*Desalination and Water Treatment*
www.deswater.com

doi: 10.5004/dwt.2017.11446

63 (2017) 442–448
February

# Evaluation on the nutrient reduction in sewage systems using ultra-rapid coagulation

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Received 15 December 2014; Accepted 2 February 2016

**ABSTRACT**

This study was conducted to test the removal of nutrients in combined sewer overflows (CSOs) by using an ultra-rapid coagulation (URC) process. Alum and poly aluminum chloride (PAC) coagulants were each used to treat ten separate CSOs caused by rainfall events. Optimal injection ratios of 3 mol P/mol Al for alum and 2 mol P/mol Al for PAC were used, as determined by a preparatory experiment based on the phosphorous concentration of raw water. Total nitrogen (TN) removal efficiency was low for both coagulants, with only 33.5% for alum and 37% for PAC. In contrast, total phosphorus (TP) removal efficiency for alum and PAC were 86.1% and 91.4%, respectively. Removal efficiency of various phosphorous forms, including orthophosphate (Ortho-P), organic-phosphate (Org. P), and polyphosphate (Poly-P), in the raw water was highest for Ortho-P (alum 99.3%, PAC 99.6%). The sum of Poly-P and Org. P removal efficiency was lower than Ortho-P alone (alum 69.1%, PAC 77.9%). Removal efficiency of phosphorus increased with higher concentrations of particulate matter in the raw water.

*Keywords:* CSOs; Nonpoint source pollutant; Precipitation; Storm water; Runoff; URC process

## 1. Introduction

Nonpoint pollution source (NPS) generated during the wet season has not been properly treated in South Korea because governmental regulation of water quality pollution has concentrated on managing point sources. This has resulted in an increase in eutrophication in rivers and lakes and damage to aquatic ecosystems. Therefore, in order to improve the water quality, NPS together with the point source pollutants have been giving more attention.

NPS that came from dust, waste, fertilizer, agricultural pesticides, animal and plant carcasses, and atmospheric acid and deposition are carried by storm water runoff during rainfall events. Also, combined sewer overflows (CSOs) that is a part of NPS includes pollutants such as organic matter, bacteria, ammonia, turbidity, total suspended solids (TSS), and toxic and acidic materials [1,2]. Based on the 2012 Korean standard, the combined sewer system (CSS) is 45,682,547 m long. The separated sewer system (SSS) is divided into two parts, the sewer pipe, which is 41,739,397 m long, and the storm sewer, which is 29,080,459 m long. The length of the CSS is almost twice the length of the sewer pipe portion of the SSS, leading to CSOs greatly influencing pollutant loads in rivers.

In 2012, four major rivers in Korea were transformed from free-flowing to lake-like because of four river projects. Therefore, concern about controlling eutrophication-generating materials, such as nitrogen and phosphorus, has been increasing. Because algae use nitrogen and phosphorus to grow, it is important to control their discharge into rivers to prevent algal blooms. About 75% of the nitrogen and 50% of

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*Presented at Diffuse Pollution Conference/Asian Regional Conference (DIPCON/ARC 2014) Kyoto University, Japan, 3–4 September 2014.*

1944-3994/1944-3986 

the phosphorus attributed to eutrophication in Korea is from NPS pollution [3]; therefore, control of NPS pollution, including CSOs, is needed to prevent algal blooms.

To control NPS pollution from CSOs, advanced countries have been using physicochemical treatment processes that can sensitively cope with changes in flow rate. Some of these treatments, such as Actiflo and DensaDeg, use a high-speed coagulation sedimentation process [4,5]. In addition to using ultra-rapid coagulation (URC) as a coagulation sedimentation process, Korea has also treated CSOs with vortex separators [6] and swirl systems [7].

The advantages of a URC process include the ability to cope with changes in runoff flow and water quality during wet weather and to remove high levels of phosphorus, a major cause of eutrophication and algae blooms [8]. In an attempt to utilize these advantages, this study treated CSOs using a URC process that, in particular, treated nutrient salts with aluminum sulfate (alum) and poly aluminum chloride (PAC) as coagulants. The objectives of this study were to determine nitrogen and phosphorus removal efficiency by the coagulants alum and PAC and to determine how the removal efficiency was affected by the concentration of particulate matter or the nutrient form in the influent.

## 2. Study methods

### 2.1. Raw water and URC process

The URC process was installed in a sewage treatment plant (STP) and CSOs generated during rainfall events were used as the source for raw water samples. The total drainage area is 998 ha and includes a large-scale industrial complex immediately upstream, with residences and businesses farther upstream. Detailed land-use includes 52.6% industrial complex, 32.5% residential area, 21.3% forest, and 2.6% farmland. During rainfall events, storm water runoff discharged from the 998 ha area are combined with sewage and the resulting mixture flowed into the URC system.

The URC system used in this study has a 30,000-ton capacity (Fig. 1). This URC system is composed of a grit chamber, a retention tank, a fast reactor, a slow flocculation tank, and a settling tank. The grit chamber removes coarse-grained sands and the retention tank completely mixes contaminants through underwater aeration. In the fast reactor tank, the experimental coagulants are injected to combine with pollutants in the raw water. A polymer coagulant injected in the slow flocculation tank coheres fine flocs and makes large flocs that sediment quickly. Generated flocs poured into the settling tank are precipitated out quickly by the inclination plate settler and extracted with a pump.

This study used alum ($Al_2(SO_4)_3$•$18H_2O$, 8% as $Al_2O_3$) and PAC (17% as $Al_2O_3$) as coagulants to cohere pollutants and an anionic polymer coagulant was used as a polymer coagulant. An alkaline environment is important for coagulation. During preliminary research, the pH of the raw water was found to be high, but NaOH was injected to keep the water proper alkaline during the experiment. A theoretical calculation of the optimal dose of alum and PAC is possible. However, determining efficient and economical coagulant dosages through jar testing is thought to be better because the amount of injected coagulants can change depending on the raw water composition [9, 10]. Experiments to determine the optimal dosage of coagulants through jar testing was conducted before this study started (Table 1). Removal efficiency and economics were best balanced at the dose of 3.0 mol P/mol Al for alum and 2 mol P/mol Al for PAC, and therefore, these coagulant dosages were used in this study.

### 2.2. Sample collection and analysis methods

Sample collection was conducted during twenty rainfall events between 2011 and 2012. The characteristics of the rainfall events are shown in Table 2. Antecedent dry days (ADD) ranged from 2 to 12 and rainfall amount widely ranged from 3 to 165 mm. Average rainfall intensity ranged from 0.25 to 4.79 mm/h. During a storm event, 12 influent samples were collected at 1-h intervals from the retention tank during complete mixing. In consideration to the first operation state of URC process, sampling of treated water was collected 2 h after the operation has stabilized. To determine

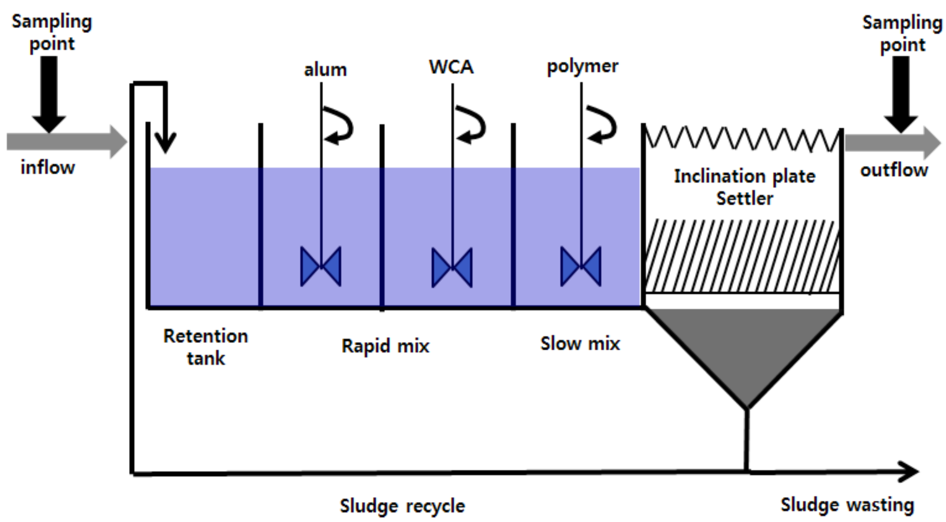


Fig. 1. Schematic diagram of the system used in this study.

Table 1
Proper injection ratios of coagulants

| mol P/mol Al | | 0.5 | 1 | 1.5 | 2 | 2.5 | 3 | 3.5 | 4 | 4.5 | 5 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Initial TP concentration (mg/L) | | 8.3~10.3 | | | | | | | | | |
| Residual phosphorus concentration (mg/L) | Alum 1 | 4.89 | 3.11 | 2.32 | 1.32 | 0.91 | 0.46 | 0.31 | 0.36 | 0.37 | 0.36 |
| | Alum 2 | 4.12 | 2.98 | 2.13 | 1.45 | 0.76 | 0.45 | 0.41 | 0.38 | 0.37 | 0.39 |
| | Alum 3 | 3.91 | 3.01 | 2.61 | 1.03 | 0.75 | 0.41 | 0.39 | 0.38 | 0.34 | 0.31 |
| | Average | 4.03 | 3.03 | 2.35 | 1.2 | 0.8 | 0.44 | 0.37 | 0.37 | 0.36 | 0.35 |
| | PAC 1 | 3.49 | 2.51 | 1.09 | 0.51 | 0.49 | 0.38 | 0.39 | 0.42 | 0.35 | 0.38 |
| | PAC 2 | 4.34 | 2.25 | 1.01 | 0.59 | 0.41 | 0.32 | 0.35 | 0.31 | 0.33 | 0.31 |
| | PAC 3 | 3.87 | 2.53 | 0.81 | 0.38 | 0.44 | 0.35 | 0.34 | 0.33 | 0.29 | 0.31 |
| | Average | 3.9 | 2.43 | 0.97 | 0.49 | 0.44 | 0.35 | 0.36 | 0.35 | 0.32 | 0.33 |

Table 2
Summary of rainfall data and concentration of pollutants for monitoring events

| Event No. | ADD (d) | Rainfall (mm) | Aver. rainfall intensity (mm/h) | Aver. TN (mg/L) | Aver. TSS (mg/L) | Aver. TP (mg/L) |
|---|---|---|---|---|---|---|
| Min | 2.0 | 3.0 | 0.3 | 1.5 | 140.5 | 2.6 |
| Max | 12.0 | 065.5 | 4.8 | 21.8 | 430.0 | 20.0 |
| Average | 6.4 | 22.9 | 1.2 | 15.5 | 239.8 | 8.5 |
| Standard deviation | 2.8 | 38.4 | 1.2 | .5 | 77.1 | 5.2 |

the removal efficiency of phosphorus and nitrogen forms, the water quality parameters of total nitrogen (TN), ammonia nitrogen ($NH_4$-N), organic nitrogen (Org. N), nitrite+nitrate ($NO_2$+$NO_3$, NOx), total phosphorous (TP), organic phosphate (Org. P), polyphosphate (Poly-P), orthophosphate (Ortho-P), and total suspended solids (TSS) were analyzed in the laboratory in accordance with standard methods [11]. $NO_2$ and $NO_3$ were analyzed individually, and the sum of these concentration values was reported as $NO_x$. The sum of Org. P and Poly-P concentration values were presented as Org.-Poly P.

## 3. Results and discussion

### 3.1. Removal efficiency by nitrogen forms

The removal efficiency of TN, Org. N, $NH_4$-N, and NOx by the alum and PAC coagulants is shown in Tables 3 and 4, respectively. While treating CSOs using the alum coagulant, the inflow TN concentration ranged from 10.8 to 21.7 mg/L and the average TN concentration was 15.3 mg/L. The inflow TN concentration during events treated by the PAC coagulant ranged from 12.7 to 20.7 mg/L with a mean of 16.7 mg/L. When alum and PAC were used as coagulants, the removal efficiency of TN was 33.5% and 37%, respectively. TN removal efficiency by the PAC coagulant is a little higher than that of the alum coagulant. However, both coagulants showed low TN removal efficiency. Kim [9] explained that this is because TN is removed much better through co-precipitation than chemical flocculation. Our study determined the removal efficiency of $NH_4$-N, Org. N, and $NO_x$, which are the nitrogen forms that compose TN. As a result, the removal efficiency by both coagulants was better for Org. N than either $NH_4$-N or $NO_x$. Org. N removal efficiency of the alum coagulant was 42.2%, while those of $NH_4$-N and $NO_x$ were determined to be the relatively low amounts of 17.7% and 12.7%, respectively. Org. N removal efficiency after injecting PAC was 42.1% and was higher than the 28.6% and 17.4% removal efficiency of $NH_4$-N and NOx, respectively. The higher percentage of removal efficiency of Org. N is likely due to the particulate portion of Org. N making coagulation relatively easy. Overall, removal efficiency of TN, $NH_4$-N, and $NO_x$ when PAC was injected was higher than when injecting alum.

### 3.2. Phosphorus removal efficiency

The removal efficiency of TP per coagulant type is shown in Table 5. The amount of metallic salts needed to cohere phosphorus is in the range of 1.5 to 2.5 mol Al/mol P, which is about 1.5 times more than the quantity demanded in stoichiometry [12]. However, our preliminary jar testing found the ratios of 3 mol Al/mol P for alum and 2 mol Al/mol P for PAC showed the most appropriate balance of removal efficiency and economics. Therefore, these coagulant ratio dosages were used in this study.

The concentrations of TP measured in rainfall event inflows in which alum was used were in the range of 3.3 to 19 mg/L with a mean of 8.6 mg/L. The concentrations of TP in the outflows were determined to range from 0.4 to 1.9 mg/L with a mean of 0.8 mg/L. Meanwhile, TP concentrations in rainfall event inflows where PAC was used were in the range of 2.6 to 20.0 mg/L with a mean of 8.3 mg/L. Concentrations of TP in the outflows when using PAC were discovered to be from 0.2 to 1.3 mg/L and the average TP was 0.6 mg/L. These results show treatment with PAC has higher TP removal efficiency than treatment with alum. In comparison, TP removal efficiency of alum was 86.1% and PAC was 91.4%. A lower dose of PAC was injected than of alum, yet PAC showed higher removal efficiency. The outflow concentrations and removal efficiencies after arranging TP inflow concentrations in ascending order are shown in Figs. 2 and 3 for alum

Table 3
Nitrogen removal efficiency of alum coagulant

| | Inflow concentration (mg/L) | | | | Outflow concentration (mg/L) | | | | Removal efficiency (%) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | TN | $NH_4$-N | Org. N | $NO_x$ | TN | $NH_4$-N | Org. N | $NO_x$ | TN | $NH_4$-N | Org. N | $NO_x$ |
| Min. | 10.8 | 2.8 | 7.2 | 0.3 | 7.0 | 2.1 | 4.3 | 0.2 | 25.7 | 11.2 | 34.1 | 6.1 |
| Max. | 21.7 | 7.1 | 14.8 | 1.3 | 13.2 | 6.2 | 9.2 | 1.1 | 40.0 | 26.3 | 51.3 | 20.3 |
| Average | 15.3 | 4.9 | 10.1 | 0.6 | 10.5 | 4.0 | 5.7 | 0.5 | 33.5 | 17.7 | 42.2 | 12.7 |

Table 4
Nitrogen removal efficiency of PAC coagulant

| | Inflow concentration (mg/L) | | | | Outflow concentration (mg/L) | | | | Removal efficiency (%) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | TN | $NH_4$-N | Org. N | $NO_x$ | TN | $NH_4$-N | Org. N | $NO_x$ | TN | $NH_4$-N | Org. N | $NO_x$ |
| Min. | 12.7 | 2.7 | 7.2 | 0.3 | 7.8 | 2.1 | 4.3 | 0.3 | 29.2 | 15.8 | 30.3 | 4.6 |
| Max. | 20.7 | 7.5 | 13.2 | 0.9 | 14.2 | 5.1 | 9.2 | 0.8 | 45.1 | 41.9 | 51.2 | 27.7 |
| Average | 16.7 | 5.7 | 10.3 | 0.5 | 10. | 4.0 | 5.9 | 0.5 | 37.0 | 28.6 | 42.1 | 17.4 |

Table 5
Total phosphorus removal efficiency

| | Alum | | | PAC | | |
|---|---|---|---|---|---|---|
| | TP inflow concentration (mg/L) | TP outflow concentration (mg/L) | Removal efficiency (%) | TP inflow concentration (mg/L) | TP outflow concentration (mg/L) | Removal efficiency (%) |
| Min. | 3.3 | 0.4 | 74.5 | 2.6 | 0.2 | 86.3 |
| Max. | 19.0 | 1.9 | 91.4 | 20.0 | 1.3 | 93.4 |
| Average | 8.6 | 0.8 | 86.1 | 8.3 | 0.6 | 91.4 |

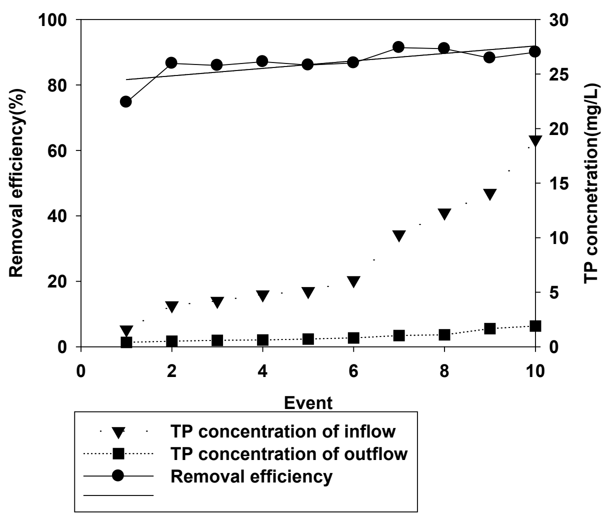


Fig. 2. Removal efficiency vs. concentration of TP (alum).

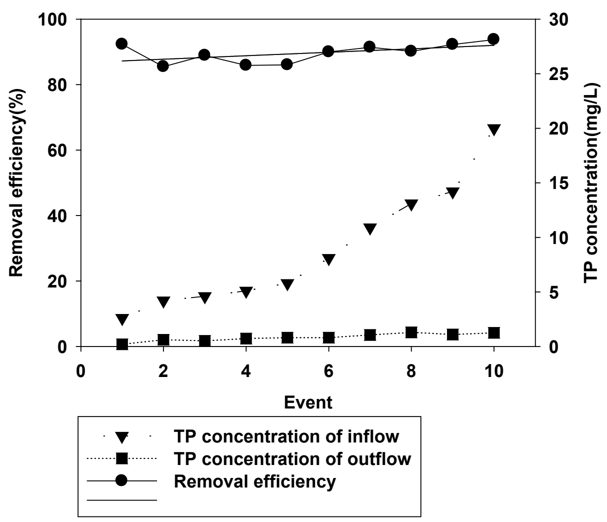


Fig. 3. Removal efficiency vs. concentration of TP (PAC).

and PAC, respectively. As TP concentration increased, the removal efficiencies of both the alum and PAC coagulants also increased.

Table 6 displays the variation of TP removal efficiencies based on inflow TP concentration. When the average TP concentration in the inflow was lower than 5 mg/L, the TP removal efficiency of the alum coagulant was 83.8%, whereas that of the PAC coagulant was 90%. Meanwhile, when the average TP concentration in the inflow was higher than 15 mg/L, TP removal efficiency of the alum coagulant was 91.4%, whereas that of the PAC coagulant was 93.4%. It is noteworthy that even though TP removal efficiency increased with higher inflow TP concentrations, residual TP concentrations in the outflows also increased. This is important to consider when trying to remove TP from CSOs, because it means that if TP concentration in the inflow is increased, an additional treatment process should be installed to remove more of the TP or an increase in coagulant dosage should be

Table 6
Total phosphorus removal efficiency by TP concentration

| TP concentration (mg/L) | Alum | | | PAC | | |
|---|---|---|---|---|---|---|
| | TP inflow concentration (mg/L) | TP outflow concentration (mg/L) | Removal efficiency (%) | TP inflow concentration (mg/L) | TP outflow concentration (mg/L) | Removal efficiency (%) |
| <5 | 4.2 | 0.7 | 83.8 | 4.2 | 0.4 | 90.0 |
| 5~10 | 7.0 | 1.0 | 85.9 | 6.50 | 0.45 | 93.08 |
| 10~15 | 12.2 | 1.5 | 87.7 | 12.6 | 0.9 | 92.6 |
| >15 | 19.0 | 1.63 | 91.4 | 20.03 | 1.31 | 93.46 |

Table 7
Removal efficiency by phosphorus form (alum)

| | Inflow concentration (mg/L) | | Outflow concentration (mg/L) | | Removal efficiency (%) | |
|---|---|---|---|---|---|---|
| | Ortho-P | Org- Poly. P | Ortho-P | Org- Poly. P | Ortho-P | Org- Poly. P |
| Min. | 1.9 | 1.4 | 0.02 | 0.3 | 99.0 | 41.2 |
| Max. | 10.2 | 8.8 | 0.04 | 1.9 | 99.7 | 85.5 |
| Average | 4.7 | 3.9 | 0.03 | 1.0 | 99.3 | 69.1 |

Table 8
Removal efficiency by phosphorus form (PAC)

| | Inflow concentration (mg/L) | | Outflow concentration (mg/L) | | Removal efficiency (%) | |
|---|---|---|---|---|---|---|
| | Ortho-P | Org- Poly. P | Ortho-P | Org- Poly. P | Ortho-P | Org- Poly. P |
| Min. | 1.4 | 1.2 | 0.005 | 0.22 | 99.4 | 53.4 |
| Max. | 13.2 | 6.8 | 0.03 | 1.28 | 99.9 | 87.2 |
| Average | 4.9 | 3.4 | 0.01 | 0.64 | 99.6 | 77.9 |

used to decrease TP loads discharged into rivers. However, the amount of alum and PAC should be applied according to the directions because the use of increased amounts of alum and PAC could cause harm to aquatic ecosystems [13].

### *3.3. Treatment efficiency according to phosphorus form*

Phosphorus in water is divided into Org. P, Poly-P, and Ortho-P. All inorganic phosphate is gradually hydrolyzed and transformed to Ortho-P. According to Elisabeth and Roland [14], the phosphate in raw water combines with the aluminum salts in coagulants, forming insoluble particles. Because TP treatment methods can vary depending on the phosphorus form found in CSOs, this study examined the treatment efficiency by phosphorus form in influents (Tables 7 and 8).

In the 10 events when the alum coagulant was used, the inflow Ortho-P concentration widely ranged from 1.9 to 10.2 mg/L with a mean of 4.6 mg/L. The concentration of Org.-poly P ranged from 1.4 to 8.8 mg/L with a mean of 3.9 mg/L. In the 10 events using PAC, the concentration of Ortho-P and Org.-poly P ranged from 1.4 to 13.2 mg/L and 1.2 to 6.8mg/L, respectively, and the averages were 4.9 mg/L and 3.41 mg/L, respectively. In case of alum coagulant, Ortho-P accounted for 55.4% of TP in the inflow. In the events when PAC was used as the coagulant, as Ortho-P and org.-poly P was 59.1% and 40.8% respectively, it was examined that Ortho-P concentration in the inflow when using PAC accounted for slightly higher rate than events when alum was used. Ortho-P in treated water was discharged at very low concentrations for both coagulants and both coagulants showed high Ortho-P removal efficiency as seen by the average removal efficiency of alum of 99.3% and 99.6% for PAC. It is likely that the reason, Ortho-P which showed high removal efficiency was due to the combine processes of Al(III) ions that were included in the alum coagulant and dissolved phosphorus which induced the formation of insoluble or low solubility salts that was precipitated [15]. Meanwhile, the average concentration and removal efficiency of Org.-poly P in alum treated water was 1.0 mg/L and 69.1%, respectively. Water treated with PAC had an average concentration and removal efficiency of org.-poly P of 0.64 mg/L and 77.9%, respectively. Therefore, PAC showed higher org.-poly P treatment efficiency than alum. As shown in Figs. 4–7, Ortho-P concentrations in the inflow differed in each rainfall event. However, most of the Ortho-P was treated using alum or PAC regardless of the Ortho-P concentration in the inflows. Most of the TP in the outflows was Org. P and Poly-P.

It is well known that PAC has a higher pollutant removal efficiency than alum. Moreover, based on this study, the TP concentration in the inflow of the events treated with PAC was higher than the inflow TP concentration of events when alum was used.

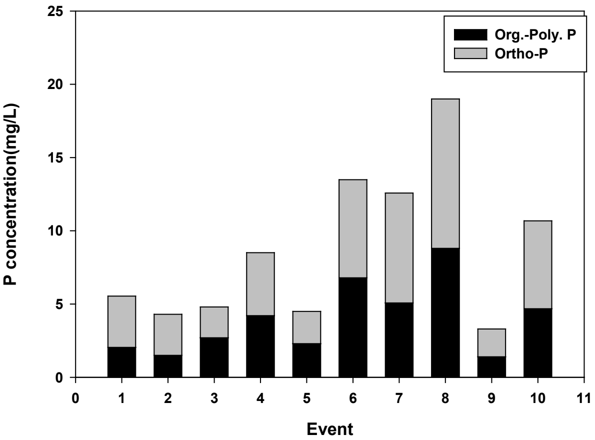


Fig. 4. Phosphorous speciation in the inflow (alum).

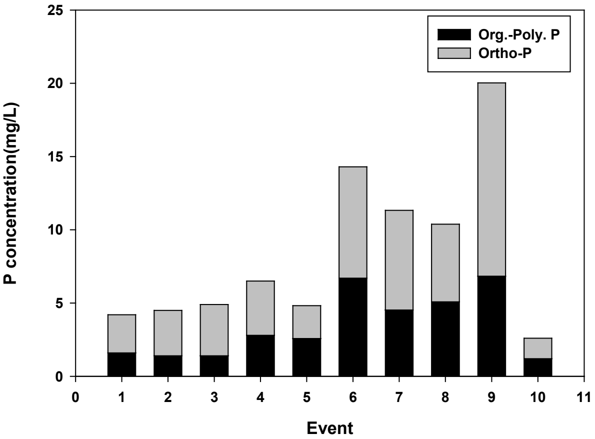


Fig. 5. Phosphorous speciation in the inflow (PAC).

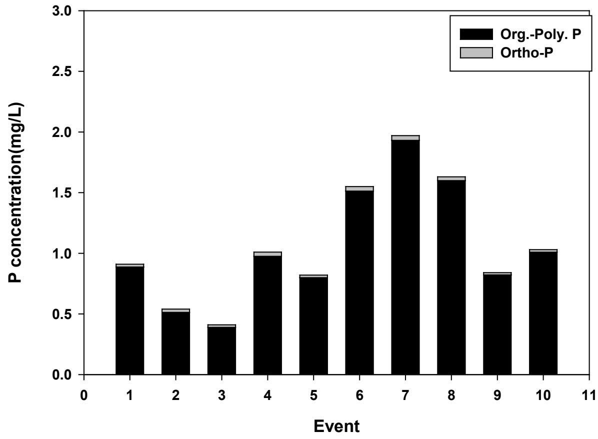


Fig. 6. Phosphorous speciation in the outflow (alum).

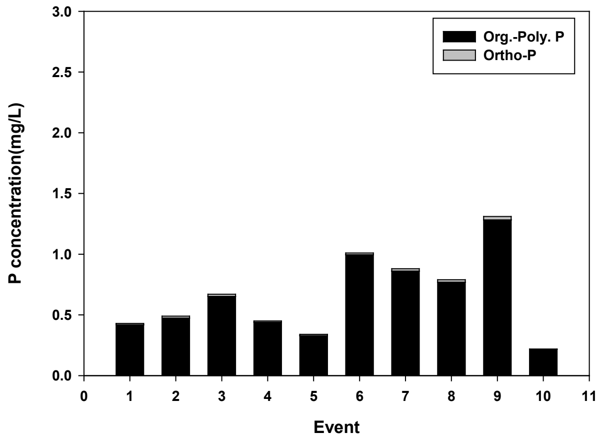


Fig. 7. Phosphorous speciation in the outflow (PAC).

### *3.4. Phosphorus removal efficiency by inflow TSS concentration*

Examination of TSS concentration in the inflows with respect to the TSS concentration, which was classified to be above or below 200 mg/L, was performed in order to determine the influence of TSS on TP removal efficiency in the URC process (Table 9).

Three of the 10 rainfall events when the alum coagulant was used had a TSS concentration lower than 200 mg/L, whereas the TSS concentration for the seven other events was higher than 200 mg/L. Five of the 10 rainfall events when the PAC coagulant was used had a TSS concentration lower than 200 mg/L and five of the events had TSS concentrations higher than 200 mg/L. When the alum coagulant was used in events with TSS concentrations above 200 mg/L, TP removal efficiency (87.4%) was slightly higher than removal efficiency (84.8%) when TSS concentrations were below 200 mg/L. The same result was also seen when PAC was used. The removal efficiency of TP when TSS concentrations were above 200 mg/L was 92.7% and it was 91.1% when TSS concentrations were below 200 mg/L. It is likely this is because with higher amounts of suspended solids, it is easier for flocs to form [16,17] and pollutants such as TSS, biochemical oxygen demand (BOD), chemical oxygen demand (COD), and phosphorus that exists as particle materials to be aggregated

Table 9
TP removal efficiency by TSS concentration

| Coagulant type | TSS concentration (mg/L) | Average TSS concentration (mg/L) | Inflow average TP concentration (mg/L) | Outflow average TP concentration (mg/L) | Efficiency (%) |
|---|---|---|---|---|---|
| Alum | <200 | 167.9 | 9.3 | 1.1 | 84.8 |
| | >200 | 236.1 | 8.4 | 1.0 | 87.4 |
| PAC | <200 | 185.3 | 7.8 | 0.7 | 90.1 |
| | >200 | 342.7 | 8.9 | 0.6 | 92.7 |

by the coagulants. As mentioned in the previous section, as TP concentration increased in the inflow, TP removal efficiency also increased. The results in this section show that when the alum coagulant was used, the TP concentration in the inflow when the TSS concentration was above 200 mg/L was lower than that when the TSS concentration was below 200 mg/L. However, TP removal efficiency was higher when the TSS concentration was above 200 mg/L. In the results above, it is thought that with increased TP concentration in the inflow, the URC process removal efficiency increases, and the amounts of suspended solids influence the TP removal efficiency.

## 4. Conclusions

To examine the removal efficiency of nitrogen and phosphorus forms by CSOs, a real scale URC process using the alum and PAC coagulants was performed, and the results are shown below.:

(1) Higher TN removal efficiency was seen with PAC (37%) than with alum (33.5%). Of the nitrogen forms, $NH_4$-N, Org. N, and $NO_x$, both coagulants removed Org. N. with the highest efficiency.
(2) Higher TP removal efficiency was seen with PAC (91.4%) than with alum (86.1%). In the analysis of TP removal, efficiency depended on the TP concentration in the inflow. It was determined that when the TP concentration in the inflow increased, the TP removal efficiency of both the alum and PAC coagulants increased.
(3) Investigation of the removal efficiency of the phosphorus forms, Ortho-P, Org. P, and Poly-P, showed that Ortho-P removal with the alum coagulant was 99.3% and that when the PAC coagulant was used, it was 99.6%. Regardless of rainfall events characteristics, Ortho-P showed high removal efficiency. The sum of Org. P and Poly-P removal efficiency by the alum and PAC coagulants was 69.1% and 77.9%, respectively.
(4) The removal efficiency of TP depended on the particulate matter concentration in the inflow for both the alum and PAC coagulants, with increased particulate matter concentrations showing increased removal efficiency. Therefore, this study determined that TP concentration and the amount of particulate matter in the inflow influences TP removal efficiency.

## Acknowledgement

This research was supported by a grant (E416-00020-0602-0) from Public Welfare Technology Development Program funded by Ministry of Environment of Korean government.

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