

Effect of pH on phosphorus, copper, and zinc elution from swine wastewater activated sludge

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ABSTRACT

With the goal of reducing the amounts of phosphorus (P), copper (Cu), and zinc (Zn) discharged from swine wastewater activated sludge treatment facilities, we studied the elution of these elements from activated sludge at various pH values. Sludge samples with neutral pH collected from three farms were incubated at pH values ranging from 3 to 10. The soluble concentrations of these elements changed dramatically with pH and were highest at pH 3. We assumed that P present in the sludge under neutral and alkaline conditions was in insoluble form bound up with magnesium (Mg) and calcium (Ca), because Ca and Mg also eluted from the sludge at low pH. To clarify forms of Zn and Cu in the sludge, we performed a sequential extraction analysis. Zinc in adsorbed, organically bound, and sulfide fractions made up a large proportion of the total Zn. Copper in organically bound, carbonate, and sulfide fractions made up a large proportion of the total Cu. The soluble P concentrations were lowest at pH 9 or 10 (11–36 mg/L), the soluble Zn concentrations were lowest at pH 8 or 9 (0.07–0.15 mg/L), and the soluble Cu concentrations were lowest at pH 6–9 (0.2 mg/L, the detection limit).

Key words | activated sludge treatment, copper, phosphorus, swine wastewater, zinc

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INTRODUCTION

Phosphorus (P), copper (Cu), and zinc (Zn) are added to swine feeds in high concentrations; the latter two elements are particularly common additives and are used to improve feed efficiency and swine health (Nicholson *et al.* 1999; Takada 2005; JLIA 2013). However, swine can absorb only 10–20% of the Cu and Zn they ingest and only 15–40% of the P in commonly used feed ingredients; the rest is excreted (Takada 2005). Consequently, swine excrement contains high concentrations of these three elements, which can thus be found as environmental contaminants in many regions of the world (Steinfeld *et al.* 2006). Swine slurry, which is a mixture of urine and feces, also contains relatively high concentrations of these three elements, and their distributions in the slurry and its treatment by-products have been well studied, along with the effects of application on arable land (Beline *et al.* 2004; Bolan *et al.* 2004; Steinmetz *et al.* 2009).

As might be expected, swine wastewater, which contains service water in addition to urine and feces, also contains high concentrations of P, Zn, and Cu. For example, raw swine wastewater has been reported to contain, on average,

431, 499, and 330 mg/L of total P (Kunz *et al.* 2009; Suzuki *et al.* 2010), as well as 3.7 and 12 mg/L of total Zn and 1.8 and 4.7 mg/L of total Cu (Suzuki *et al.* 2008, 2010). In Japan, swine wastewater is generally treated by means of an activated sludge process. Most of the P, Zn, and Cu in the raw swine wastewater exists in solid form (72–91%), and large portions of the solid-associated P, Zn, and Cu are removed when suspended solids (SS) are removed during the pretreatment process and the subsequent activated sludge process. Therefore, most of the P, Zn, and Cu in the effluent from the activated sludge process (71–93%) is in soluble form (Suzuki *et al.* 2008, 2010). Any metals that are removed from the aqueous phase in an aeration tank are removed by transfer to the activated sludge; volatilization of metals to the atmosphere is insignificant. The removal of metals during the activated sludge process occurs via a complex process that depends on many inter-related physical, chemical, biological, and operational parameters (Ziolko *et al.* 2011).

A survey of 11 swine farms in Japan revealed that in the effluents from the activated sludge processes, the average

total P, total Zn, and total Cu concentrations were 35, 0.3, and 0.2 mg/L, respectively. However, relatively high concentrations of soluble Zn and Cu were observed when the pH of the effluent was either unusually low or unusually high; for example, 1.9 mg/L Zn and 1.5 mg/L Cu at pH 4.7 and 2.3 mg/L Cu at pH 8.6 (Suzuki et al. 2010). The acidification of sewage sludge is known to promote elution of metals from the sludge (Reimers 1983), and pH strongly affects the solubility of Cu and Zn bound to organics, hydroxides, and sulfides (Snoeyink & Jenkins 1980; Tchobanoglous et al. 2003). The removal of P from activated sludge is also affected by pH under certain circumstances. Crystallization of P as magnesium ammonium phosphate (MAP, $\text{NH}_4\text{MgPO}_4 \cdot 6\text{H}_2\text{O}$) and precipitation with calcium (Ca) in the form of hydroxyapatite [HAP, $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$] are ways to remove P during wastewater treatment. The suitable pH conditions for these processes are pH 8.5–9.5 (Le Corre et al. 2009) and up to 12 (Stumm & Morgan 1981), respectively. Crystallization of MAP can occur unexpectedly during treatment of anaerobic wastewater, and the resulting crystals can cause mechanical difficulties at treatment facilities (Snoeyink & Jenkins 1980; Le Corre et al. 2009). Removal of P in the form of Ca phosphate material at high pH upon addition of calcium hydroxide ($\text{Ca}(\text{OH})_2$) to swine wastewater has been reported (Szogi & Vanotti 2009). In addition, substantial amounts of metal-P forms (particularly with Ca and magnesium (Mg)) have been shown to be naturally present in the activated sludge from swine wastewater (Bortone et al. 1992). All these results indicate that the elution of P, Zn, and Cu from activated sludge can be expected to vary with the pH in the aeration tank. However, little information about elution of these elements over a wide pH range is available.

Recently, the water pollution control law in Japan lowered the uniform effluent standard value of Zn to 2 mg/L for large-scale facilities, and the standard values of Cu and P are 3 and 8 mg/L, respectively. With the goal of preventing discharge of these elements into the public water supply, we investigated the relationships between pH and P, Zn, and Cu concentrations in swine wastewater activated sludge, and we evaluated the forms of Zn and Cu in the sludge.

MATERIALS AND METHODS

Samples

An activated sludge sample (mixed liquor sample) was collected from the aeration tank of the activated sludge

Table 1 | Characteristics of sludge samples collected from swine farms

	Farm A	Farm B	Farm C
pH	6.8	8.1	8.4
MLSS (mg/L)	7650	5350	8200
MLVSS (mg/L)	5950	4650	6350
N-NH ₄ ⁺ (mg/L)	7.1	460	3.4
N-NO ₂ ⁻ (mg/L)	0	0	0
N-NO ₃ ⁻ (mg/L)	81	2.3	46

MLSS, mixed liquor suspended solids; MLVSS, mixed liquor volatile suspended solids.

treatment facility of each of three farms (A, B, and C): farm A treats a mixture of swine wastewater and small dairy wastewater, and farms B and C treat swine wastewater. The samples were mixtures of SS and wastewater, and each sample was tested at the SS concentration that existed in the corresponding aeration tank. The characteristics of the sludge samples are shown in Table 1.

Incubation of activated sludge samples under various pH conditions

Activated sludge samples with pH values ranging from 3 to 10 were prepared by the addition of hydrochloric acid (HCl) or sodium hydroxide (NaOH) to the samples from the three farms. Three 50-mL aliquots at each pH value were incubated in flasks for 24 h with stirring at room temperature (23–29 °C). After incubation, the samples were centrifuged at 3,000 rpm (1,000 g) for 10 min, and the soluble P, Mg, Ca, Zn, and Cu concentrations in the supernatants were measured.

Sequential extraction of Zn and Cu

Zn and Cu in the mixed liquor samples obtained from the aeration tanks at the farms were sequentially extracted according to the method described by Stover et al. (1976). In brief, the samples were centrifuged at 10,000 g for 10 min, and the concentrations of soluble Zn and Cu in the supernatants were measured. Then, a 1-g sample of each of the separated solids was sequentially extracted with 1 M potassium nitrate (KNO_3), 0.5 M potassium fluoride (KF) (pH 6.5), 0.1 M sodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7$), 0.1 M ethylenediaminetetraacetic acid (pH 6.5), and 1 M nitric acid (HNO_3) to obtain the exchangeable, adsorbed, organically bound, carbonate, and sulfide fractions, respectively, as well as the residual fraction remaining after sequential extraction. Sequential extraction was performed in triplicate for each activated sludge sample.

Analytical methods

To determine the concentrations of mixed liquor SS and mixed liquor volatile suspended solids (VSS), we centrifuged the samples at 800 g, decanted the supernatants, re-suspended the solids in deionized water, and dried them at 105 °C and then at 600 °C.

Solution pH was measured with a glass electrode (D-13; Horiba, Japan).

The concentrations of nitrate and nitrite ions (NO_3^- and NO_2^-) in the mixed liquor samples were determined with an HIC-VP Super ion-exchange chromatograph (Shimadzu Co., Japan), and NH_4^+ concentrations were determined with a DX-120 ion-exchange chromatograph (Dionex Co., Japan).

Soluble P, Mg, Ca, Zn, and Cu concentrations after incubation of the samples under various pH conditions were measured by inductively coupled plasma optical emission spectrometry (730-ES, Varian Medical Systems, USA). In some of the samples, the Cu concentration was below the detection limit (0.2 mg/L); for these samples, the concentration was set at 0.2 mg/L.

The Zn and Cu concentrations in the fractions obtained by sequential extraction were measured by inductively coupled

plasma mass spectrometry (7500c, 7500ce, Agilent Technologies, USA) after nitric acid and perchloric acid digestion.

RESULTS AND DISCUSSION

Phosphorus elution from sludge

The total P, total Mg, and total Ca concentrations in the sludge samples used in this study ranged from 37 to 51 mg/g-SS (48–65 mg/g-VSS, 260–390 mg/L), from 13 to 21 mg/g-SS (17–24 mg/g-VSS, 100–120 mg/L), and from 46 to 62 mg/g-SS (53–81 mg/g-VSS, 250–510 mg/L), respectively (Table 2). The total P concentrations were similar to those reported for sewage activated sludge (16–64 mg/g-VSS according to Mino *et al.* (1983)).

The pH in activated sludge aeration tanks at swine farms is usually approximately neutral, and in our study, the pH range of the mixed liquor samples obtained from the aeration tanks was 6.8–8.4 (Table 1). However, it has been reported that the pH in the mixed liquor at swine farms can occasionally be as low as 3.7 or as high as 9.0 (Waki *et al.* 2010). Therefore, the pH range used for our incubation experiments was 3–10. We found that the soluble P, Mg, and Ca concentrations after incubation of the sludge varied dramatically with pH (Figure 1). For all three elements, the soluble concentrations were highest at pH 3, accounting for 46–69% of total P, 75–87% of total Mg, and 76–95% of total Ca in the original sludge samples. The lowest concentrations were observed at pH 9 or 10: 11–36 mg/L for P, 4.6–23 mg/L for Mg, and 12–28 mg/L for Ca.

It has been reported that the P in the activated sludge from swine wastewater exists mostly in cell-P,

Table 2 | Total P, Mg, Ca, Zn, and Cu concentrations in sludge samples collected from swine farms

	Farm A	Farm B	Farm C
Total P (mg/g-SS)	51	48	37
Total Mg (mg/g-SS)	13	21	15
Total Ca (mg/g-SS)	59	46	62
Total Zn (mg/g-SS)	2.6	1.3	1.2
Total Cu (mg/g-SS)	0.77	0.79	0.52

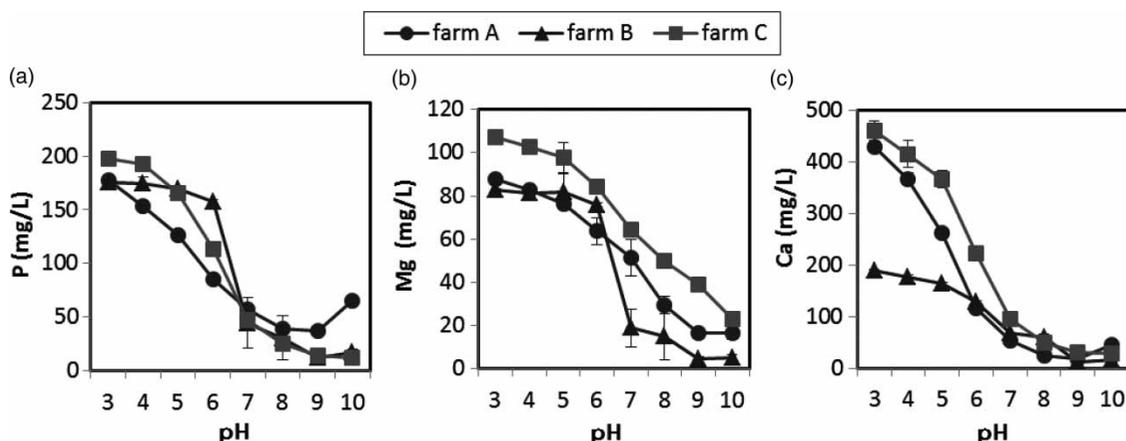


Figure 1 | pH dependence of soluble (a) P, (b) Mg, and (c) Ca concentrations. Error bars indicate standard deviations ($n = 3$).

polyphosphate-P, and metal-P forms (particularly with Ca and Mg) (Bortone *et al.* 1992). Because swine wastewater contains high concentrations of P, Mg, Ca, and ammonium (NH_4^+), the potential for crystallization or precipitation of P in the form of MAP, HAP, or amorphous calcium precipitates is high (Suzuki *et al.* 2001). In fact, the crystallization of MAP (Suzuki *et al.* 2002) and the formation of amorphous precipitates with Ca (Fernandes *et al.* 2012) in swine wastewater have been reported. The pH is one of the main factors controlling the crystallization process. MAP crystallization is reported to be maximal at pH 8.5–9.5 (Le Corre *et al.* 2009), and HAP crystallization increases with increasing pH up to 12 (Stumm & Morgan 1981). The pH at which we observed the lowest soluble P concentrations (pH 9 or 10) is conducive to MAP or HAP crystallization, although the formation of other precipitates containing P and Ca is possible. The Mg:P molar ratio of MAP is 1, and the Ca:P molar ratio of typical HAP is 1.7. In the present study, the P:Mg:Ca molar ratios that resulted from lowering the pH from 9 (at which the concentrations of soluble P were the lowest) to 3 were 1:0.65:2.25, 1:0.61:0.84, and 1:0.48:1.81 for farms A, B, and C, respectively. We assumed that crystallization or precipitation of P with Mg and Ca contributed substantially to the insolubility of P in the swine wastewater activated sludge aeration tanks.

For sludge from farms B and C, the soluble P concentration at pH 8 differed from that at pH 9; when the pH was increased from 8 to 9, the soluble P concentration decreased from 30 to 11 mg/L for farm B and from 25 to 13 mg/L for farm C. This result indicates that increasing the effluent pH to 9 by adding alkali may be useful for reducing the soluble P concentration without the need for flocculants. However, because the Japanese uniform standard for P concentration in effluent from large-scale facilities is quite low (e.g., average 8 mg/L) and because the standard for effluent pH is 5.8–8.6 in some areas, an additional P removal process may be required if swine wastewater effluent is to meet these rigorous standards. Elevating the pH by adding $(\text{Ca}(\text{OH})_2)$ and removal of P in the form of a Ca phosphate (Szogi & Vanotti 2009), as well as the use of a P removal material (Okano *et al.* 2013; Yamashita *et al.* 2013), may be suitable options.

Zinc and copper elution from sludge

The total Zn and Cu concentrations in the sludge samples analyzed in this study were 1.2–2.6 mg/g-SS (1.5–3.4 mg/g-VSS, 7.0–20 mg/L) and 0.52–0.79 mg/g-SS (0.68–0.99 mg/g-VSS, 4.2–5.9 mg/L), respectively (Table 2). These

concentrations were slightly higher than those reported for sewage activated sludge (0.1–0.75 mg/g-SS and 0.05–0.6 mg/g-SS, respectively; Kawashima *et al.* 2003). The soluble Zn and Cu concentrations varied dramatically with pH (Figure 2). For all three farms, the concentrations were maximal at pH 3, accounting for 67–86% of total Zn and 12–29% of total Cu in the original samples. The lowest soluble Zn concentration (0.07–0.15 mg/L) was observed at pH 8 for farms A and B and at pH 9 for farm C. This concentration range was quite low and was lower than the standard value for effluent from large-scale facilities in Japan (2 mg/L). In fact, at pH 5–10, the soluble Zn concentration was lower than the standard value.

The soluble Cu concentrations were the lowest (0.2 mg/L, the detection limit) at pH 6–9; however, under all pH conditions, the concentrations were below the standard value for effluent from large-scale facilities (3 mg/L). The total Cu concentrations in the sludge samples from the aeration tanks at the three farms were high (4.2–5.9 mg/L); however, when enough of the SS that included solid-form Cu was removed, the risk of exceeding the standard value was low because Cu elution was minimal even at pH 3.

The Zn and Cu in the sludge existed in various forms (Figure 3). Zinc in the adsorbed, organically bound, and sulfide fractions made up a considerable portion of the total Zn. Copper in the organically bound, carbonate, and sulfide fractions accounted for a considerable portion of the total Cu. In sewage activated sludge, a large portion of the Zn was in the organically bound and carbonate fractions, and a large portion of the Cu was in the sulfide fraction in excess sludge (Kawashima *et al.* 2003). The forms of Zn and Cu in swine sludge did not differ significantly from

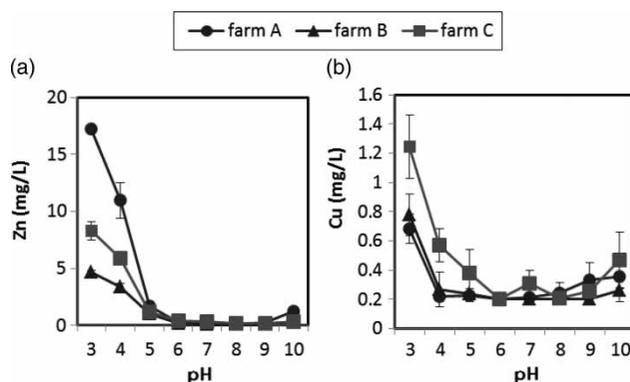


Figure 2 | pH dependence of soluble (a) Zn and (b) Cu concentrations. Error bars indicate standard deviations ($n = 3$).

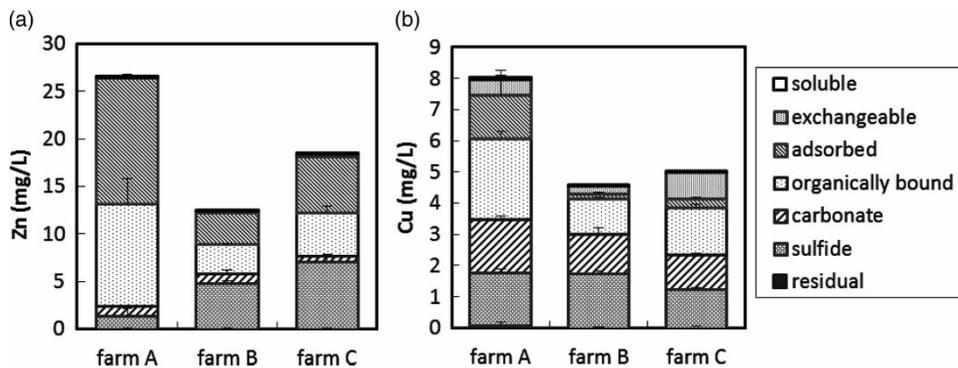


Figure 3 | Forms of (a) Zn and (b) Cu in activated sludge, as indicated by sequential extraction. Error bars indicate standard deviations ($n = 3$).

those of sewage sludge, although the total metal concentration was rather high in swine wastewater.

Metal removal during activated sludge treatment is a complex process (Ziolko *et al.* 2011). Acidification to pH 2.0 reportedly promotes the solubilization of Zn and Cu in sewage sludge (Reimers 1983). Metal sulfides and metal hydroxides dissolve under acidic and alkaline conditions (Snoeyink & Jenkins 1980; Tchobanoglous *et al.* 2003). Humic substances, which are soluble at high and low pH values, combine with metal ions (Snoeyink & Jenkins 1980). Therefore, at very high or very low pH values, Zn and Cu could be eluted from these compounds. Our results indicate that pH is an important factor for lowering Zn and Cu concentrations in effluent from activated sludge treatment facilities on farms.

CONCLUSIONS

We studied the elution of P, Cu, and Zn from swine wastewater activated sludge at pH values ranging from 3 to 10. Elution of these elements was highest at pH 3. We assumed that P eluted from the sludge as P crystallized or precipitated with Mg and Ca. Zinc in the adsorbed, organically bound, and sulfide fractions accounted for a substantial portion of the total Zn, and Cu in the organically bound, carbonate, and sulfide fractions accounted for a substantial portion of the total Cu. We assumed that some of the Cu and Zn in these fractions eluted from the sludge at high and low pH values.

The soluble P concentrations were lowest at pH 9 or 10 (11–36 mg/L), the soluble Zn concentrations were lowest at pH 8 or 9 (0.07–0.15 mg/L), and the soluble Cu concentrations were lowest at pH 6–9 (0.2 mg/L, the detection limit). The soluble Zn and Cu concentrations were quite low, lower than the corresponding standard values for

effluent from large-scale facilities in Japan (2 mg/L for Zn and 3 mg/L for Cu). However, the P concentration was higher than the standard value for P (e.g., an average concentration of 8 mg/L is the uniform standard value for large-scale facilities in Japan).

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