14000 μg/kg in MC) were extremely higher than those reported by Rasschaert et al. [34] (up to 200 μg/kg, Table 3). Zhou et al. [53] reported up to 17000 μg/kg of LCM in piglet feces, which is comparable with our observations. Similarly, TMP concentrations reported elsewhere from Germany (200 μg/kg; [45]) and China (250 μg/kg; [53]) were of the same orders of magnitude as that reported in the present study. In contrast, the TML residue levels in this study were several orders higher than those reported in Germany and Belgium [34,45]. The maximum AM concentrations in the end products (MCs and SFPs) in this study were higher

Table 3
Global comparison of AM maximum residual levels in swine feces, manure and composts with the present study and earlier reports.

Country	Sample matrix	TCs				FQs					
		TC	OTC	CTC	DOXY	ERFX	CPFX	NFLX	MBFX	DNFX	OBFX
Netherlands	Gut feces[a]	n.e.[d]	1500	n.e.	95000	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
Belgium	Manure[a]	n.e.	2000	n.e.	23000	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
Belgium	Manure[a]	26	3900	60	14000	34	8.1	n.e.	110	n.e.	n.e.
Austria	Manure[b]	23000	29000	46000	n.e.	130–750		n.e.	n.e.	n.e.	n.e.
Germany	Manure[b]	300000	210000	55000	380000	4700	n.e.	n.e.	n.e.	n.e.	n.e.
Germany	Digestates[b]	2100	<MQL	900	11000	300	n.e.	n.e.	n.e.	n.e.	n.e.
Spain	Manure[a]	n.e.	720	560	1400	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
USA	Manure[a]	410		1000	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
China	Feces[b]	n.e.	59000	21000	14000	33000	34000	5500	n.e.	2900	n.e.
China	Feces[a]	31000	57000	22000	n.e.	2200	960	3200	n.e.	n.e.	n.e.
China[e]	Feces[b]	9300	1700	98000	2200	17	9	48	n.e.	n.e.	n.e.
China	Manure[a]	44000	180000	27000	n.e.	n.e.	4300	n.e.	n.e.	n.e.	n.e.
China	Manure[a]	98000	350000	140000	37000	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
China	Manure[b]	13000	19000	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
China	Manure[b]	1200 [f]	59000	21000	1400 [f]	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
China[g]	Manure[b]	95	19	81	95	<MDL	200	8	n.e.	n.e.	n.e.
China[f]	Compost[b]	55	71	334	74	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.
China[g]	Compost[b]	26	11	32	24	<MDL	41	490	n.e.	n.e.	n.e.
Japan	Compost[a]	15	13	280	n.e.	n.e.	6	n.e.	n.e.	n.e.	n.e.
Japan	ESF/RM[b]	250	24000	1500	2000	320	150	2800	480	<MDL	4100
Japan	MC/SFP[b]	830	3700	9500	910	170	80	3600	310	<MDL	2900

Country	Sample matrix	**SAs**					**MLs**		**Others**				**Reference**
		SMX	**SMMX**	**SDMX**	**SDZ**[c]	**SMZ**[c]	**TS**	**TMS**	**LCM**	**TML**	**TMP**	**MRT**	
Netherlands	Gut feces[a]	n.e.	n.e.	6	216	n.e.	7700	n.e.	2	4	n.e.	n.e.	[3]
Belgium	Manure[a]	n.e.	n.e.	n.e.	3000	n.e.	<MDL	n.e.	n.e.	n.e.	6	n.e.	[38]
Belgium	Manure[a]	n.e.	n.e.	n.e.	1400	3.0	5600	220	3200	120	4.3	n.e.	[34]
Austria	Manure[b]	n.e.	n.e.	n.e.	n.e.	20000	n.e.	n.e.	n.e.	n.e.	<MDL	n.e.	[25]
Germany	Manure[b]	n.e.	n.e.	500	7300	23000	6400	n.e.	n.e.	1400	200	n.e.	[45]
Germany	Digestates[b]	n.e.	n.e.	<MQL	900	<MQL	<MQL	n.e.	n.e.	<MQL	<MQL	n.e.	[45]
Spain	Manure[a]	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[7]
USA	Manure[a]	n.e.	n.e.	n.e.	n.e.	400	n.e.	n.e.	240	n.e.	2.5	n.e.	[6]
China	Feces[b]	840	4100	n.e.	140	1700	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[51]
China	Feces[a]	2100	4800	n.e.	n.e.	n.e.	1900	n.e.	n.e.	n.e.	n.e.	n.e.	[19]
China[e]	Feces[b]	n.e.	4000	n.e.	n.e.	250	n.e.	n.e.	17000	n.e.	250	n.e.	[53]
China	Manure[a]	5700	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	3800	n.e.	n.e.	n.e.	[13]
China	Manure[a]	n.e.	n.e.	n.e.	7100	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[10]
China	Manure[b]	7600	n.e.	n.e.	4900	6200	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[14]
China	Manure[b]	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[33]
China[g]	Manure[b]	6	5	n.e.	13	<MDL	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[47]
China[f]	Compost[b]	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[33]
China[g]	Compost[b]	<MDL	4	n.e.	3	<MDL	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	[47]
Japan	Compost[a]	35	210	<MDL	n.e.	n.e.	n.e.	n.e.	n.e.	n.e.	<MDL	n.e.	[28]
Japan	ESF/RM[b]	100	<MDL	<MDL	n.e.	n.e.	270	130000	9600	26000	170	3500	Present study
Japan	MC/SFP[b]	60	<MDL	<MDL	n.e.	n.e.	490	14000	10000	15000	750	4700	Present study

Abbreviations: TCs – Tetracyclines; FQs – Fluoroquinolones; SAs – Sulfonamides; MLs – Macrolides; TC – Tetracycline; OTC – Oxytetracycline; CTC – Chlortetracycline; DOXY – Doxycycline; ERFX – Enrofloxacin; CPFX – Ciprofloxacin; NFLX – Norfloxacin; MBFX – Marbofloxacin; DNFX – Danofloxacin; OBFX – Orbifloxacin; SMX – Sulfamethoxazole; SMMX – Sulfamonomethoxine; SDMX – Sulfadimethoxine; SDZ - Sulfadiazine; SMZ - Sulfamethazine/Sulfadimidine; TS – Tylosin; TMS – Tilmicosin; LCM – Lincomycin; TML – tiamulin; TMP – trimethoprim; and MRT – Morantel.
<MDL – below method detection limit
<MQL – below method quantification limit.
[a] Results represented in μg kg^{-1} wet wt.
[b] Results represented in μg kg^{-1} dry wt.
[c] Not analyzed in the present study
[d] Not estimated
[e] Maximum value of average in each growth stage and locations
[f] Maximum value of average in each locations
[g] Maximum value of average in each locations

than those in manure composts reported previously (Table 3). However, comparable residue levels of TCs were previously reported in Japan by Yoshizawa et al. [48] (10^1–10^4 μg/kg).

3.2. Relationship of AM residues to purchase quantities and composting methods

In same farms, the purchase quantities of AMs had a notable impact on the residue levels in wastewater (liquid waste after primary treatment; [44]). However, in case of end products of composting facilities, such as matured compost, the relationship between the purchase quantities and residue levels of AMs was weaker (Fig. 1). A significant positive correlation ($p < 0.05$) was observed only for FQs, indicating that reduction in usage quantities is effective for reducing the residue levels of FQs in composts. For SAs, LCM, and TMP, occasional increasing trends in their residual levels with their purchase quantities were observed, although these relationships were statistically insignificant ($p > 0.05$). Meanwhile, for TCs, MLs, and TML, these relationships were weak, and AM residue concentrations were considerably low in some samples at high purchase quantities. This suggests that residues of AMs

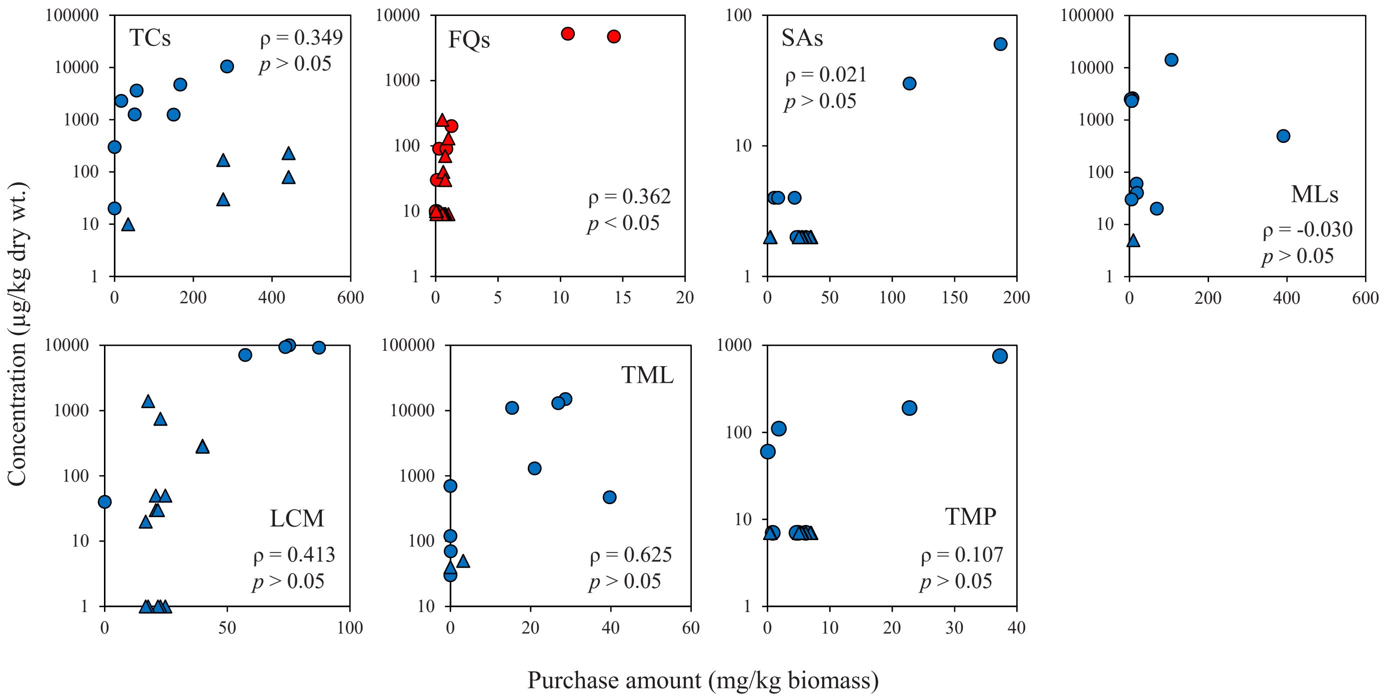

Fig. 1. Relationships between the purchase quantities and residue concentrations of AMs in mature composts (MCs, symbol: circle) from open-type facilities and secondary fermenting composts (SFPs, symbol: triangle) from enclosed-type facilities. TCs, tetracyclines; FQs, fluoroquinolones; SAs, sulfonamides; MLs, macrolides; LCM, lincomycin; TML, tiamulin; TMP, trimethoprim; and MRT, morantel.

other than FQs in swine manure composts reflect not only purchase quantities but also other factors, such as the facility-wise variation in removal efficiencies of these AMs during composting [12,26,47].

Interestingly, the residue levels of most AMs, especially TCs, in the end products of open-type facilities were lower than those in the end products of enclosed-type facilities, even with similar purchase

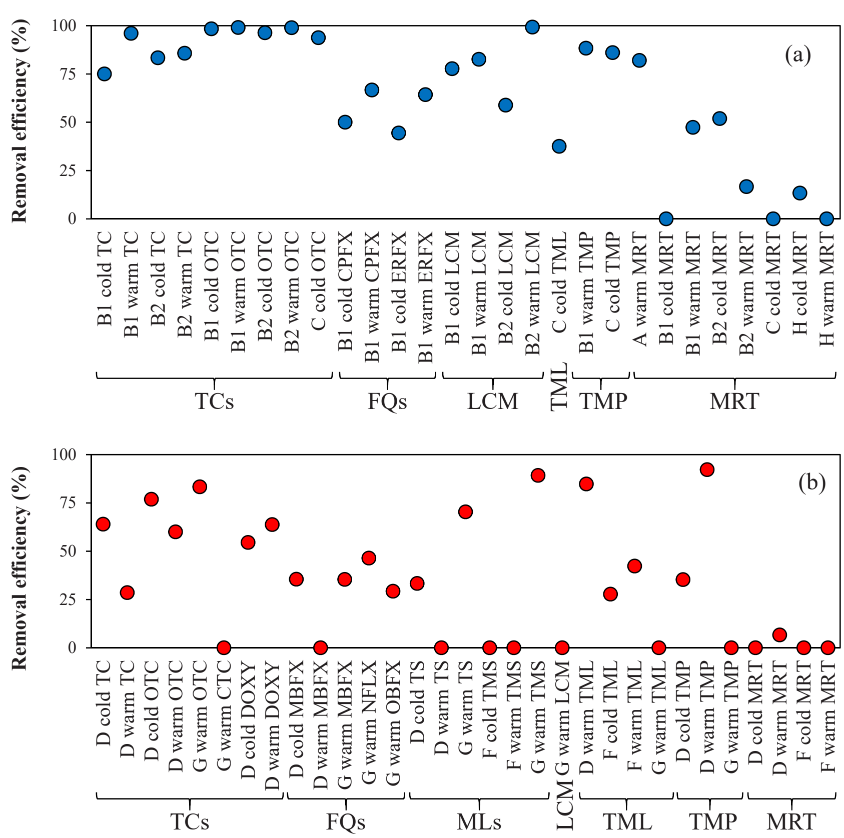

Fig. 2. Removal efficiencies of AMs during composting in open-type (a) and enclosed-type (b) facilities. The *x*-axis indicates the farm and site ID, season, and AMs. TCs, tetracyclines; FQs, fluoroquinolones; MLs, macrolides; LCM, lincomycin; TML, tiamulin; TMP, trimethoprim; and MRT, morantel.

quantities (Fig. 1). The residual concentrations of TCs, SAs, MLs, TML, and TMP in MCs were significantly lower ($p < 0.05$) in the end products of open-type facilities than in the end products of enclosed-type facilities. However, comparable residue levels for FQs, LCM, and MRT were observed (Table 2). Furthermore, the detection frequencies of AMs, except for LCM and MRT, were lower in the end products of open-type facilities than in those of enclosed-type facilities (Table 2). Of these AM classes, the purchase quantities of MLs and TML were lower in farms using open-type compositing methods than in those using enclosed-type composting methods (Fig. S1), indicating that the differences in those AM concentrations between open- and enclosed-type composting methods were also partially associated with their usage patterns. However, the purchase quantities were comparable between farms using open- and enclosed-type composting methods for the remaining AMs (Fig. S1). These results corroborate the reason why a weaker relationship was observed between the residual levels for some AMs in the end products and their purchase amounts (Fig. 1). In other words, the removal efficiencies for several AMs, such as TCs, SAs, and TMP, in open-type composting were higher than those for AMs in enclosed-type composting, whereas the removal efficiencies in both composting methods were comparable for FQs, LCM, and MRT.

3.3. Removal efficiencies

The removal efficiencies for AMs were calculated using their residual levels in the input materials (ESFs or RMs) and end products (MCs or SFPs) (Fig. 2). As input materials could not be collected from all facilities and in all seasons, AMs were detected in a limited number of samples. Thus, composting facilities where AMs were detected in the input materials on a given sampling date were only selected for calculating removal efficiencies. In some cases, the removal efficiency may have been underestimated or overestimated because the AM residue levels in some input materials were insufficient (showed low concentrations) for understanding the full range of removal efficiencies. Besides, most importantly, input materials and end products in each facility and season were sampled on the same day; generally, composting processes take approximately 1 to several months for open-type facilities and approximately 10–20 days for enclosed-type facilities. Therefore, given the above facts, the removal efficiency of AMs in composting must be considered carefully.

Nevertheless, the estimated overall removal efficiencies of AMs during open-type composting (Fig. 2a) were higher than those during enclosed-type composting (Fig. 2b). Furthermore, the removal efficiencies during enclosed-type composting varied greatly within the same AM classes, and no AM removal was observed in some cases (Fig. 2b). These findings and presence of relatively higher residue levels indicate that the removal of AMs might be low and unstable during composting in enclosed-type facilities. Although the reason remains unclear, a possible explanation could be associated with the maturity of composts. In this regard, Kobashi et al. [17] reported that composts from enclose-type facilities retain easily degradable organic matter at higher rates than those from open-type facilities. This insufficient maturity condition may be associated with the lower degradation of AMs [28]. Further detailed studies are required to unravel the reasons behind low and unstable AM degradation during enclosed-type composting.

Among different AM classes, the removal efficiencies for MRT in most cases were the lowest and nearly zero in both composting methods (Fig. 2), indicating that it was relatively resistant to degradation during composting. To the best of our knowledge, this study is the first to report on the residue and degradability of MRT during composting. Meanwhile, the removal efficiencies for FQs were moderate (44–67% in open-type facilities; 0–46% in enclosed-type facilities); however, the detected FQ compounds differed between the two composting methods (Fig. 2a & b). These lower removal rates of MRT and FQs led to their comparable concentrations in the end products between open- and enclosed-type composting (Table 2).

The removal efficiencies for FQs while composting varied widely among previous reports. For instance, Zhang et al. [50] reported that various FQs were more persistent than other ABs during composting. Similar to our results, removal efficiencies of 17–31% were reported for CPFX in a previous study [35]. In contrast, Liu et al. [22] and Cheng et al. [11] reported relatively higher removal efficiencies for CPFX (approximately 90%). Meanwhile, Lin et al. [20] observed that copper promoted FQ removal under mesophilic conditions of composting in lab-scale experiments, suggesting that the coexisting residual copper can influence the variation in removal efficiencies for FQs.

High removal efficiencies for TCs were observed in open-type composting (Fig. 2a; 75–99%). Wang et al. [42] achieved up to 89% OTC degradation in thermophilic composting methods using swine manure. Similar observations for TCs were reported elsewhere [11,22,35]. In contrast, the removal efficiencies in enclosed-type composting (0–83%) were lower and more unstable than those in open-type facilities (Fig. 2). Similarly, LCM and TMPs were likely to be removed in greater amounts in open-type composting (Fig. 2a), although data in this regard are limited. Such a difference may affect residue levels in end products from open- and enclosed-type facilities (Table 2).

Interestingly, most AMs in open-type composting showed lower removal efficiencies in the cold season than in the warm season in the same facilities (Fig. 2a), whereas a clear trend was not observed in enclosed-type facilities (Fig. 2b). Such differences between composting methods may be explained by the effect of ambient temperature. Composts in open-type facilities are directly exposed to a wide area of ambient air, resulting in a decline in the temperature in the top layer of compost in the cold season compared with that during warmer periods. In contrast, composts in enclosed-type facilities are less influenced by ambient temperature because of heat-insulated cylinders and heated aeration air in some cases. Similar trends for SAs and FQs were observed in a study on industrial-scale composting [22].

The temperature during composting is one of the key factors for removing some AM classes from livestock manure [1,21,40,41,8]. Wang et al. [40] found increasing removal efficiencies for SDMX with increasing temperature. Similarly, Liu et al. [21] reported that temperature was a significant factor in the dissipation of SMZ and SMX. Arikan et al. [1] concluded from the results of lab-scale experiments that the concentrations of CTC and its epimer and isomers decreased faster at compost temperatures of 55 °C than at 25 °C.

3.4. Intrafacility variations

In most of the studied facilities, we conducted biannual surveys to understand temporal variations during warm and cold seasons. The ratio based on the residual concentrations in different seasons (cold season/warm season) in the end products (MCs and SFPs) is shown in Fig. 3. This ratio for MRT was nearly 1, suggesting minimal seasonal variation, likely due to consistent usage of this AM as a feed additive. Both residue levels and purchased quantities of FQs also showed limited seasonal variation. In most facilities, the ratio for TCs was over 1, indicating higher residue levels in cold season. This can be explained by a seasonal usage pattern, with residual high quantities of TCs during the cold season in most farms. Similar to TCs, seasonal variations in the residue levels of MLs, LCM, and TML mostly aligned with the purchase quantities. These findings suggest that the purchase quantity significantly influences the AM residue levels in MCs in a facility.

Facilities B1 and B2 were surveyed seven times while facility F1 was surveyed four times on different dates (Table 4), to understand temporal intrafacility variations in AM residue levels in the end products (MCs and SFPs). OTC usage in Facilities B1 and B2 was terminated during this sampling period, whereas other AMs were used as usual. Moreover, no alternative drug to OTC was administered. Hence, AMs other than TCs in the end products from Farms B1 and B2 are discussed here (the fate of TC residues after OTC withdrawal is discussed in Section 3.5).

The MRT concentrations in each facility were relatively consistent

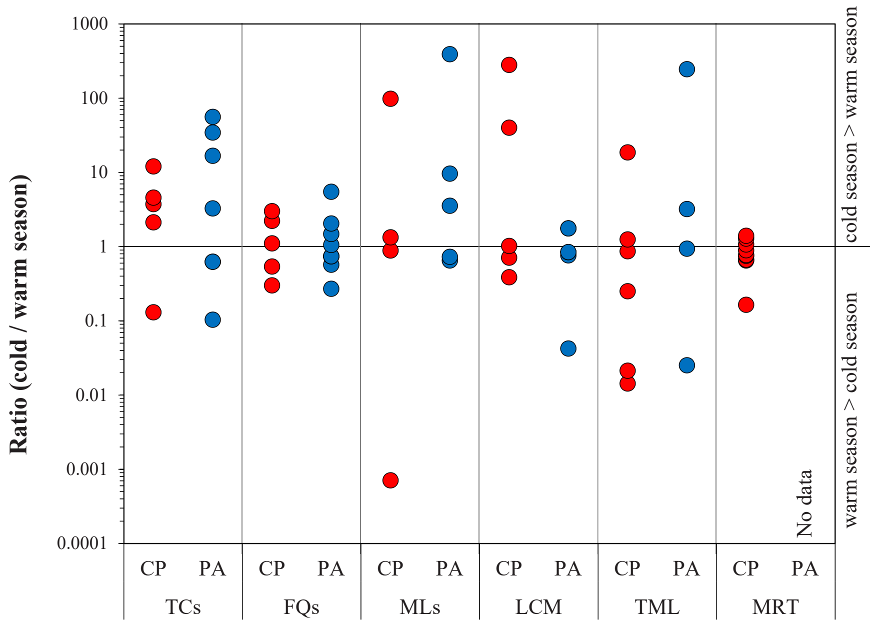

Fig. 3. Ratio of AM concentrations in the end products (MCs and SFPs) and purchase quantities in same farms during cold and warm seasons. There were no data for SAs and TMP because they were detected from limited samples in the end products of composts. There were no data for purchase quantities of MRT because it is used as a feed additive. CP, composts (end products); PA, purchase amounts; TCs, tetracyclines; FQs, fluoroquinolones; MLs, macrolides; LCM, lincomycin; TML, tiamulin; and MRT, morantel.

Table 4
Intrafacility variations of AM residue concentrations in end products from facility B1, B2, and F1 at different sampling times.

	FQs	MLs	LCM	TML	MRT
Facility B1 (n = 7)					
average conc. (μg kg^{-1} d.w.)	100	-	430	-	1400
median conc. (μg kg^{-1} d.w.)	70	-	170	-	1300
maximum conc. (μg kg^{-1} d.w.)	250	-	1400	-	1900
minimum conc. (μg kg^{-1} d.w.)	30	-	20	-	940
detection frequency (%)	71	0	86	0	100
Facility B2 (n = 7)					
average conc. (μg kg^{-1} d.w.)	-	-	110	-	1200
median conc. (μg kg^{-1} d.w.)	-	-	30	-	1000
maximum conc. (μg kg^{-1} d.w.)	-	-	280	-	2300
minimum conc. (μg kg^{-1} d.w.)	-	-	30	-	530
detection frequency (%)	0	0	43	0	100
Facility F1 (n = 4)					
average conc. (μg kg^{-1} d.w.)	-	2300	-	12000	430
median conc. (μg kg^{-1} d.w.)	-	2400	-	12000	430
maximum conc. (μg kg^{-1} d.w.)	-	2600	-	15000	480
minimum conc. (μg kg^{-1} d.w.)	-	1800	-	9100	380
detection frequency (%)	0	100	0	100	100

(Table 4). Similarly, the residue concentrations of MLs and TML in Facility F1 varied across different sampling dates. This indicates consistent use of these AMs in the farm and limited degradation during composting, possibly due to the enclosed-type facility. In contrast, the concentrations of FQs in Facility B1 and LCM in Facilities B1 and B2 varied on different sampling events (Table 4). These concentrations in near-input materials (ESF) varied across different sampling events (Table S5; minimum–maximum: 50–470 μg/kg, 710–4300 μg/kg, and 40–1300 μg/kg, respectively). In addition, seasonal variations in the removal efficiencies for FQs and LCM during composting in these facilities were recorded (Fig. 2). These results imply that within each facility, the residue levels of AMs in swine manure composts can differ widely altered depending on AM usage and/or removal efficiencies during composting.

3.5. Fate of TCs upon discontinuation of OTC usage

To the best of our knowledge, no real farm study has so far investigated the fate of AMs in composted manure after completely stopping their usage. Therefore, residual concentrations of TCs in two open-type facilities were measured for 12 months after the cessation of OTC usage (Facilities B1 and B2) to understand the dynamics of TCs during composting. This farm had not used TCs other than OTC. Moreover, these facilities practiced windrow-type composting with mechanical turning systems, and the composting period lasted for several months.

The dynamics of residual TCs in ESFs and MCs are shown in Fig. 4. The residual concentrations in nearly all input materials (ESFs) from both facilities logarithmically decreased and were not detected at 6 and 12 months after OTC withdrawal in Facilities B1 and B2, respectively. Similar descending trends were observed in wastewater from pigsty outlets and the inlet of a treatment facility in Farm B [44], indicating that AM withdrawal effectively reduced AM residues in swine waste.

The residue concentrations in the end products (MCs) were approximately two orders of magnitude lower than those in the input materials (ESF) before OTC withdrawal (Fig. 4) because of the relatively higher removal efficiencies for TCs in open-type composting (Fig. 2). The concentrations of TCs in MCs from Facility B1 were relatively consistent until 4 months after OTC withdrawal (Fig. 4a), whereas those in MCs from Facility B2 were sparsely detected at 0 and 4 months (Fig. 4b). Considering the composting periods of several months at these facilities, residues in composts may reflect OTC usage before withdrawal. After 4 months, TCs were not detected in the MCs from both facilities, indicating that the decrease in the AM residues in the end products after withdrawal was delayed depending on the composting periods in the facilities.

4. Conclusion

Livestock manure is an important source of natural fertilizer and plays a crucial role in sustainable food systems. In Japan, swine solid waste is primarily composted after separating the liquid phase. It is then reused as an organic fertilizer and/or an ameliorant. This study evaluated the residue status and patterns of 19 AMs in swine manure composts from 12 facilities in Japan. It also examined the relationship between AM residue levels and their purchasing quantities and removal efficiencies. Importantly, the post-usage dynamics of TCs in composts were evaluated on a farm where their usage was discontinued to understand their fate. The composts from Japanese swine farms occasionally contained high concentrations of various types of AMs, reaching levels of approximately 10 mg/kg. The residue levels of AMs in composts from same facilities were affected largely by their purchase

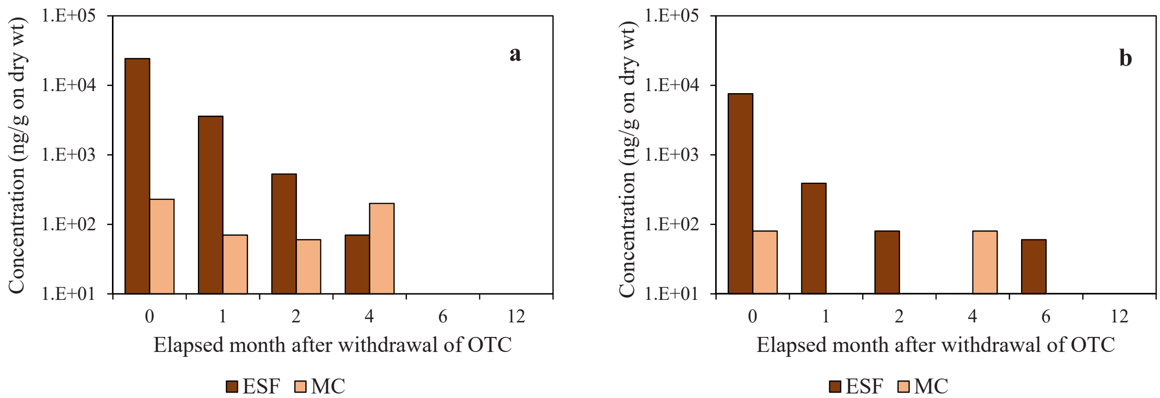

Fig. 4. Fate of residual TCs in the input material (ESF) and mature compost (MC) after withdrawal of OTC administration at Farm B1 (a) and Farm B2 (b).

quantities. However, the variation in removal efficiencies depending on composting methods or AM compounds affected the residue levels from different facilities. In enclosed-type composting, certain AMs exhibited lower and less stable removal efficiencies than those in open-type composting, resulting in higher residue levels in the composts derived from enclosed-type composting. In Japanese swine farms, enclosed-type composting methods are preferred over open-type composting methods as they can be installed in smaller spaces, are more heat efficient and time efficient, and can control the foul odor and greenhouse gases. However, this study reports that open-type composting is more effective in removing AMs during manure composting. Therefore, additional efforts are required to establish appropriate operating conditions for enclosed-type composting to ensure high and consistent AM removal efficiencies.

CRediT authorship contribution statement

M. Watanabe: Methodology, Investigation, Formal analysis, Data curation, Writing – original draft. **P. Goswami:** Data curation, Writing – review & editing. **K. Kure:** Resources, Investigation. **I. Yamane:** Investigation. **S. Kobayashi:** Project administration. **M. Akiba:** Supervision. **K.S. Guruge:** Conceptualization, Methodology, Supervision, Funding acquisition, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.132310.

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