Open-air storage with and without composting as post-treatment methods to degrade pharmaceutical residues in anaerobically digested and dewatered sewage sludge

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HIGHLIGHTS

- Fate of pharmaceuticals and hormones was investigated during open storage of sewage sludge with and without composting.
- No estrogenic hormones or antibiotics were detected after one year of composting.
- 93% of $\sum$ antibiotics in sludge degraded within 180 days of composting.
- Half-life of estrogen in sludge compost was 100 days.
- One year of open-air storage of sludge removed 95% of $\sum$ Antibiotics.

GRAPHICAL ABSTRACT
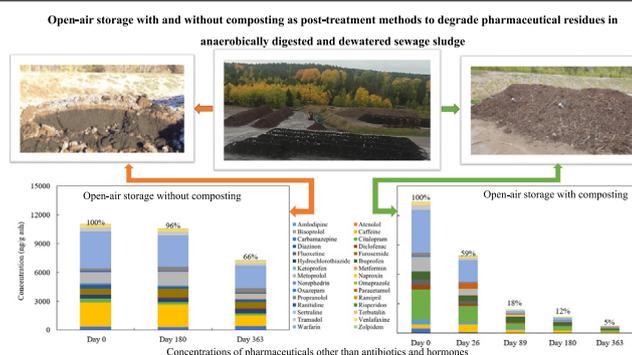
ARTICLE INFO

ABSTRACT

Over a period of 12 months, the fate of three hormones, 12 antibiotics and 30 pharmaceutically active substances (PhACs) was investigated during open-air storage without and with composting of anaerobically digested and dewatered sewage sludge. The effect of oxidation conditions during storage on degradation of hormones and PhACs in the sludge biomass was also examined. Under summer and winter conditions in Uppsala County, Sweden, two field-scale sludge windrows were constructed: open-air storage of sewage sludge windrow without composting (NO-COM)) and open-air storage windrow with composting (COM). NO-COM achieved effective removal of $\sum$ Hormones (85%) and $\sum$ Antibiotics (95%), but lower removal of $\sum$ PhACs (34%), during the study year. The top layers of the sludge pile had significantly lower concentrations of $\sum$ PhACs (3100–5100 ng/g ash) than deeper layers (8000–11,000 ng/g ash). After one year of composting, the degradation in the COM windrow resulted in concentrations of $\sum$ Hormones (<LOD), $\sum$ Antibiotics (<LOD), while the $\sum$ PhCAs was 5% (730 ng/g ash) of initial (13,000 ng/g ash). The half-life of substances during composting in COM was within 7–100 days for all substances except ibuprofen (156 days). The first-order degradation constant (K) was the lowest for ibuprofen (0.0045 day^{-1}) and the highest for oxazepam (0.0805 day^{-1}). In conclusion, composting of sludge was effective in degrading the target hormones, antibiotics, and PhACs.

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1. Introduction

Anaerobically digested and dewatered sewage sludge, a product also known as biosolids, is a sustainable fertilizer and soil conditioner because of its valuable content of organic matter, plant nutrients, and trace elements. Around 45–53% of the digested sludge produced in the European Union and USA is used in agriculture (Dubey et al., 2021; Kelessidis and Stasinakis, 2012). For example, in Sweden, 211×10^3 tons of sewage sludge (dry weight) are produced per year, of which around 39% is spread on agricultural land (SMED, 2018). It has been well documented that sewage sludge contains pharmaceutically active substance (PhACs) and other micropollutants and heavy metals (Bisognin et al., 2021; Fijalkowski et al., 2017; Stahl et al., 2018). Following land application of sludge, pharmaceutical residues and other pollutants may be lost from the soil and can enter surface water via runoff or leaching to the groundwater, or can be assimilated by vegetation or other living organisms (Topp et al., 2008; Wu et al., 2010). It has been shown that pharmaceuticals present in soil can be taken up by plants, constituting a route of involuntary intake of pharmaceuticals which could be a risk for human health (Carter et al., 2014).

Anaerobic digestion of sludge provides stabilization and the additional benefit of reduced biological activity in the biomass (Chen et al., 2016). However, it has been shown that anaerobic digestion is not sufficient for removal of pharmaceuticals with removal efficiencies of >80% reduction for 5 pharmaceuticals out of 23 and > 30% reduction for a further 3 out of 23 while the remaining were reduced by less than 30% (Malmborg and Magnér, 2015). In anaerobic digestion experiment of fecal sludge, Gros et al. (2020) showed that out of 17 substances only two (oxazepam and losartan) showed a removal of ≥50%, seven substances (atenolol, metoprolol, carbamazepine, lamotrigine, venlafaxine, valsartan and lidocaine) were removed at rates of 10–37%. Remaining eight substances were poorly removed (<10%).

Common post-treatment methods for sludge include incineration, composting, and open storage. The latter two methods are used before usage of sludge for land application in agriculture, or use for road construction or as a landfill cover (Guo et al., 2020). Storage of sludge for at least six months is applied to ensure decay of *Escherichia coli* and *Salmonella* spp. in the digested and dewatered sludge. In a laboratory-scale experiment, sludge storage was found to reduce the concentrations of some substances by between 5 and 85%, while the concentrations of 21 substances increased by 4–283% (Cidh, 2014). Effects of sewage sludge storage on fate and behavior of pharmaceuticals is not well documented especially under field conditions.

Composting is a treatment method used to enhance the degree of sanitization of sludge as composting is exothermic reaction process i.e. produces heat which increases the temperature of the substrate to >55 °C for several days, which in its turns destroy the bacteria (Manga et al., 2021). During composting, organic molecules undergo both abiotic degradation, for instance through hydrolysis or oxidation, biodegradation, and evaporation (Haug, 1993). It has been shown that persistent molecules, including PhACs, also undergo these processes and can be effectively degraded (Guo et al., 2020; Wu et al., 2010). A study on a full-scale liquid composting plant for blackwater recorded a 30–80% reduction in PhACs, including substances such as atenolol, metoprolol, citalopram, furosemide, and atorvastatin (Gros et al., 2020). Other PhACs (carbamazepine, lamotrigine, venlafaxine, lidocaine, diazepam, and losartan) showed no or low reduction (<50%) during liquid composting, but propranolol, valsartan, and hydrochlorothiazide showed high overall removal rates (>80%) during the treatment (Gros et al., 2020). In a laboratory study, Vasskog et al. (2009) observed reductions in five PhACs during the first 10 days of a 19-day experiment in which a mixture of sludge, bark, and garden waste was composted. The same study analyzed a number of metabolites and found that concentrations after composting were approximately 10% of those of the parent substances (Vasskog et al., 2009). In a study by Cidh (2014), it was found that 61 days of composting of sludge and animal manure

decreased the mass of PhACs by 4–96% for 28 pharmaceutical compounds, while concentrations of seven compounds increased by 3–193% on a mass basis. However, those studies were performed in the laboratory under controlled conditions. The promising effects of composting on degradation of PhACs at laboratory scale warrant scaling up of compost trials to field scale, to better assess the potential of composting for eliminating PhACs in sludge. The effect of storage on the PhACs content in sludge is not clear and to our knowledge, no previous field study has examined the fate of PhACs during storage of sludge and there are no experimental data on the effects of composting on degradation of PhACs under real environmental conditions in a Northern temperate climate.

The main aim of this study was to assess the fate of PhACs and hormones during post- treatment of digested and dewatered sewage sludge in field-scale experiments under Northern temperate climate conditions. Specific objectives of the work were to: (i) determine the reduction in concentration and mass of PhACs and hormones in mesophilically digested and dewatered sludge during 12 months of open-air storage, ii) investigate the stratification of PhACs and hormones with depth in a sludge windrow during 12 months of open-air storage under environmental conditions, and (iii) investigate the degradation of hormones and PhACs during 12 months of full-scale composting of the dewatered sludge. The hypothesis tested was that better oxidation conditions in the sludge result in high and fast degradation of hormones and PhACs in sludge.

2. Materials and methods

2.1. Set-up of the field experiment

Degradation of PhACs was investigated in mesophilic digested sludge stored in two field-scale windrows mimicking two types of post-treatment conditions: (i) Open-air storage of mesophilic digested and dewatered sludge during 12 months of storage in a Northern temperate climate (NO-COM) and stratification of PhACs at different depths in the NO-COM windrow during storage; and (ii) co-composting of mesophilic digested and dewatered sludge and garden waste during 12 months in Northern temperate climate conditions (COM). Weather data of average daily air temperature and total daily precipitation were obtained from Hovgården weather station for the entire period of the study (31 May −2 June 2018 to 29–31 May 2019).

The Anaerobically digested and dewatered sludge was obtained from Henriksdal wastewater treatment plant in Stockholm, Sweden. The feed to the Henriksdal digester consists of 90% primary sludge and 10% of sludge from the biological and chemical wastewater treatment. The hydraulic retention time of the digester is 14 days and the degree of degradation is 58%. After being digested under mesophilic conditions (37 °C) the sludge was mixed with Sedifloc 683CHH polymer (Kemira) and de-watered by centrifuge. The freshly dewatered sludge was delivered to Hovgården solid waste management station in Uppsala. The TS content in the delivered sludge was 27.6 ± 1.3% upon delivery.

A mass of 76.1 tons of sludge was used to construct the NO-COM windrow, which was kept in the open air under environmental conditions for 12 months. In the COM windrow, 61.8 tons of sludge and 21.7 tons of garden waste (crushed branches) were mixed initially and then mixed/turned once every week during weeks 1–3, once every three weeks during weeks 4–13, and after 6 months of storage. The storage period for both windrows was one year (31 May −2 June 2018 to 29–31 May 2019). Immediately after construction of NO-COM and COM, windrow dimensions, field density, porosity, and total solids (TS) were determined using methods described in the following sections and section SI1 in the Supporting Information (SI) and summarized in Table 1.

2.2. Sampling and sample preparation

Samples from the NO-COM windrow were collected after 0, 180, and 363 days of storage, while samples from the COM windrow were collected 0, 26, 89, 180, and 363 days after start of composting.

Table 1

Dimensions of windrows, weight of sludge, total solids (TS) content, field density, and porosity of the open-air storage without composting (NO-COM) and with composting (COM) windrows immediately after construction.

Windrow	Dimensions (m) (H × W × L)	Weight of sludge (ton)	TS (%)	Density (kg/L)	Porosity (%)
NO-COM	1.6 × 4.8 × 15.5	86.3	27.6 ± 1.3	0.69 ± 0.02	34.4 ± 2.0
COM	3.2 × 6.1 × 16.3	83.5[a]	34.1 ± 6.6[b]	0.38 ± 0.04	64.3 ± 4.0

[a] Consisting of 61.8 tons of sludge and 21.7 tons of garden waste.
[b] TS in the mixture of sludge and garden waste.

Samples were collected from COM as follows: prior to sampling, the material was mixed using a wheel loader and dumped in a windrow. Samples were then collected from seven locations randomly distributed along the surface of the windrow. Each of these seven samples consisted of three sub-samples collected at each location and wood chips and branches with size >1 cm were removed before pooling the samples.

For NO-COM, the sampling on day 0 (at windrow formation) followed the procedure described for COM. Collection of samples at 180 and 363 days was done as follows: material from a 3-m stretch of windrow was removed by wheel loader. In the clean-cut ed. surface of the windrow, samples used for the depth study were then collected at different depths (5–15, 25–35, 45–55, 65–75, and 145–155 cm, measuring from the top surface of the windrow). To analyze the substances in the whole windrow, sludge was consistently sampled in three replicates. Each replicate was composed by mixing (homogenizing) seven subsamples taken from different sites in the clean cut of the windrow according to the sampling scheme shown in Fig. S1 in SI.

2.3. Field tests and analyses of physical and chemical parameters

The temperature in NO-COM and COM was measured continuously, using one temperature sensor (Telldus 433 MHz) placed in NO-COM and three sensors placed in COM (all about 60 cm deep in the windrow). More sensors were used in COM in order to capture the variation in temperature in the windrow during composting. The temperature was measured every five minutes and sent to a data server at Telldus Technologies AB (Sweden). On each sampling day, field density of the windrows was measured in situ and samples of the windrow material were collected for laboratory analyses. These involved determining the concentrations of three hormones, 12 antibiotics, and 30 PhACs with other therapeutic effects, (TS), ash, volatile solids (VS), pH, phosphorus (P), ammonia-nitrogen (NH_4-N), nitrite-nitrogen (NO_2-N), nitrate-nitrogen (NO_3-N), and Kjeldahl-nitrogen (Kj-N). Determination of TS, ash, VS, pH, P, NH_4-N, NO_2-N, NO_3-N, and Kj-N was performed according to the following methods: Total solids content was determined by measuring the weight of sludge samples before and after drying at 105 °C for 24 h (SIS, 2007). Ash was determined by incinerating the dried samples at 550 °C for 4 h (SIS, 2000). Volatile solids content was estimated as the difference between TS and ash content. Other parameters were analyzed by the commercial laboratory ALS Scandinavia AB in Prague, Czech Republic, according to these methods: CSN ISO 11732 and CSN ISO 13395 for NO_2-N, NO_3-N, CSN EN 25663, CSN EN 13342 and CSN ISO 7150-1 for Kj-N, CSN ISO 7150-1 for NH_4-N, CSN ISO 10390, CSN EN 12176:1999, CSN EN 13037, CSN EN 15933, CSN 465735, ÖNORM L 1086–1, US EPA 9045D; US EPA 9040C for pH, and CSN EN 14671 and CSN EN ISO 6878 for P.). Determination of field porosity and density is described in section SI1 in the supporting information.

2.4. Analyses of antibiotics, pharmaceuticals, and hormones

The term PhACs as used here refers to all substances with therapeutic effects other than antibiotics and hormones. The compounds analyzed were three hormones (estradiol, estrogen, 17α-ethinyl estradiol, 12 antibiotics (benzylpenicillin, rifampicin, sulfamethoxazole, ciprofloxacin, clarithromycin, clindamycin, erythromycin, linezolid, metronidazole, moxifloxacin, trimethoprim, and ofloxacine) and 30 PhACs (atenolol, amlodipin, bisoprolol, caffeine, carbamazepine, citalopram, diazinon, fluoxetine, ketoprofen, metformin, metoprolol, norephedrine, omeprazole, oxazepam, paracetamol, propranolol, ranitidine, risperidone, sertraline, terbutaline, tramadol, venlafaxine, zolpidem, diclofenac, furosemide, hydrochlorothiazide, ibuprofine, naproxine, ramipril, warfarin). Chemical properties of these compounds are shown in Table S1. Isotopically labeled internal standards (IS) used in the analyses were diclofenac $^{13}C_6$, hydrochlorothiazide $^{13}C_6$, carbamazepine ^{13}C ^{15}N, and ibuprofind-d_3. (Merck, Darmstadt, Germany).

2.4.1. Extraction of antibiotics

The extraction method used for antibiotics in sludge was a method similar to a previously published method (Gago-Ferrero et al., 2015) with an addition of a separate extraction step using Dimethyl Sulfoxide (DMSO). 0.5 g portions of freeze-dried windrow material were weighed into two 14 mL Falcon tubes, to which a 100 ng IS mixture was added. Then 0.5 mL of 2.0 M $MgNO_3$ solution was added to each of the tubes, and 5 mL acetonitrile and 5 mL of 0.20 M citric acid were added to one tube, while 10 mL DMSO in water (1:1) were added to the other. All samples were extracted for 5 min in an ultrasonic bath and then agitated for 30 min on a shake table. The samples were centrifuged and the supernatant was decanted into a 50 mL Falcon tube, containing 0.20 g Na 2 EDTA, and diluted to 50 mL with deionized water. The resulting 50 mL samples were concentrated and purified using solid phase extraction (SPE) with Oasis HLB cartridges (Waters, Oasis HLB, 200 mg) according to the following protocol: (i) The cartridges were washed with 5 mL methanol and conditioned with 5 mL deionized water, (ii) the 50 mL sample was percolated through the cartridge, (iii) the cartridges were washed with 2 ml MQ water and then eluted with 5 ml of methanol, followed by 5 ml of acetone, (iv) the eluted samples were evaporated to dryness using dry nitrogen, and (v) the dried sample was reconstituted in methanol:water (1:1) containing 0.1% Na 2 EDTA, centrifuged, and transferred to 1.5 mL glass vials for analysis by high performance liquid chromatograph coupled with mass spectrophotometry (HPLC-MS/MS).

2.4.2. Extraction of pharmaceuticals other than antibiotics

Around 0.25–0.5 g samples of freeze-dried windrow material were weighed into 14 mL Falcon tubes, to which 100 ng of IS mixture and 10 mL acetonitrile:dichloromethane (1:1) were added. The samples were treated for 5 min in an ultrasonic bath and then agitated for 30 min on a shake table. The tubes were centrifuged, and the supernatant was decanted into new Falcon tubes and evaporated to dryness under nitrogen flow. The dried samples were reconstituted in 1.0 mL methanol:water (1:1) containing 0.1% Na 2 EDTA, transferred to Eppendorf tubes, and centrifuged at 10,000 rpm or filtered through 0.45 μm spray filter. The samples were finally transferred to 1.5 mL glass vials for HPLC-MS/MS analysis.

2.4.3. Extraction of hormones

The extraction and cleanup procedure for the hormones was adapted from an earlier presented method (Vulliet et al., 2007). Around 0.25 g of freeze-dried windrow material was combined with IS (10 ng of D_5-β-estradiol or D_4-ethinylestradiol) and 14 mL methanol were added

to each sample. The samples were pre-extracted in a microwave extractor for 25 min at 110 °C and the extract was then evaporated to about 2 mL of solution and diluted with 3 mL buffered MQ water (pH <2) before purification and concentration by SPE. This was done using a mixed mode reversed phase cartridge (Isolute ENV + 500 mg, 6 cc, Biotage AB, Uppsala, Sweden) conditioned with methyl tertbutyl ether (MTBE), methanol, and deionized water before application of the sample. The cartridge was washed with 40% methanol solution and the analytes were eluted with a mixture of ethanol and MTBE (1:9). The samples were then evaporated to dryness and reconstituted in a methanol: water mixture (1:1). Finally, the samples were filtered through a syringe filter with particle size 0.45 μm.

2.5. Instrumental analyses of PhACs, hormones, and antibiotics

The masses of the target substances were determined using an HPLC-MS/MS method further developed from a similar method previously published (Gros et al., 2006). All analyses were carried out at the laboratories of the Swedish Environmental Institute (IVL) using a binary Shimadzu AD20 UFLC HPLC system with automatic sample changer and column furnace coupled to an ABSciex API — 4000 mass spectrometer. The samples were analyzed in positive and negative electrospray ionization (ESI) mode. A Waters XBridge BEH C18 column (100 mm × 2.1 mm with 3 μm opening size) was used. The eluents used in the mobile phase were A: 10 mM acetic acid in deionized water and B: methanol. The gradient used was a linear gradient from 0 to 90% methanol for 17 min with a final plateau at 90% methanol for 4 min, before a rapid return to 100% A and a final recovery and equalization period of 2 min. Each analyte was quantified by internal quantification using an eight-point calibration curve (0, 5, 10, 20, 50, 100, 200, and 500 ng).

2.6. Quality control

Limit of detection (LOD) and limit of quantification (LOQ) were determined at a signal to noise (S/N) ratio of 3 and 10, respectively. The LOD and LOQ range for the samples was 0.04–35 ng/g ash and 0.13–116 ng/g ash, respectively (Table S2 in SI). For assessment of recovery, 100 ng of standard PhACs and 40 ng of each of the hormones were added to freeze-dried compost with zero background concentrations of the analytes, determined by chemical analysis. These samples were extracted and analyzed in the same manner as the other samples. The recovery rate was estimated as: Measured concentration/Added concentration. The recovery values were 80–120% except for estrogen (191%) and 17 α-ethinyl estradiol (161%). The concentrations of PhACs and hormones in the samples of windrow material were then corrected for recovery rates.

2.7. Calculations and statistical analysis

Concentration of hormones, antibiotics and PhACs were calculated on ash basis to allow estimation of total mass loss of the substances, i.e. the mass loss that considered degradation of organic matter in the substrate. The biodegradation of organic matter in windrow material was calculated as using Eq. (1) reported by Haug (1993):

$$Biodegradation = 1 - \left(\frac{Ash_i}{VS_i} \div \frac{Ash_f}{VS_f} \right) \quad (1)$$

where VS and ash content are expressed as percentage of TS, and i and f denote the initial and final measurements. The degradation of the substances were modeled using first-order exponential decay models as using Eq. (2) reported by Cheng et al. (2019):

$$C_t = C_0 e^{-Kt} \quad (2)$$

where C_0 is initial concentration of the substance, C_t is the concentration at time (t), and K is the first-order decay constant (day^{-1}). First order

exponential decay model is a common model used for fitting the degradation of pharmaceuticals in soils and other environmental samples (Al-Khazrajy et al., 2018; Cheng et al., 2019; Li and Zhang, 2010; Yu et al., 2013). We expect that of the behavior of pharmaceuticals in sludge compost ca be similar to this in soil and therefore used the first order exponential decay model to explain the degradation and dissipation of the studied substances in the current study. The models were run using GraphPad Prism version 6.04 for Windows (GraphPad Software, La Jolla California USA). The accuracy of the models was analyzed using R^2 and sum of squares.

Two-way analysis of variance (ANOVA) at 95% confidence level was used to assess the difference in concentrations between: (i) treatments and (ii) different sampling events. All statistical analyses were performed using STATISTICA version 10 (Statsoft Inc., Tulsa, OK, USA). When a statistically significant difference was found, Tukey multiple comparison of means (95% confidence level) was performed.

3. Results and discussion

3.1. Physico-chemical characteristics of sludge during storage and composting

During the entire storage period, the density of the sludge in NO-COM ranged from 0.69 to 1.02 kg/L and porosity from −0.1 to 34.4%. The density of the material was lower in COM (0.38–0.55 kg/L) and the porosity was higher (49–64%). These results indicate that possibilities for oxygen transfer was significantly lower in NO-COM compared with COM, which, according to the hypothesis, could affect the degradation of organic matter, hormones, antibiotics and PhACs in the two treatments.

The temperature in NO-COM sludge at the depth of about 60 cm followed the temperature pattern in the ambient environment, but delayed and averaged (Fig. 1). Storage of sludge in NO-COM did not seem to change TS, VS, or ash content during the first 180 days of storage, and low degradation rate was estimated ($p > 0.05$) (Table 2). There was high variation in TS, VS, and ash content measured at day 363, which made it difficult to assess whether the sludge had degraded after one year of storage. The Kj-N content in NO-COM during storage was in the range 54,000–65,000 mg/kg, with no significance difference between the days of measurement (Table 3). The samples collected from the entire NO-COM did not show nitrification (Table 3).Yet, the surface samples (i.e samples collected from the top 5–15 cm) of NO-COM showed some levels of nitrate (320 mg/kg) on day 363. This indicates better conditions for nitrification i.e. higher oxygen levels at the surface of the windrow compared to the lower sections of it.
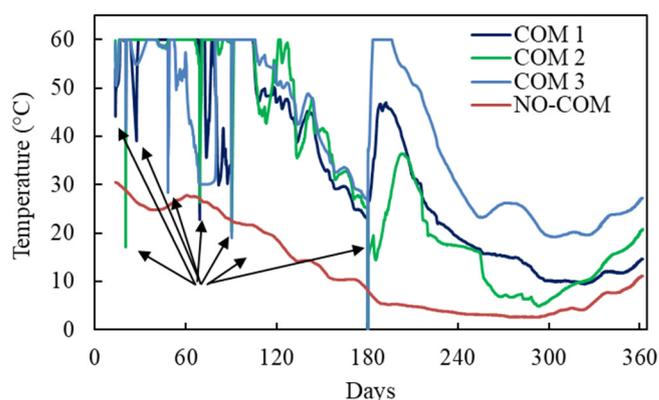
Fig. 1. Variations in temperature (°C) about 60 cm below the surface in the open-air storage without composting (NO-COM) and with composting (COM) windrows during the 363 days of operation (31 May 2018–30 May 2019). COM 1–3 denote temperature profiles in the COM in three different locations along the windrow. The arrows indicate drops in temperature measured when temperature sensors were removed from the windrow when it was mixed. This figure was reported in our technical report by Jönsson et al. (2020).

Table 2
Concentrations of total solids (TS), volatile solids (VS), ash, field density, porosity, and biodegradation of material in the open-air storage without composting (NO-COM) and with composting (COM) windrows. Values shows are mean ± standard deviation (Std). Number of replicates (n) = 3 for all samples unless otherwise stated.

Treatment	Day	TS		VS (% TS)		Ash (% VS)		Density (Kg/L)		Porosity (%)		Biodegradation (%VS)	
		Mean	Std	Mean	Std	Mean	Std	Mean	Std	Mean	Std	Mean	Std
NO-COM	0	28	1	67	1	33	1	0.69	0.02	34	2	0	0
	180	29	12	67	8	34	8	1.06	0.08	0	7	-4	44
	363	21	10	47	22	53	22	1.02	0.05	4	6	48	34
COM	0	34	7	70	7	30	7	0.38	0.04	64	4	0	0
	26	51	11	63	3	37	3	0.44	0.08	61	8	21	38
	89	56	13	57	6	43	6	0.47	0.10	59	9	38	30
	180	66	1	55	5	45	5	0.47	0.03	62	2	43	25
	363	55	7	51	2	49	2	0.55	0.07	49	7	54	11

The temperature variation in the COM windrow during the composting process followed three stages: a heating or initial mesophilic phase (37–40 °C), a thermophilic phase (>55 °C), and a cooling phase (Fig. 1). The mesophilic phase cannot be seen in Fig. 1 as it had already passed when the temperature sensors were ready on day 12, when they also were installed . Immediately after installation, the temperature increased rapidly to exceed 60 °C by day 14. This was the maximum temperature that could be measured by the sensors, and thus the actual temperatures probably have been higher than 60 °C from day 14 to 110. When the compost was mixed on days 19, 26, 47, 68, 89, and 180, the temperature sensors were removed before mixing and then inserted again after mixing, which explains the decline in temperature during these specific days, as indicated by the arrows in Fig. 1. The temperature in COM stayed above 60 °C until day 110, reflecting active biological degradation of organic matter in the compost. After 110 days the temperature started to decline gradually, but increased again after mixing on day 180. The windrow was not mixed between 89 and 180 days, which could have led to zones of limited oxygen supply and dryness in the compost, resulting in a decline in bacterial activity. The VS content in the COM material declined from $70 \pm 7\%$ on day 0 to $51 \pm 2\%$ after 363 days of composting (Table 1). The degradation of organic matter in COM reached $54 \pm 11\%$ after 363 days of composting. The temperature profile and degradation rate of organic matter suggested compost maturity (Toledo et al., 2019). The high temperature over an extended period indicated that the compost was likely sanitized as recommended by the Swedish environmental protection agency (NV, 2013). The highest average (24 h) air temperature was 26 °C measured in July, 62 days after start of experiment (Fig. S2). The lowest average temperature was −17 °C measured 235 days after start of the experiment.

Also, nitrification activity occurred in COM. The NO_3-N content in the COM material increased from non-detectable levels on day 0 to 2200 ± 600 mg/kg TS at day 363, in parallel with a decline in NH_4-N from 4000 to 6300 mg/kg TS during the first 26 days to 2700 mg/kg TS at day 363 (Table 3) and pH in the compost, confirming nitrification. Nitrite in COM was only detected in low concentrations in days 89 and 180 (Table 3) no significant changes in the concentration of Kj-N on TS basis was observed during the entire study period. However, the biodegradation of 54% of the VS means that the final TS of compost 32% less than the initial. Thus similar Kj-N on TS basis indicates that about one third of the Kj-N has been lost during the compost treatment. Mixing of COM led not only to better oxygen transport, but also to lower density and higher porosity compared with NO-COM, allowing better aeration (Table 2). We did not measure loss of greenhouse gases, e.g. NH_3, CH_4, N_2O or other gases from COM.

3.2. Initial concentrations of hormones, antibiotics and PhACs in sludge before treatment

Concentrations of the individual hormones, antibiotics and PhACs, in the freshly delivered sludge (day 0), as measured in NO-COM, are shown in Table S4 in SI. Of the target substances, two hormones, six

antibiotics, and 19 other PhACs were found at detectable levels in the freshly delivered sludge. $\sum$Hormones were 200 ± 100 ng/g ash as measured in the NO-COM and COM windrows. The $\sum$Antibiotics was 1300 ± 300 ng/g ash (Tables S4 and S5 in SI). $\sum$PhACs was $11,000 \pm 2000$ ng/g ash.

Among the target hormones, estrogen was detected at the highest levels (180 ± 100 ng/g ash), while estradiol concentrations were less than 13 ± 0.1 ng/g ash. Fick et al. (2011) previously reported <10 ng/g TS of estrogen, estradiol, and 17a- ethinyl estradiol in sludge from Henriksdal treatment plant, compared with 53, 2.5, and 6.8 ng/g TS in the present study. Levels of estrogen were comparable to the 48–137 ng/g TS reported for digested and dewatered sludge in Seine Aval (Mailler et al., 2017). Among the antibiotics, ciprofloxacin (800 ng/g ash; 62% of $\sum$Antibiotics) and ofloxacine (300 ng/g ash; 23% of $\sum$Antibiotics) recorded the highest concentrations. The dominant PhACs were sertraline (35% of $\sum$PhACs), citalopram (23% of $\sum$PhACs), and metoprolol (10% of $\sum$PhACs), which were present in concentrations of 3900, 2500, and 1100 ng/g ash, respectively. The concentrations of caffeine and paracetamol were low compared with these usually found in wastewater influents. These substances have high solubility and affinity to partition to water and therefore were found at low levels in the sludge. The concentrations found in the current study were comparable to those reported previously for Henriksdal sludge (Fick et al., 2011), except for sertraline (380 vs 1300 ng/g TS in this study), diclofenac (570 vs 130 ng/g TS in this study), and ibuprofen (<LOQ vs 220 ng/g TS in this study).

3.3. Fate of hormones, antibiotics, and PhACs during open-air storage of sludge

First-order exponential decay models failed to explain the degradation of hormones, antibiotics, or PhACs in NO-COM at 95% confidence interval ($p > 0.05$; data not shown). Among the measured hormones, estrogen was the most persistent during open-air storage of sludge in NO-COM. Only 16% of estrogen was removed during the first 180 days, while estradiol was well removed (final concentrations <LOD) during the first 180 days (Fig. 2). After 363 days, about 16% of the initial hormone mass was still present in the sludge, but consisting solely of estrogen (Table S4). Xuan et al. (2008) reported rapid degradation of estradiol, with half-life in non-sterile soil of 0.17 day. Also, 17 α-Ethynylestradiol at 0.1–10 mg/kg were reported to reduce rapidly ($t_{1/2}$ = few hours – few days) in loamy soils at 30 °C, while estrone was reduced at slower rates than the 17 α-Ethynylestradiol (Colucci et al., 2001; Colucci and Topp, 2001).

In total, 14% of $\sum$Antibiotics was degraded during the first 180 days of open-air storage in NO-COM (Fig. 2). After one year of storage, 95% of $\sum$Antibiotics degraded and only 65 ng/g ash remained in the sludge, compared with 1300 ng/g ash measured at day 0 (Table S4). Ciprofloxacin, moxifloxacin, and ofloxacine, which were initially detected at high levels in the sludge (Table S5), degraded at intermediate rates (69, 31, and 55% respectively) during the first 180 days of storage. Interestingly, the analyzed concentration of clindamycin increased during the first

Table 3
Mean pH and concentrations of Kjeldahl nitrogen (Kj-N), ammonium (NH$_4$-N), nitrate (NO$_3$-N), nitrite (NO$_2$-N), and total phosphorus (Tot-P) in the open-air storage without composting (NO-COM) and with composting (COM) windrows. All concentrations are in mg/kg TS except for pH. Values shown are mean ± standard deviation (Std). Number of replicates (n) = 3 for all samples unless otherwise stated.

Treatment	Days	pH		Kj-N		NH$_4$-N		NO$_3$-N		NO$_2$-N		Tot-P	
		Mean	Std	Mean	Std	Mean	Std	Mean	Std	Mean	Std	Mean	Std
NO-COM	0[a]	8.1	0.0	54,000	1600	8600	480	<8	0	<1	0	19,000	2500
	180	8.4	0.1	59,000	7500	3400	500	<1	0	<1	0	23,000	2900
	363	8.6	0.2	65,000	11,000	15,000	1300	<4	0	<1	0	21,000	4700
COM	0[b]	8.0	–	30,000	–	4000	–	<4	0	<1	–	2[c]	–
	26	8.1	0.2	33,000	2700	6300	100	<15	0	<1	0	13,000	1800
	89	7.6	0.1	29,000	2900	4900	750	44	70	33	58	2[c]	–
	180	7.5	0.1	34,000	1900	2000	340	290	230	23	29	17,000	1000
	363	5.7	0.3	25,000	5800	2700	2500	2200	600	1	1	20,000	3900

[a] Values for NO-COM (3 replicates), which was the same sludge as used in COM.
[b] Values for COM were measured after mixing the sludge 3:1 with garden waste (1 replicate).
[c] Outlier values.

180 days of storage, to 570 ± 200 ng/g ash, and then decreased during the 1ast 183 days to 50 ± 20 ng/g ash. This could have been due to transformation of metabolites back to the original compound during storage (Evgenidou et al., 2015). Other possible explanations can be changes in the chemical conditions in the sludge during storage, decrease in the number or mass of particles to which clindamycin was adsorbed, or ion saturation in the electrospray during analysis of the start samples. Generally, there is public concern about the spread of
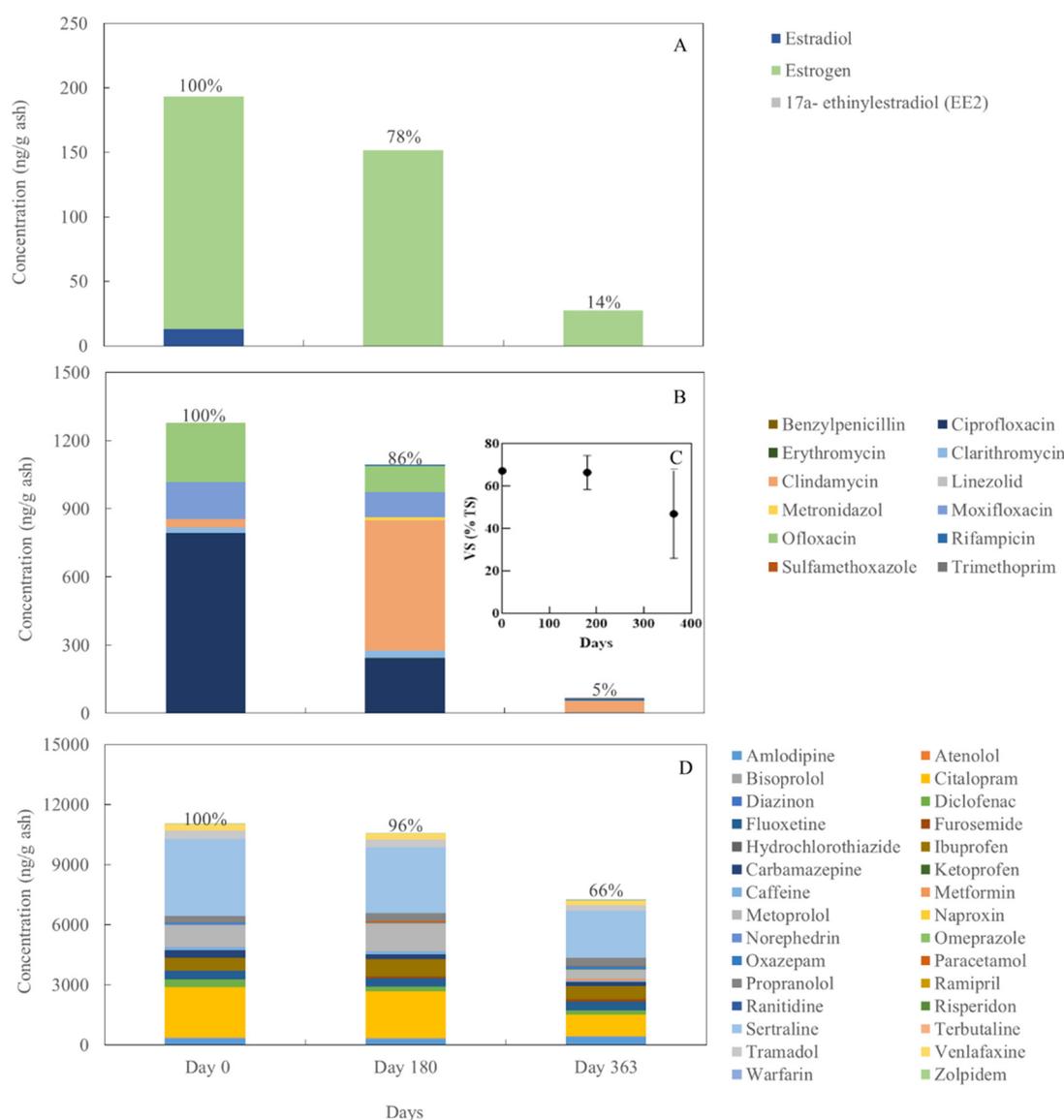
Fig. 2. Concentrations of (A) hormones, (B) antibiotics, (C) volatile solids (VS) and (D) other pharmaceuticals (PhACs) in NO-COM windrow during 363 days of open-air storage under field conditions. Values on bars indicate ∑Antibiotics and ∑PhACs at Day$_i$ relative to Day 0. Graphs show average concentrations ($n = 3$) for each day of measurements.

antibiotics and development of antibiotic resistance in bacteria following use of sewage sludge as fertilizer on arable land (Gros et al., 2020). However, the present study showed that effective removal (95%) of antibiotics was achieved with just open-air storage of sludge and studies of the soil after application shows that the impact on the microbial population is small and short (Rutgersson et al., 2020).

Degradation of the other PhACs studied was generally poor in NO-COM, with only 4% of ∑PhACs degraded during the first 180 days and 34% during 363-day storage period (Fig. 2). A number of substances proved to be resistant to degradation during open-air storage in NO-COM, specifically amlodipine, fluoxetine, oxazepam, propranolol, zolpidem, and ibuprofen. These compounds showed no significant change in their concentrations during the entire period of storage (Table S4). During the first 180 days, only oxazepam and ranitidine showed removal rate > 50% (Fig. 2, Table S4). However, ranitidine was initially detected at low concentration (7.5 ± 0.8 ng/g ash) and the concentration of oxazepam increased during the last 183 days of storage. After 363 days of storage, six substances showed removal rate > 50% (bisoprolol 79%, caffeine 59%, citalopram 56%, metoprolol 63%, ranitidine 93%, and hydrochlorothiazide 94%). The trend in removal of PhACs during the storage period was similar to biodegradation of organic matter in NO-COM (as indicated by VS removal in Fig. 2B). There was no significant loss of VS during the first 180 days of storage, when the weather was shifting towards autumn-winter and the temperature in NO-COM was decreasing (Fig. 1). In fact, the external surface of NO-COM was frozen at some points during the winter even though the measured temperature 60 cm below surface was not below 3.1 deg. C (Fig. 1). The temperature in NO-COM increased towards the second half of the storage period, accompanied by decreases in the

concentrations of substances including VS (Fig. 2B). How large the loss of VS was however uncertain, as indicated by the large standard deviation in Table 2. Generally, the porosity of NO-COM was lower and the density was higher at the end of the storage period compared with the start (Table 1). This indicates that aeration within the windrow was low, but effective degradation of the target hormones, antibiotics, and some PhACs was still achieved (Fig. 2). This suggests that the degradation could have been due to other factors, which lead to anaerobic and/or aerobic degradation of the substances. In addition, day length and exposure to sunlight and solar radiation of the external surface of the windrow decreased in the first half of the storage period (June–December) but increased in the second half. Pharmaceuticals can absorb sunlight (Chong et al., 2010), and highly oxidative products such as reactive oxygen species can develop and react with the pharmaceutical molecules, causing mineralization (Ikehata and El-Din, 2006). It would probably have been better to start the storage experiment early in the spring season, to capture more warm and sunny weather to enhance degradation of the sludge during storage. In a laboratory-scale experiment with no sun light, sludge storage reduced the concentrations of some substances by 5–85%, but increased the concentrations of 21 substances by 4–283% (Cidh, 2014).

The stratification of antibiotics and PhACs with depth in the NO-COM windrow is shown in Fig. 3. At the first measurement (day 0), the windrow was mixed and the concentrations at different depths could not be specified. After 180 days of storage, the top 5–15 cm of NO-COM showed the highest concentrations of ciprofloxacin (1600 ng/g ash; 93% of ∑Antibiotics) (Fig. 3A). Clindamycin showed opposite trend, i.e., the top layer of NO-COM had the lowest concentration of all layers after 180 and 363 days of storage (Fig. 3A). The
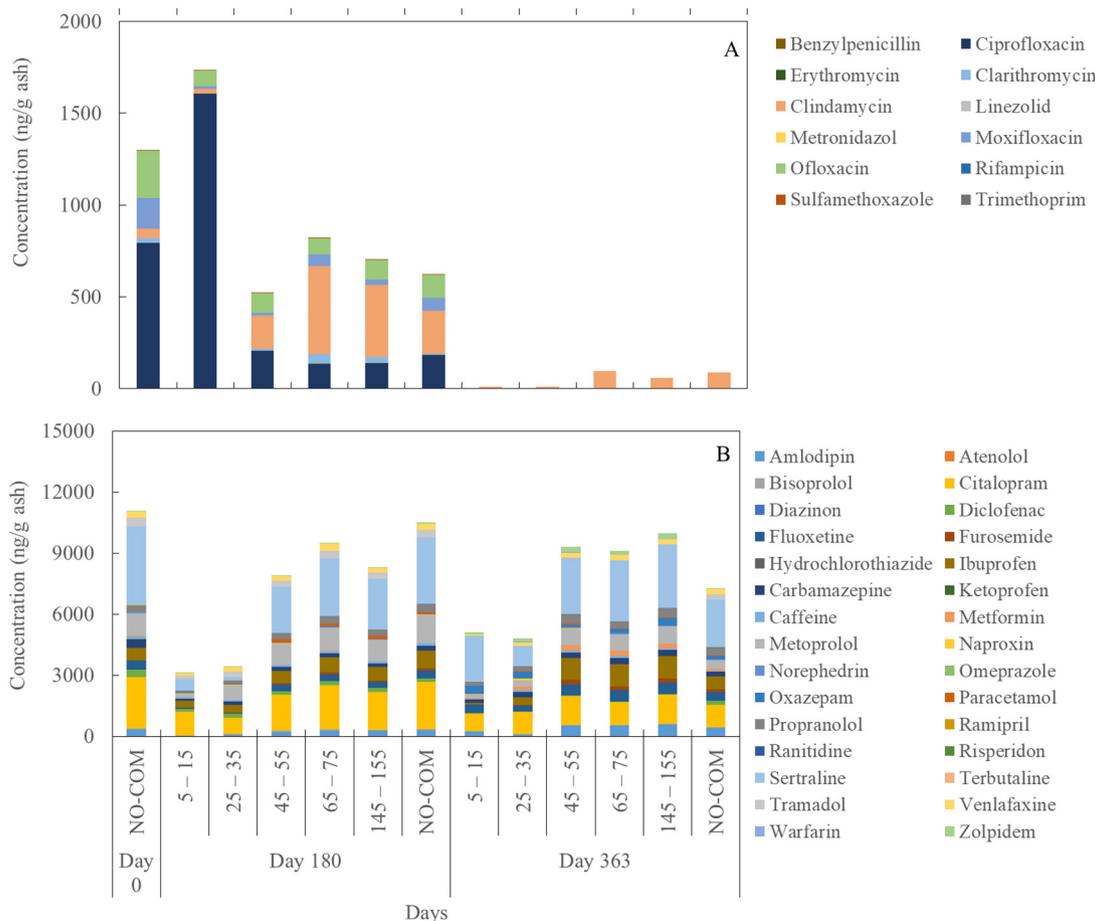
Fig. 3. Concentrations of (A) antibiotics and (B) other pharmaceuticals (PhACs) at different depths in windrow NO-COM during 363 days of open-air storage under field conditions. Concentrations expressed in ng/g ash, graphs show single point measurements (n = 1) for the samples at the different depths. For "NO-COM" columns, n = 3.

occurrence of PhACs was strongly influenced by depth in the windrow (Fig. 3B; Table S5). Upper layers (5–15 cm and 25–35 cm) had the lowest concentrations of $\sum$PhACs (3100–5100 ng/g ash) at the 180- and 363-day measurements, while $\sum$PhACs in deeper layers fluctuated within 7900–9900 ng/g ash (Fig. 3B). Among the target PhACs, sertraline had the highest concentrations at all depths in NO-COM, but the amount of sertraline in the top 5–15 cm layer was only 24–32% of that at 145–155 cm (Fig. 3B).

The low levels of PhACs in the upper layers of NO-COM can be explained by the higher oxidation potential in top layers compared with lower layers. The upper layers of NO-COM showed some nitrification (i.e., production of nitrite and nitrate) (Table S3), confirming aerobic conditions and supporting the hypothesis that better oxygen conditions in sludge windrows during storage result in greater degradation of PhACs in the material. The calculated weighted averages over the different layers of $\sum$Hormones, $\sum$Antibiotics, and $\sum$PhACs in NO-COM on day 363 were 30, 60, and 8000 ng/g ash, respectively. These amounts agree well with the 28, 65, and 7248 ng/g ash measured for

$\sum$Hormones, $\sum$Antibiotics, and $\sum$PhACs in composite samples of NO-COM at day 363 (Table S4). This confirms that the 16% mass reduction in PhACs seen by day 363 mostly occurred at the external surface of the windrow, i.e., in material exposed to air. It would have been interesting to have another NO-COM windrow that combined storage and regular mixing/turning of the sludge during the storage period.

3.4. Degradation of hormones, antibiotics, and PhACs during composting of sludge

The measured concentrations of estrogen and estradiol in COM increased by 3 and 9-fold respectively during the first 26 days of composting, and then declined to non-detectable levels on day 363 (Fig. 4A). The high initial increase in estrogen might be explained by deconjugation of steroid conjugates such as glucuronides and sulfates in the liquid phase of the sludge (water content 49–66%) and to a small degree by oxidation of estradiol to estrogen under aerobic conditions (Atkinson et al., 2012; Hamid and Eskicioglu, 2012). Instrumental
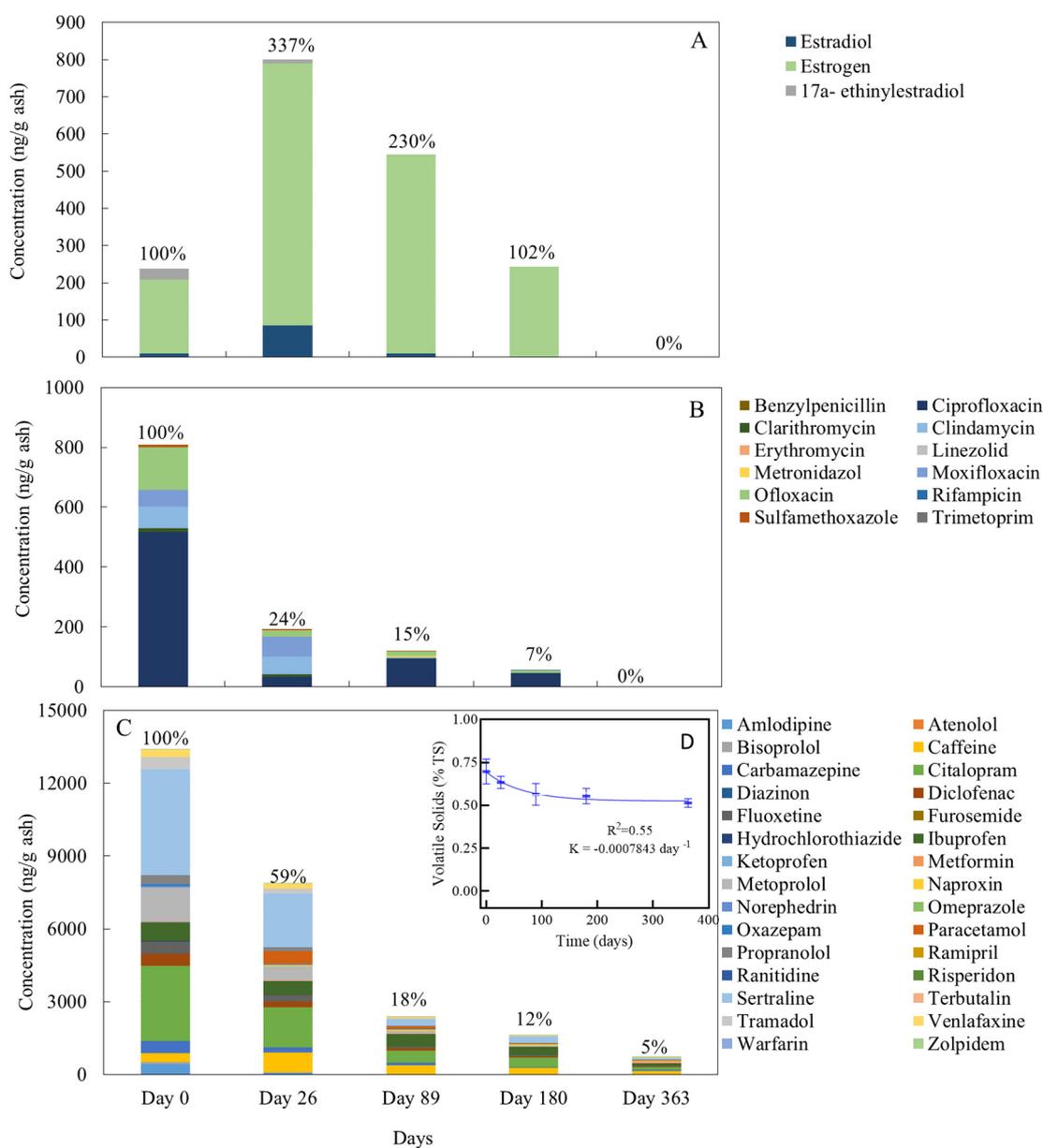
Fig. 4. Concentrations of (A) hormones, (B) antibiotics, (C) other pharmaceuticals (PhACs) in windrow and (D) Volatile Solids in COM during the 363-day sludge composting experiment. Values on bars indicate $\sum$Antibiotics and $\sum$PhACs at Day$_i$ relative to Day 0. Concentrations expressed in ng/g ash, graphs show average concentrations ($n = 3$) for each day of measurements.

challenges i.e. saturation of the electron spray by similar ions/substance could have occurred and caused the detection of initial concentration to be too low. The compound 17a-ethynylestradiol was non-detectable already by day 89. Considering only the data from the periods during which estrogen and estradiol showed degradation (i.e., days 26–363), the rate of degradation, represented by the first-order degradation rate constant K, was 0.0069 and 0.0334 day^{-1} for estrogen and estradiol, respectively (r^2 = 0.56 and 0.76, respectively). The half-life (t$_{1/2}$) of these two hormones was 100 and 21.8 days, respectively. The K value and half-life time of estradiol were comparable to those reported for sterile silt soil (K = 0.024 ± 0.002 day^{-1}, t$_{1/2}$ = 29 days) (Xuan et al., 2008). Biodegradation (aerobic and anaerobic) is a common mechanism for removal of estrogenic hormones (Ilyas and van Hullebusch, 2020). The high oxygen transfer in COM did not seem to have significant effects on removal of the hormones. Estrogen and estradiol were also removed effectively in the NO-COM windrow after 363 days of open-air storage, despite lower oxygen availability than in COM. In fact, Joss et al. (2004) reported that estrogen and estradiol are biodegraded under all (aerobic, anoxic, and anaerobic) conditions, but that biodegradation of 17a-ethynylestradiol is strictly aerobic. In spite of this, 17a-ethynylestradiol was in our study degraded also in NO-COM.

More than 75% of ∑Antibiotics were degraded within 26 days of composting and 93% within 180 days, (Fig. 4B). During the first 26 days, the compost was mixed once a week, which can be expected to have accelerated the composting process due to better oxygen transfer and the microbes finding new material to degrade. No antibiotics were detected in the COM after 363 days of composting. The temperature in the compost was high (>60 °C) during the first 110 days. However, it is not likely that thermal non-biotic degradation of antibiotics had occurred in COM, due to the thermal stability of the substances. High removal of antibiotics was also achieved in NO-COM, which had significantly lower temperature (Fig. 1). Svahn and Björklund (2015) reported high stability of a number of antibiotics (including ciprofloxacin, trimethoprim, and penicillin) at 50–150 °C, while Ottosen and Petersen (2016) reported that an oxidative degradation process, i.e., composting, decreases high concentrations of PhACs and antibiotics in digested and dewatered sludge. Ottosen and Petersen (2016) observed a large reduction in the concentration of tetracycline (86%). Mitchell et al. (2015) reported 95% reductions in florfenicol, sulfamethazine, sulfamethazine, and triclosan within 21 days of thermophilic composting of animal manure, which is in line with findings in the present study. Among the antibiotics, ciprofloxacin, clindamycin, and ofloxacin were still detected in the compost during the first six months of composting, with ciprofloxacin showing the highest concentrations (Table S4, Fig. 4B). The highest degradation constant was found for ciprofloxacin (K = 0.105 day^{-1}; R^2 = 0.77) followed by ofloxacin (K = 0.073 day^{-1}; R^2 = 0.9), which led to t$_{1/2}$ of 6.6 and 9.5 days, respectively (Table 4, Fig. 5). The degradation rate constant for the other antibiotics ranged from 0.01 to 0.027 day^{-1}. The longest half-life was 66.6 days, for moxifloxacin. Similarly, Cheng et al. (2019) observed effective degradation of ciprofloxacin in a pilot-scale swine manure composting study, with K = 0.09–0.1 day^{-1} and t$_{1/2}$ = 7 days. In batch experiments on saline sewage sludge, Li and Zhang (2010) reported first-order degradation of ofloxacin in an activated sludge system, with K = 0.24 day^{-1} and t$_{1/2}$ = 2.8 days.

Also, the other PhACs degraded effectively during the composting, with 41% of ∑PhACs degraded during the initial 26 days of composting, 85% during the first 180 days, and 95% during one year of composting, leaving ∑PhACs of 725 ng/g ash on day 363 (Fig. 4C). Only citalopram and caffeine showed concentrations >100 ng/g ash after one year of composting (Table S6). The other PhACs measured had individual residual concentrations of <LOD - 65 ng/g ash on day 363. Biological degradation and chemical oxidation probably were the main contributers to loss of PhACs in COM. Addition of garden waste to COM considerably increased the porosity in this treatment (Table 2). The regular mixing of COM during the first 89 days of composting aerated the windrow. This

Table 4

First-order exponential decay model parameters for hormones, antibiotics and other pharmaceuticals (PhACs) during 363 days of degradation in open air storage with composting in windrow (COM).

Substance	C$_o$ (ng/g ash)	K^a (Day^{-1})	Half- life (t$_{1/2}$; days)	R^2
Hormones				
Estrogen[c]	878.2	0.0069	100.4	0.5649
Estradiol[c]	41.86	0.0334	20.79	0.7683
17α-ethinyl estradiol[b]	27.81	0.0342	20.28	0.2956
Antibiotics				
Benzylpenicillin[b]	–		–	–
Rifampicin	–		–	–
Sulfamethoxazole	8.285	0.0137	50.56	0.7405
Ciprofloxacin	516.3	0.1049	6.607	0.7698
Clarithromycin	12.8	0.0207	33.56	0.9385
Clindamycin	77.58	0.0190	36.49	0.7587
Erythromycin[b]	1.451	0.0268	25.83	0.2468
Linezolid	0.1578	−0.0085	−81.49	0.9142
Metronidazole[b]	–		–	–
Moxifloxacin	63.38	0.0104	66.57	0.7023
Trimethoprim	–		–	–
Ofloxacin	143.4	0.0733	9.455	0.9089
PhACs				
Amlodipine	442.2	0.0729	9.512	0.8971
Atenolol	–		–	–
Bisoprolol	65.78	0.0282	24.59	0.8091
Caffeine[b]	579.2	0.0037	187.9	0.3117
Carbamazepine	466.5	0.0248	28	0.8417
Citalopram	3055	0.0205	33.76	0.8142
Diazinon	–		–	–
Diclofenac	468.6	0.0224	30.91	0.7911
Fluoxetine	515	0.0261	26.56	0.8027
Furosemide	5.281	0.0081	86.02	0.8471
Hydrochlorothiazide	27.66	0.0716	9.685	0.9165
Ibuprofen	722.5	0.0045	155.7	0.7419
Ketoprofen	33.55	0.0085	82.02	0.7915
Metformin[b]	17.37	0.0028	−247.7	0.2813
Metoprolol	1379	0.0322	21.53	0.7956
Naproxen[b]	–		–	–
Norephedrine	60.32	0.0102	67.86	0.778
Omeprazole	2.946	0.0081	85.41	0.7439
Oxazepam	73.01	0.0805	8.616	0.9207
Paracetamol[b]	304	0.007	99.59	0.1911
Propranolol	348.7	0.0287	24.14	0.8399
Ramipril	–		–	–
Ranitidine	8.674	0.0076	90.66	0.7986
Risperidone	–		–	–
Sertraline	4403	0.0277	24.99	0.89
Terbutaline	–		–	–
Tramadol	495	0.0312	22.21	0.8928
Venlafaxine	299.3	0.0149	46.53	0.8605
Warfarin	–		–	–
Zolpidem	17.97	0.012	57.97	0.8689

[a] A minus sign of rate constant K implies degradation.

[b] The regression models did not show good fit with the measured data.

[c] Degradation of estrogen and estradiol was modeled using data for days 26–363 for estrogen and estradiol, during which these hormones showed decay after an initial increase in concentration.

increased both the biological activity and the possibility for biotic and abiotic degradation of the target substances. The loss of organic matter and the elevated temperature in the COM windrow confirmed the occurrence of an increased biological activity (Fig. 1). It should be noted that the added garden waste may have adsorbed some of the hormones, antibiotics, and PhACs, since it was composed of organic material including wood chips, and adsorption of PhACs and other substances to surface of such material is possible. We did not analyze the PhACs content in composted garden waste separately from the composted sludge, either at start or later. Big wood chips (>1 cm) were removed from the samples for chemical analysis, while smaller chips were included.

Interestingly, diclofenac, which is resistant to degradation in biological wastewater treatment systems, showed good degradation (K = 0.022 day^{-1}, t$_{1/2}$ = 30 days). The fate of diclofenac in the sludge
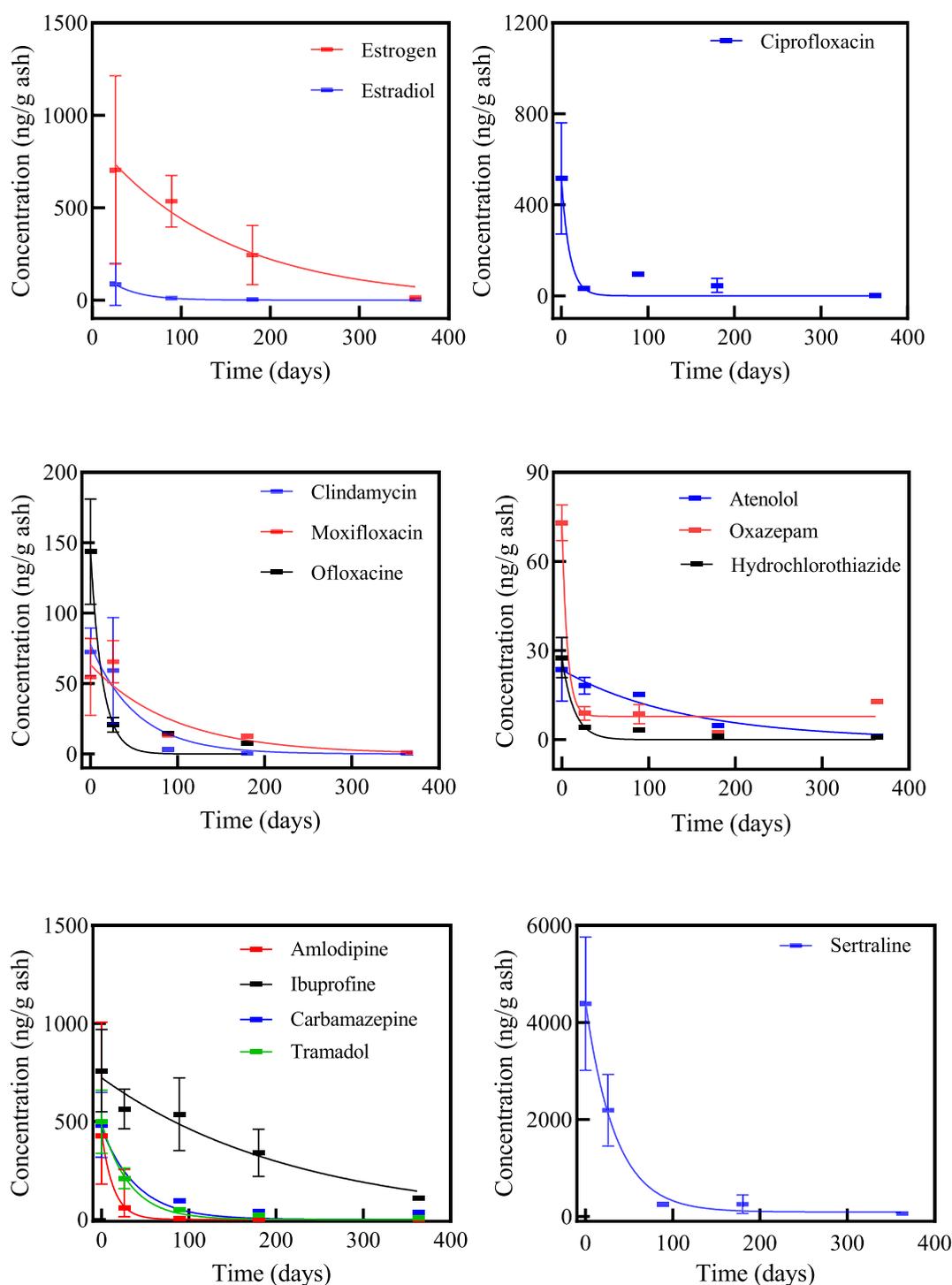
Fig. 5. First-order exponential decay models of selected hormones, antibiotics and other pharmaceuticals (PhACs) in COM windrow. Symbols show mean measured concentrations, bars show standard deviation of the mean, and continuous lines show modeled concentrations.

compost was comparable to that reported for Palouse silty loam agricultural soil with high (5.4%) organic matter content (K = 0.034 day^{-1}, t$_{1/2}$ = 20.4 days) (Xu et al., 2009). Sertraline had the highest initial concentration (4387 ng/g ash), but was degraded by about 98% (K = 0.028 day^{-1}, t$_{1/2}$ = 25 days). Oxazepam had the highest degradation constant (K = 0.0805 day^{-1}), followed by hydrochlorothiazide and amlodipine (K = 0.0716 and 0.0729 day^{-1}, respectively) (Fig. 5 and Table 4). The longest half-life was seen for ibuprofen (t$_{1/2}$ = 156 days, K = 0.0045 day^{-1}). The degradation of ibuprofen was significantly lower than that reported for agricultural

soil (t$_{1/2}$ = 1–6 days, K = 0.11 to 0.77 day^{-1}) (Xuan et al., 2008). Despite the low initial concentration of ranitidine and furosemide, they persisted for a long time in the compost (t$_{1/2}$ = 90.7 and 86.0 days, respectively; K = 0.0076 and 0.0081 day^{-1}).

Removal of different substances was significantly better in COM than in NO-COM, due to higher biological degradation and oxidation in COM (Fig. 5). Previous studies have reported varying removal of a wide range of micropollutants during composting of biomass (animal manure, fecal sludge, sewage sludge), including PhACs, antibiotics, and polyaromatic hydrocarbons (Cheng et al., 2019; Gros et al., 2020; Guo et al., 2020;

Ottosen and Petersen, 2016). In more details, Ottosen and Petersen (2016) reported low removal of anti-inflammatory drugs (ibuprofen 35%, diclofenac 52%) in oxidative degradation (composting) of digested and dewatered sludge. Also, in a full-scale liquid composting plant treating fecal sludge, it was found that the reduction in PhACs ranged from 30 to 80%, including for substances such as atenolol, metoprolol, citalopram, furosemide, and atorvastatin (Gros et al., 2020). Other PhACs (carbamazepine, lamotrigine, venlafaxine, lidocaine, diazepam, and losartan) showed no to low reduction (<50%) during liquid composting, but propranolol, valsartan, and hydrochlorothiazide showed high overall removal rates (>80%) during treatment (Gros et al., 2020).

There are general concerns regarding application of sewage sludge as fertilizer to agricultural land, due to risks related to persistence and eco-toxicological effects of PhACs and spread of antibiotic-resistant bacteria and genes. From the results in the present study, it is obvious that sludge composting is an effective method for treating sewage sludge before land application, to decrease levels of hormones, antibiotics, and PhACs. Even sludge storage without composting was shown to be effective for removal of hormones and antibiotics. The remaining 5% of $\sum$PhACs in sludge compost can be effectively degraded by the soil microflora. Effective removal of PhACs in agricultural soil has been reported (Xu et al., 2009). For sludge from the Nordic countries, very small risks for antibiotic resistance and soil ecosystem has also been reported (Rutgersson et al., 2020) It is believed that composting of sludge can be suitable treatment to achieve degradation of hormones, antibiotics and other pharmaceutical in a wide range of climate and weather conditions.

4. Summary and concluding remarks

Fate of hormones, antibiotics, and PhACs in sludge during storage with and without composting was studied. The sludge had very low concentrations of hormones, antibiotics, and PhCAs after one year of composting. The sludge stored in the open air showed lower, but still high, removal of hormones and antibiotics, while the removal of PhACs was low. Top layers of the sludge windrow displayed higher removal than deeper layers. Both aeration and biological activity seemed to be important in removal of pharmaceuticals. Composting during six months is thus a method to achieve high removal of pharmaceuticals in sludge.

Significant amounts of hormones, antibiotics, and PhACs were still present in non-composted sludge after six months of the storage, but storage for one year was effective in removal of hormones and antibiotics. Storage of sludge for six months is not sufficient for removal of pharmaceutical residues before land application.

CRediT authorship contribution statement

The following authors has contributed to the article as described below.

Sahar Dalahmeh: participated in planning the experiment, collected samples, conducted part of the analyses, analyzed the data and wrote the manuscript.

Gunnar Thorsen: conducted the analyses of antibiotics, hormones and other pharmaceuticals, and reviewed the article.

Håkan Jönsson: planned the experiment and reviewed the article.

Declaration of competing interest

The authors have no competing interests to declare, or financial or personal relationships with other people or organizations that could have inappropriately influenced this work.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.151271.

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