Long-term investigation of phosphorus removal by iron electrocoagulation in small-scale wastewater treatment plants
I. Mishima, M. Hama, Y. Tabata and J. Nakajima

ABSTRACT
Small-scale wastewater treatment plants (SWTPs), called Johkasou, are widely used as decentralized and individual wastewater treatment systems in sparsely populated areas in Japan. Even in SWTPs, nutrients should be removed to control eutrophication. An iron electrolysis method is effective to remove phosphorus chemically in SWTPs. However, it is necessary to determine the precise conditions under which phosphorus can be effectively and stably removed in full scale SWTPs for a long period. Therefore, long-term phosphorus removal from SWTPs was investigated and optimum operational conditions for phosphorus removal by iron electrolysis were analyzed in this study. Efficient phosphorus removal can be achieved for a long time by adjusting the amount of iron against the actual population equivalent. The change of the recirculation ratio had no negative effect on overall phosphorus removal. Phosphorus release to the bulk phase was prevented by the accumulated iron, which was supplied by iron electrolysis, resulting in stable phosphorus removal. The effect of environmental load reduction due to phosphorus removal by iron electrolysis was greater than the cost of power consumption for iron electrolysis.

Key words | Fe/P molar ratio, iron electrolysis, phosphorus removal, small-scale wastewater treatment plants

INTRODUCTION
Small-scale wastewater treatment plants (SWTPs), called Johkasou, are widely used as decentralized and individual wastewater treatment systems in sparsely populated areas in Japan (JECES 2017). They can treat domestic wastewater from fewer than 10 households directly. Even in SWTPs, nutrients should be removed in order to control eutrophication in Japan. SWTPs are often installed in agricultural regions with a small population, and the phosphorus removed and accumulated in SWTPs may be used as agricultural fertilizer. Studies on phosphorus recovery from sludge incineration have also been progressing in recent years. As a process prior to the application of these techniques, a method for chemically and stably removing phosphorus in small-scale facilities such as SWTPs would be effective.

An iron electrolysis method is an electrocoagulation technique (Mollah et al. 2001; Pulkka et al. 2004) that uses electrochemical technology (Cong et al. 2016), which has been recently developed as a new method for the addition of iron as a coagulant into the wastewater treatment process (Irdemez et al. 2006; Mishima & Nakajima 2011; Tran et al. 2012). Ferrous ions are supposed to be released from the anode of the iron electrodes, and then oxidized by dissolved oxygen to become ferric ions. These ferric ions in turn combine with phosphate ions to remove phosphate from the wastewater. Phosphorus can easily be removed by means of the formation of compounds such as FePO₄, as conducted in this study.

With the iron electrolysis method, because an iron plate can be kept in an aerobic tank, it is not necessary to install the liquid coagulant tank used at large-scale treatment plants. Furthermore, the hydrogen gas and the hydroxide ion are consequently produced in a cathode (Omwene & Kobya 2018), therefore the pH decrease involved when adding liquid coagulant does not occur. In addition, microorganism respiratory activities were not reduced using iron electrolysis in a full scale wastewater treatment plant.

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It is also known that iron oxides formed in the anode may prevent the iron electrode from dissolving from the electrode, resulting in deteriorated phosphorus removal performance. This problem was addressed by interchanging the anode and cathode in several periods, and this technology enabled a long-term phosphorus removal operation (Morizumi et al. 1999).

However, problems still remain with regard to the stability of phosphorus removal. Therefore, the behavior of phosphorus and Fe in SWTPs must be investigated in detail in order to clarify the factors contributing to phosphorus removal. Furthermore, it is necessary to determine the precise conditions under which phosphorus can be effectively and stably removed in full scale SWTPs for a long period. Therefore, long-term phosphorus removal from SWTPs was investigated and optimum operational conditions for phosphorus removal by iron electrolysis were analyzed using the data collected. Then, the effects of both the internal phosphorus behavior in the tank and the recirculation ratio on the overall phosphorus removal were also investigated in plants. Finally, the economy of this phosphorus removal system was calculated using an LCA tool to evaluate the cost effectiveness of this system.

**MATERIALS AND METHODS**

**Investigated wastewater treatment plants**

This study targeted five full scale SWTPs in Saitama prefecture, Japan (S1 to S5, each treating wastewater from five people equivalents), in which the iron electrolysis equipment for phosphorus removal was installed at the top of the aerobic tank. Each plant had two anoxic tanks and one aerobic tank, as shown in Figure 1. The volumes of anoxic tank 1, anoxic tank 2, and the aerobic tank were 0.93, 0.95, and 0.73 m³, respectively. Fluidized media were placed into the aerobic tank for biological oxidation of organic matter and nitrogen. Filtration using the fluidized media was performed at the bottom of the aerobic tank, and the wastewater in the aerobic tank was returned to anoxic tank 1 for denitrification. Four iron electrodes (two units), which were about 300 mm in height, were installed in the aerobic tank (Mishima & Nakajima 2011). The weight of each iron electrode was approximately 1.2 kg, and a direct current was allowed to flow in order to release iron ions for phosphorus removal. Iron electrodes were replaced by new ones every 4 months.

**Operational conditions of wastewater treatment plants**

The operational conditions of the five SWTPs are shown in Table 1. Each SWTP can serve five people equivalents; however, the actual population using each was two to five people. The Fe addition and recirculation conditions were adjusted during the experiment in order to assess their effect on phosphorus removal performance. Iron was not added during the first period for all plants nor for all periods of S2 as the control experiment. The amount of Fe addition was calculated according to Faraday’s law, based on the electrical current. The electrical current corresponding to the Fe addition of 8, 14, and 19 g Fe/d was 0.4, 0.6, and 0.8 A, respectively. The Fe/P molar ratio (molar ratio of planned P load to amount of Fe addition) and recirculation ratio (ratio of returned flow rate as recirculation to influent flow rate) were also calculated, and are shown in brackets in the row of Fe addition and recirculation in Table 1. Hyphens and n mean no Fe addition or no recirculation and the number of sampling in Table 1, respectively. Backwashing was carried out twice a day by means of intensive aeration to move the sludge produced in the aerobic tank to anoxic tank 1. The biologically and chemically produced sludge was stored in anoxic tank 1.

These SWTPs had been operated for about 10 years. Removal of the accumulated sludge in each SWTP was conducted before beginning the sampling. Effluent grab sampling was carried out every 2 weeks for an entire year from January 2014 to January 2015. Biochemical oxygen
Table 1 | Operational conditions of investigated SWTPs

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<td>S2</td>
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<td>Fe addition gFe/d</td>
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RESULTS AND DISCUSSION

Outline of treatment performance

Figure 2 shows average values for the amount of flow rate, SS, BOD, DOC, T-N, NO₃-N, S-Ca, and S-Fe in the effluent during each period, along with the standard deviations. According to the calculation using the amount of flow rate in Figure 2 and population in Table 1, the average daily amounts of water use per person for S1 to S5 were 200, 310, 220, 170, and 170 L/d/person, respectively. Although it was low in S2 due to low population, these values were almost equivalent to 200 L/d/person, which is used as the designed wastewater flow for SWTPs in Japan (JECES 2017). This indicates that the actual wastewater flow can be estimated if the population has been obtained. Therefore, the influent phosphorus load was calculated using the product of the power consumption due to iron electrolysis and the electric energy charge for each unit of power consumption in the case of period 3 in S4. The environmental load reduction by T-P removal was estimated by the product of the decrease in T-P discharge load and T-P damage factor (973.9 Japanese yen/T-Pkg), and the economic value conversion factor (1 Japanese yen/Japanese yen) according to LIME2. The decrease in T-P discharge load was calculated by the product of the difference in T-P concentration between the assumed phosphorus concentration in the influent of 5.0 mgP/L (JECES 2017) and the average effluent T-P concentration in the case of period 3 in S4, and the planned flow rate of 1.0 m³/d.

Evaluation of phosphorus removal system

LIME2 (Itsubo & Inaba 2010; Itsubo et al. 2012), which is an LCA tool that can estimate the integrated environmental load as an economic value (Japanese yen), was introduced to evaluate the phosphorus removal system. Since this phosphorus removal system consumes electric power, the cost of electric power consumption was compared to that of the environmental load reduction by T-P removal.

The cost of electric power consumption for iron electrolysis (14 gFe/d of Fe addition) was estimated according to the product of the power consumption due to iron electrolysis and the electric energy charge for each unit of power consumption in the case of period 3 in S4. The environmental load reduction by T-P removal was estimated by the product of the decrease in T-P discharge load and T-P damage factor (973.9 Japanese yen/T-Pkg), and the economic value conversion factor (1 Japanese yen/Japanese yen) according to LIME2. The decrease in T-P discharge load was calculated by the product of the difference in T-P concentration between the assumed phosphorus concentration in the influent of 5.0 mgP/L (JECES 2017) and the average effluent T-P concentration in the case of period 3 in S4, and the planned flow rate of 1.0 m³/d.
wastewater flow obtained from the population and a general phosphorus concentration of 5 mgP/L (JECES 2017). The molar ratios of the Fe addition to phosphorus loads shown in Table 1 were also calculated from these values and used as an index for the phosphorus removal analysis described later.

The values of SS and BOD were generally 10 mg/L or less in every SWTP, with the exception of period 5 for S1. During this period, it was thought that the sludge in the tank reached an excessive concentration and not all of it could be trapped by filtration using the fluidized media. As a result, SS flowed out in the effluent, leading to an increase in SS and BOD. In fact, following this investigation period, the accumulated sludge was removed from S1 and the additional operation continued for approximately 2 months. The average values of SS and BOD decreased to 4.8 and 6.0, respectively, in this period.

The fact that T-Ca and S-Ca had almost the same concentration indicated that the majority of calcium existed as a soluble form. The values of S-Ca were nearly constant during all periods, between 20 and 25 mg/L. In the periods where iron electrolysis was not performed, T-Fe in the effluent was 0.2 to 0.3 mg/L. The amount of T-Fe in the effluent exhibited an increase through the iron electrolysis. However, the added Fe was thought to be in a particulate state, because, the values of S-Fe with iron electrolysis were in the same range without iron electrolysis.
During period 4, with the recirculation set to 0, a decrease in alkalinity, an accumulation of NO₃-N and an associated increase in T-N were clearly observed. It was implied that biological denitrification and the associated alkalinity supply ceased because the nitrified wastewater was not circulated to the anoxic tank. As a result, the nitrogen removal performance, an important function of the SWTP, was lost. However, neither nitrification inhibition nor a significant pH decrease occurred, because an alkalinity of 40 mg/L or greater remained. Actually, the pH values were within a neutral range in every case. As described above, when the recirculation was changed, the effect thereof on nitrogen removal was confirmed. In other periods, the T-N values were close to 10 mg/L or less, which indicated that nitrification and denitrification progressed effectively in each period. Changes in T-N corresponding to changes in electrolysis conditions have not been specifically observed; therefore, it was concluded that the iron electrolysis did not obstruct nitrogen removal.

**Phosphorus removal performance**

Regarding the T-P values during period 1 and all the periods of S2, the value of S1 was lower than those in other periods, at approximately 2 mgP/L, while that of other periods was approximately 4 mgP/L on average. The difference in phosphorus concentration was observed among SWTPs.

The values of both T-P of S2 did not show remarkable variations during any periods because S2 was not subjected to electrolysis. The mean effluent PO₄-P concentration and the standard deviation for each measurement period are shown in Figure 3. A decrease in PO₄-P was observed, corresponding to the increase in Fe addition by means of electrolysis during periods 2 to 5, indicating effective phosphorus removal by iron electrolysis. For S1 and S4, it was confirmed that the PO₄-P values during period 3 were 1 mg/L or less. However, for S3 and S5, the PO₄-P values in the effluent during the operation in period 3 were 1 to 2 mg/L. Accordingly, when S3 and S5 were operated with a further increased Fe addition amount during period 4 to 5, it was confirmed that the PO₄-P values in the effluent decreased further, to 1 mg/L or less. Therefore, the conditions affecting phosphorus removal by less than 1 mg/L differed among the SWTPs.

**Relationship between phosphorus and Fe/P**

The relationships between the effluent PO₄-P and Fe/P molar ratio are shown in Figure 4. The PO₄-P concentration correlated with the Fe/P molar ratio (P < 0.001), indicating that efficient phosphorus removal could be achieved by adjusting the addition of Fe with the iron electrolysis against the actual population equivalent in each SWTP. This result corresponded with the previous results, in which the addition of iron as a coagulant by Fe/P molar ratio of 2.0 was enough to reduce the phosphorus concentration below 1.0 mg P/L (Rittmann & McCarty 2001).

For S1 and S4, in which iron electrolysis was performed, the recirculation ceased during period 4, and the recirculation ratio was set to 10 or higher in period 5. Compared to period 3, with a normal recirculation ratio, a significant difference in the average PO₄-P value was recognized only in period 4 for S1 (P < 0.01). Average PO₄-P in period 4 was lower than that in period 3 in S1. This indicated that deterioration of phosphorus removal was not observed even if the recirculation stopped or operated at a very high ratio. The fact that a significant difference was found implied that there was a possibility that the highly accumulated iron in the aerobic tank, by stopping the recirculation, contributed effectively to phosphorus removal. In contrast, during period 5, where the recirculation ratio was increased, phosphorus removal did

![Figure 3](https://iwaponline.com/wst/article-pdf/78/6/1304/504457/wst078061304.pdf)  
**Figure 3** | Change of PO₄-P.

![Figure 4](https://iwaponline.com/wst/article-pdf/78/6/1304/504457/wst078061304.pdf)  
**Figure 4** | Relationships between Fe/P molar ratio and PO₄-P.
not decrease compared to period 3. The large part of the added Fe was returned into the anoxic tank due to the high recirculation ratio. The Fe accumulated in the anoxic tank would contribute to the prevention of the phosphorus release. For further improvement of phosphorus removal, analysis of the Fe and phosphorus reaction mechanisms in the aerobic and anoxic tanks will be required.

It had been reported that phosphorus removal improved when calcium was rich (Li et al. 2013; Mishima et al. 2017). Mishima et al. (2017) investigated improvement in phosphorus removal through iron electrolysis with calcium addition. In some cases where calcium was lacking, phosphorus removal performance was poor, and when calcium was added, phosphorus removal improved. If the amount of calcium became more than 20 mg/L, an improvement was hardly observed. Therefore, Mishima et al. (2017) pointed out that it was desirable for the calcium amount to be 20 mg/L or more for the efficient phosphorus removal. In this study, the calcium amount of calcium was approximately 20–25 mg/L; therefore, it was suggested that phosphorus removal inhibition due to a lack of calcium did not occur because enough calcium was in the investigated SWTPs.

Mishima et al. (2017) also reported that the amount of Fe binding to phosphorus decreased because the organic matter formed a complex with iron when the DOC in the bulk of the wastewater was high, and phosphorus removal consequently deteriorated. In this study, the average DOC was 9.2 mg/L during all periods; however, at times, the DOC values were high, with a maximum of 18 mg/L. It had the possibility that the phosphorus removal performance was improved further if DOC could be decreased.

**Behavior of phosphorus in tanks**

The effects of changing PO$_4$-P in anoxic tank 1, the aerobic tank, and effluent in S2 and S4 are illustrated in Figure 5. The level of PO$_4$-P decreased slightly from the anoxic tank to the effluent in S2 without iron electrolysis, and was almost constant during period 1 in S4 without iron electrolysis. PO$_4$-P in the anoxic tank during period 2 in S4 was higher than that during period 1 in S1 and during all periods in S2. Moreover, PO$_4$-P in the effluent was higher than that in the aerobic tank in S4. These results indicate that the amount of accumulated phosphorus in the tank as a result of the effect of iron electrolysis increased, and part of the accumulated phosphorus was released in an anaerobic condition at the bottom of the tank. PO$_4$-P existed in a low concentration from the anoxic tank to effluent during period 3 in S4. This result indicated that the Fe addition by iron electrolysis contributed to not only the direct production of FePO$_4$ in the aerobic tank, but also the prevention of anaerobic phosphorus release from the sludge.

**Role of iron**

Figure 6 shows the relationships between the Fe content and PO$_4$-P in the filtrate in anoxic tank 1 and the aerobic tank, during periods 1 to 4. In this case, the maximum Fe content in the aerobic tank was approximately 400 mg/g. The Fe content in the aerobic tank was higher than that in anaerobic tank, because iron electrolysis was performed directly in
the aerobic tank, and substances such as organic matter suspended in the influent were accumulated in the anoxic tank. In particular, the PO₄-P in the filtrate was a maximum of 29 mgP/L in the anoxic tank (during period 2 of S5), which was much higher than the influent phosphorus concentration. The maximum PO₄-P in the aerobic tank was 4.8 mgP/L. The PO₄-P in the filtrate in both the anoxic and aerobic tanks decreased with an increase in Fe content in the sludge samples. It was indicated that phosphorus release from the sludge to the bulk phase was prevented by the Fe, which was supplied by iron electrolysis and accumulated at the bottom of the SWTPs, resulting in stable phosphorus removal.

**Economy of this system**

LIME2 is an LCA tool that can estimate the integrated environmental load as an economic value; this tool had already applied to evaluate a wastewater treatment plant (Mishima et al. 2016). Therefore, the economy of phosphorus removal by iron electrolysis was evaluated using this tool.

A comparison of the calculated results of phosphorus removal costs between electric power consumption and environmental load reduction by T-P removal is shown in Table 2. The electric power consumption cost was in the same range as that of the environmental load reduction by T-P removal, with the latter being 30% higher than the former. Therefore, it was demonstrated that the effect of environmental load reduction due to phosphorus removal by iron electrolysis was greater than the cost of power consumption for iron electrolysis. Further studies are needed in order to evaluate the integrated the environmental impact discharged from SWTP during whole life cycle.

**CONCLUSIONS**

Long-term phosphorus removal from SWTPs was investigated and optimum operational conditions for phosphorus removal by iron electrolysis were analyzed in this study. Efficient phosphorus removal can be achieved for a long time by adjusting the amount of iron against the actual population equivalent. Changing the recirculation ratio had no negative effect on overall phosphorus removal. Phosphorus release to the bulk phase was prevented by the accumulated iron, which was supplied by iron electrolysis, resulting in stable phosphorus removal. The effect of environmental load reduction due to phosphorus removal by iron electrolysis was greater than the cost of power consumption for iron electrolysis.

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