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Optimizing the H_3PO_4 leaching conditions of post-precipitated sewage sludge using response surface methodology

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Abstract

The leaching procedure of post-precipitated aluminium phosphate sludge with dilute phosphoric acid was developed. The leaching offers a route to recover both critical phosphorous from sewage sludge and the metal used in precipitation. Using phosphoric acid as leaching solution makes it possible to continue the recovery process without the need to remove chloride or sulfate anions. The optimization of the leaching was based on experimental three-level-four-variable central composite face-centered design. The four variables included were acid concentration, volume of acid, temperature and time of leaching. The leaching was conducted for dewatered and water-containing sludge (total solid content 3-4 %) and for both second-order regression models were obtained. For water-containing sludge optimal conditions for leaching are solid to liquid ratio (S/L) 400 gL^{-1}, a temperature of 60°C and a leaching time of 6 hours. For the dewatered sludge, optimal leaching is attained when S/L ratio 119 gL^{-1} with 2 M acid is used at a temperature of 20 °C. The obtained results enable the developing of full-scale process where phosphate in the sludge is refined to phosphorous acid and metal used in sludge production recycled back to precipitation.

1 Introduction

High phosphorus content in water systems accelerates eutrophication (Morse et al., 1998) and because of that its removal from wastewaters is required by the directive of European Commission Council (91/271/EEC). However at the same time, phosphorus is an irreplaceable element for food production as it is used in fertilizers (Cordell et al., 2009). The current phosphate rock deposits being depleted and for that reason European Commission listed it as a critical raw material in 2014 (European Commission, 2014). In 2017 the critical raw material listing was updated and elemental phosphorus was added to the list (European Commission, 2017). This encourages research on the possibility to recover vital phosphorus from secondary sources, like sewage sludge, and process it to a valuable product, for instance phosphorous acid.

Phosphorus is most often removed from wastewaters by chemical precipitation as a phosphate or via enhanced biological treatment (Bashar et al., 2018; Pratt et al., 2012). The sludge formation in chemical precipitation is high and currently the most common ways to process the sludge is incineration, using it in landfilling or agricultural use (Kacprzak et al., 2017). However the agricultural use of the sludge is restricted in some European countries because of the heavy metals, pathogens and micropollutants in the sludge (Kelessidis and Stasinakis, 2012; Veeken, 1999). Incinerating sewage sludge is widely used in European countries, but it is energy-consuming process and the heavy metals are enriched in the ash (Franz, 2008; Kelessidis and Stasinakis, 2012). The problem with heavy metals can be overcome with chemical post-precipitation where phosphorus is precipitated with aluminum or iron as phosphate salt in the end of wastewater process while still achieving the required removal efficiency of phosphorus (Eklund et al., 1991). Formed sludge can then be processed without the need to consider the possibility of heavy metals contaminating the end-product.

Leaching in alkaline or acidic conditions is one of the most widely investigated methods for recovery of phosphorus from sewage sludge, but most studies focus on incinerated sewage sludge ash (ISSA) (Donatello et al., 2010; Donatello and Cheeseman, 2013; Franz, 2008). Few studies concentrating on the leaching of the sludge has been reported, but these are focused on the sludge formed in conventional chemical precipitation (Levlin et al., 2002). Typically the leaching solutions investigated have been ones within lower price range such as inorganic sulfuric acid H_2SO_4, and nitric acid HNO_3, and organic acids like oxalic acid $C_2H_2O_4$ (Fang et al., 2018; Li et al., 2018). However, the problem with these is that excess sulfate or chloride needs to be removed from leaching solution (Ottosen et al., 2013) which adds another process step in the recovery process. Leaching with dilute phosphoric acid (H_3PO_4) would make it possible to continue the processing without the need to remove chloride or sulfate anions. Phosphorus is recovered as phosphoric acid due the low pH (Levlin and Hultman, 2004) and the metal used in precipitation step can be circulated back to the precipitation via ion exchange or solvent extraction (Levlin et al., 2004; Mohapatra et al., 2007).

The aim of this study is to optimize the leaching of moist or dried post-precipitated aluminium phosphate sludge with dilute phosphoric acid. To achieve this goal regression equations for both types of sludge are created with response surface methodology (RSM). The methodology is commonly used in natural sciences for optimization purposes (He et al., 2018; Sen et al., 2017). The obtained results serve as a first step in developing the phosphorous recovery process where not only phosphorous is recovered but also the metal used in precipitation step can be recycled.

2 Experimental

2.1 Materials and chemicals

The batch of post-precipitated aluminium phosphate sludge was received from RAVITA pilot plant located in Viikinmäki wastewater treatment plant, Helsinki. Before use the sludge was filtrated (Whatman no. 41 filtration paper). This resulted in sludge with solid content of about 3-4 %. A part of the filtrated sludge was held at 105 °C for 16 hours in order to dewater it and then manually ground in a mortar.

All chemicals were of analytical grade and were used as purchased without further purification. The standard stock solutions of elements (1000 mg L^{-1}) were supplied by PerkinElmer. Phosphoric acid (85 wt. %) was obtained from VWR International. Nitric acid (65-68 wt. %) and hydrochloric acid (37-39 wt. %) were purchased from Sigma-Aldrich. High-purity water produced by a Purelab Ultra water purification system supplied by Elga (Buckinghamshire, U.K.) was used throughout the experiments.

2.2 Methods

2.2.1 Digestion of sludge

A sludge sample (250 mg for moist and 100 mg for dry sludge) was accurately weighed into a 50 mL centrifuge tube (Sarstedt) and 10 mL of aqua regia (HCl + HNO_3) was added. The bottle was closed and placed into a 750 W, 37 kHz, Elmasonic P ultrasonic water bath supplied by Elma Schmidbauer GmbH. The ultrasound-assisted extraction was carried out at a temperature of 60 °C. The sonication was conducted in the series of 8 x 3 minutes. After each sonification step the samples were shaken by hand. After cooling, the sample solution was filtered (Whatman no. 41 filter paper) into a 50 mL volumetric flask. The residue was washed three times with a few milliliters of water and diluted with high purity water to a volume of 50 mL.

2.2.2 Leaching of sludge

The leaching was conducted in a water bath with manual temperature control. A sludge sample (2 g for moist and 1g for dry sludge) was accurately weighed into a 50 mL centrifuge tube and the volume of phosphoric acid depending on the experimental design was added. Samples were placed in water bath and were stirred with magnetic stirrer with the speed of 250 rpm (rounds per minute). After leaching samples were centrifuged for 10 minutes with speed 3500 rcf (relative centrifugal force) with Heraeus Labofuge 400 centrifuge supplied by Thermo Scientific and filtrated (Whatman no 41 filtration paper).

Because the pH during leaching is below 2.2 (Levlin and Hultman, 2004) the dissolution reaction is as follows:

$$AlPO_4 + 3\ H^+ \rightarrow Al^{3+} + H_3PO_4^0 \qquad (1)$$

The equation (1) proves that with high leaching efficiency of aluminium also phosphorus is leached simultaneously.

2.2.3 Experimental design

Central composite face-centered design with four independent variables (acid concentration (A), volume of acid (B), temperature (C) and time of leaching (D)) and three levels (± 1, 0), including six replicates at the center point, was used to design the experiments. The variables and their values are shown in table 1. The experimental runs were randomized in order to reduce bias from extraneous or uncontrollable conditions. The response variable measured was the concentration of aluminium in acid after leaching (*Y*). The concentration of phosphorus cannot be used as a response because phosphoric acid is used as leaching solution. The design was generated with Minitab 18 software. The experimental plan and the results are presented in supplementary data (Table S1).

Factors	Levels		
	Lowest	Center	Highest
	-1	0	+1
Acid concentration (M)	0.5	1.25	2.0
Volume of acid (mL)	5	10	15
Temperature (°C)	20	40	60
Time of leaching (h)	2	4	6

Table 1. Coded and actual levels of variables in the experimental design

2.2.4 Statistical analysis

The response surface analysis of the Minitab 18 software was used to analyze the experimental data (table S1) and to produce the response surface plots. The experimental data were fitted to a second-order polynomial model and regression coefficients obtained. The generalized second-order response surface model used in the response surface analysis was as follows:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum \sum_{i<j}^{k} \beta_{ij} x_i x_j + \epsilon, \qquad (2)$$

where y is the response (aluminium concentration), β_0 is constant coefficient, β_i is the linear effect, β_{ii} is the quadratic effect, β_{ij} is the interaction effect and ϵ is the error observed in response (Montgomery, 2009). After analysis Minitab 18 software's response optimizer feature was used to achieve maximum value for aluminium concentration within the range of factor levels.

2.2.5 ICP-OES measurements

The element concentrations were determined with a PerkinElmer ICP-OES (inductively coupled plasma- optical emission spectrometer) Optima 8300 –spectrometer. The GemCone low flow –nebulizer with cyclonic spray chamber was used for sample introduction. The measurement parameters for all measurements were argon gas flow of 8 L min^{-1}, nebulizer gas flow of 0.6 L min^{-1}, auxiliary gas flow of 0.2 L min^{-1}, sample flow rate of 1.5 L min^{-1} and the radio frequency power of 1500 W. The wavelengths, calibration ranges and the method detection limits are presented for each element in supplementary data (Table S2). The method detection limits were determined according to U.S Environmental Protection Agency method 200.7 (U. S. Environmental Protection Agency, 2001).

3 Results

3.1 Analysis

The total concentration of aluminium and phosphorus in post-precipitated sludge was analyzed. The concentrations (mean ± standard error) extracted by ultrasound-assisted digestion method and determined by ICP-OES are shown in table 2. Other analyzed elements are presented in supplementary data table S3.

	moist sludge [a]	dry sludge [b]
Al (mg kg^{-1})	6500 ± 400	188400 ± 800
P (mg kg^{-1})	1580 ± 80	44900 ± 300

[a] solid content 3-4 %
[b] dried in 105 °C for 16 h

Table 2. Determined element concentrations (mg kg^{-1}) of post-precipitated phosphorus sludge (n=3)

3.2 ANOVA analysis and fitting of second-order model

The second-order model was first fitted according to equation (2), but in order to achieve an improved model for the leaching conditions, insignificant model terms were removed. A backward elimination process with Minitab 18 software was chosen to automatically remove the insignificant terms. The resulting ANOVA (analysis of variance) data for the reduced quadratic models of moist and dried sludge leaching are given in tables 3 and 4, respectively. It can be seen from table 3 that the F-value for the model of reduced quadratic model of moist sludge leaching is 155.1 and the corresponding P-value is less than 0.0001. This means that the model is significant and can be used for optimization of leaching. The significant terms in this model are volume of acid (*B*), the quadratic effect of volume (B^2), the two-level interactions of volume and time ($B*D$) and the two- level interactions of temperature and time ($C*D$). Other insignificant terms remaining in the model are there to support hierarchy. The P-value for the lack-of-fit is 0.915, which indicates that it is not significant. The goodness-of-fit of the model can be checked from the coefficient of determination (R^2). The R^2-value is high, 97.59 %, which means that only 2.41 % of the total sample variation cannot be explained by the model. The adjusted R^2-value of 96.96 % indicates that all the factors included in the model are significant and affect the response variable. High predicted R^2-value 95.89 % proves that this model can predict proper responses for new observations.

From table 4 can be seen that F-value for the reduced quadratic model, where dried sludge is used for leaching, is 41.14 and the corresponding P-value is less than 0.0001. This model is also significant and can be used for optimization of leaching. The significant terms are acid concentration (*A*), volume of acid (*B*), the quadratic effect of volume (B^2), the quadratic effect of temperature (C^2) and the two-level interactions of acid

concentration and volume of the acid (*A***B*). Other insignificant terms remaining in the model are there to support hierarchy. The P-value for the lack-of-fit is 0.769, which indicates that it is not significant. The R^2-value is high, 91.48 %, which means that only 8.52 % of the total sample variation cannot be explained by the model. The adjusted R^2-value (89.25 %) and the predicted R^2-value (85.48 %) are both relatively high, meaning that the model fits to the data.

The regression equations for the leaching of moist and dried phosphorus sludge in uncoded units were obtained as follows:

Moist sludge:

$$Y = 4004 - 393.1B - 7.21C - 2.1D + 14.00B^2 - 6.17B*D + 1.76C*D \quad (3)$$

Dried sludge:

$$Y = -19488 + 18361A + 5949B - 711C - 259.8B^2 + 8.36C^2 - 795A*B \quad (4)$$

where *Y* is the concentration of the aluminum in acid after leaching, *A* concentration of the acid, *B* acid volume, *C* temperature and *D* leaching time. The regression equations obtained can be used to predict aluminium concentration in acid after leaching within the limits of the experimental factors. The model assumption checks are presented in supplementary data along with main effects plots.

Source	DF	Adj SS	Adj MS	F-Value	P-Value	Remarks
Model	6	9552419	1592070	155.10	0.000	Significant
Volume of acid (B)	1	8526540	8526540	830.66	0.000	
Temperature (C)	1	45	45	0.00	0.948	
Time (D)	1	3111	3111	0.30	0.587	
Volume of acid*Volume of acid (B^2)	1	882558	882558	85.98	0.000	
Volume of acid*Time (B*D)	1	60865	60865	5.93	0.023	
Temperature*Time (C*D)	1	79298	79298	7.73	0.011	
Error	23	236090	10265			
Lack-of-Fit	20	170898	8545	0.39	0.915	Not significant
Pure Error	3	65192	21731			
Total	29	9788508				
Model Summary:	R^2	R^2 (adj)	R^2 (pred)			
	97.59 %	96.96 %	95.89 %			

Table 3. ANOVA table for reduced quadratic model of moist sludge leaching (response: concentration of aluminium (mg L^{-1}))

Source	DF	Adj SS	Adj MS	F-Value	P-Value	Remarks
Model	6	1436763871	239460645	41.14	0.000	Significant
Concentration (A)	1	1097519032	1097519032	188.53	0.000	
Volume of acid (B)	1	26026670	26026670	4.47	0.046	
Temperature (C)	1	12670746	12670746	2.18	0.154	
Volume of acid*Volume of acid (B^2)	1	145294521	145294521	24.96	0.000	
Temperature*Temperature (C^2)	1	38511436	38511436	6.62	0.017	
Concentration*Volume of acid (A*B)	1	142190719	142190719	24.43	0.000	
Error	23	133890714	5821335			
Lack-of-Fit	20	108523781	5426189	0.64	0.769	Not significant
Pure Error	3	25366933	8455644			
Total	29	1570654585				
Model Summary:	R^2	R^2 (adj)	R^2 (pred)			
	91.48 %	89.25 %	85.48 %			

Table 4. ANOVA table for reduced quadratic model of dried sludge leaching (response: concentration of aluminium (mg L^{-1}))

3.3 Effect of the four variables on aluminium concentration in leaching acid

Based on the second-order models created in the previous section, response surface plots were made to investigate the effects of four variables (acid concentration, volume of acid, temperature and time of leaching), on the aluminium concentration in acid after leaching of moist or dried sludge. In both models one factor proved to be insignificant and was removed from the model. Hence, only three variables remained and in each surface plot one variable is kept at the central level (0), while considering the other two variables.

3.3.1 Factor effects when leaching the water containing sludge

In the reduced quadratic model for leaching of moist sludge only two interaction terms are present volume of acid*time and temperature*time. Those interactions can be investigated by drawing the 3D response plots (Figure 1a-c). In figure 1a, where the effects of temperature and acid volume to the aluminium concentration form a plane with a curving slope, an increase in aluminium concentration when volume decreases can be seen. Because concentration proved to be insignificant factor it can be deduced that H^+ concentration in 0.5 M phosphoric acid is sufficient to leach the aluminium and along with the phosphorus from the moist sludge. In that case, the volume becomes the regulating factor for aluminium concentration and any increase in volume decreases the aluminium concentration. A slight increase in the aluminium concentration is also obtained when leaching time is increased from 2 to 6 hours, but according to ANOVA analysis the change is so minor that temperature on its own does not enhance the leaching. The maximum value of 2250 mg L^{-1} for aluminium concentration is achieved when acid S/L ratio is 400 gL^{-1} and temperature is 60 °C. Figure 1b displays the effects of temperature and time on the aluminium concentration. When both factors are increased, temperature from 20 to 60 °C and time from 2 to 6 h, a maximum aluminium

concentration is attained. The interaction between these two factors enhances the leaching, but only when both factors are in higher level. Aluminium concentration is at maximum 1300 mg L^{-1} when temperature is 60 °C and leaching time is 6 hours. Figure 1c shows the effect of acid volume and leaching time to the aluminum concentration. The plot confirms the ANOVA analysis determination that the leaching time has no significant effect in sludge leaching, even though a small increase in aluminum concentration is seen when leaching time increases. Aluminium concentration is at maximum 2330 mgL^{-1} when S/L ratio is 400 gL^{-1} and leaching time is 6 hours.

At the current factor value ranges, the volume of acid is the dominant process variable. However, the leaching time and temperature have a synergetic effect and therefore cannot be discarded when considering optimal conditions for leaching process.

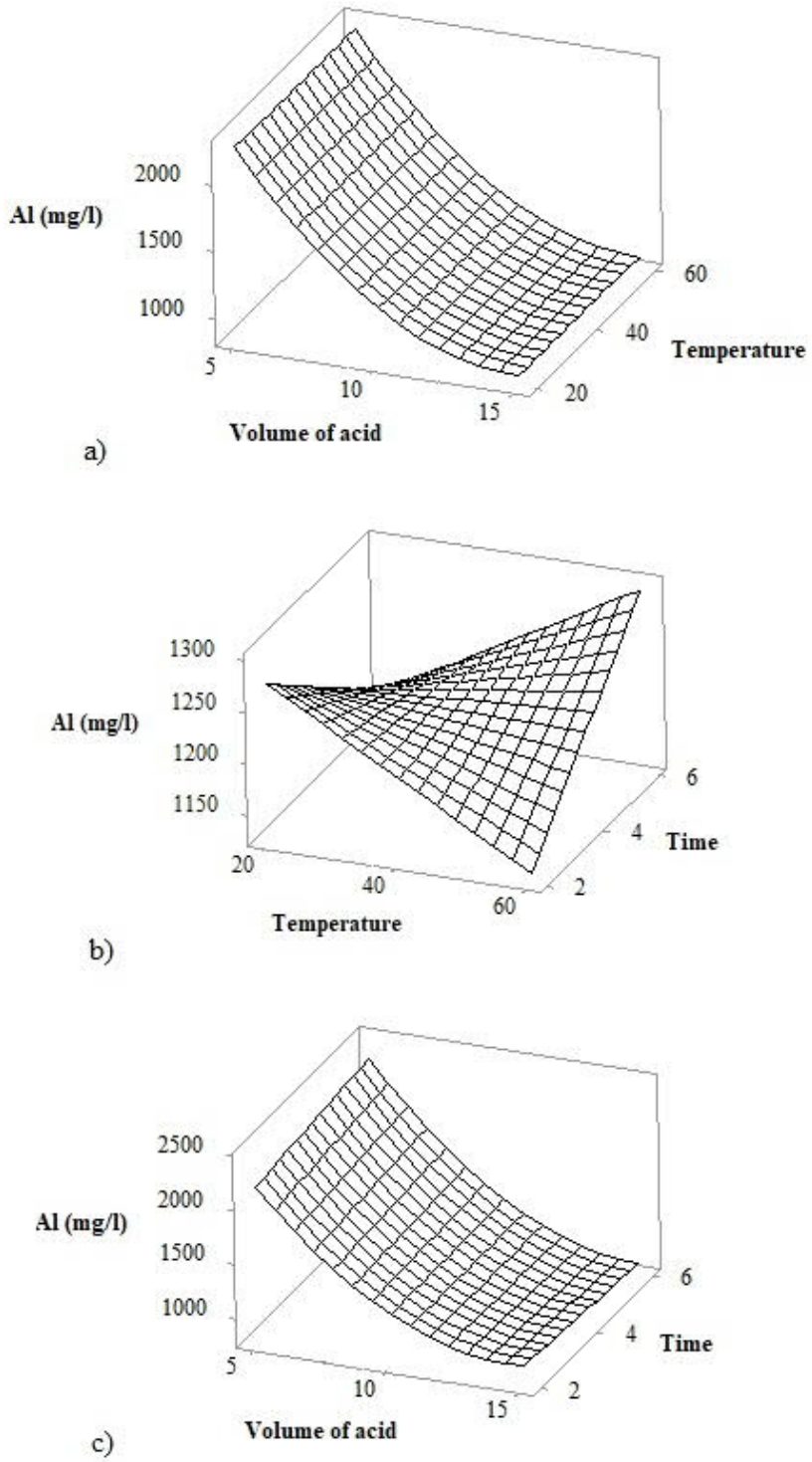
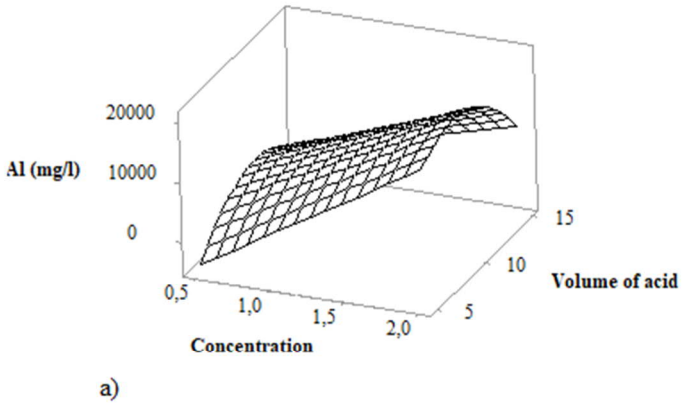


Figure 1. Changes in aluminium concentration (Al mg/l) with respect to: a) volume of acid (mL) and temperature (ºC), b) temperature (ºC) and time (h), c) acid volume (mL) and time (h) when leaching the moist sludge (m= 2 g)

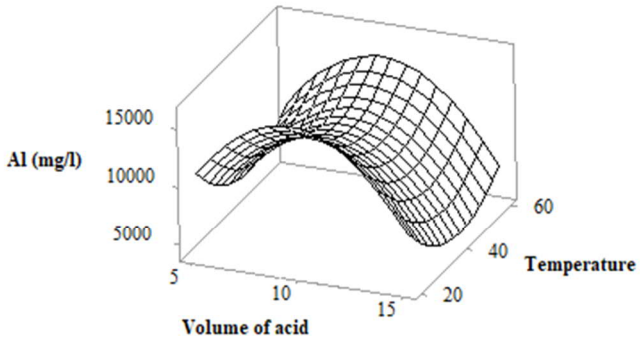
3.3.2 Factor effects when leaching the dewatered sludge

There is only one interaction term present in the reduced quadratic model for leaching of dewatered sludge, but both temperature and acid volume have significant quadratic effects in the model. In figure 2a can be seen that higher acid concentration increases the amount of leached aluminium. The rising ridge surface indicates that 2 M phosphoric acid is needed to maximize the leached aluminium. Due to the factor ranges it cannot be said if higher H^+ concentration is needed for better leaching efficiency. However, this would mean increasing the phosphate concentration in the leaching solution. This will cause re-precipitation of aluminium phosphate due to common ion effect and it is not for that reason a viable option. Within the factor levels a maximum aluminium concentration of 20.5 gL^{-1} is achieved when acid concentration is 2 M and S/L ratio is 100 gL^{-1}. Figure 2b shows the effects of temperature and volume of acid on the aluminium concentration. Due to the quadratic effects of both factors a saddle is formed. The highest aluminium concentration, 16.2 gL^{-1}, is reached at a low temperature (20 °C) and S/L ratio 102 gL^{-1}. Figure 2c displays effects of acid concentration and temperature on the aluminium concentration. Again the substantial effect of the leaching acid concentration on aluminium concentration is seen, while temperature has no influence. The highest value for aluminium concentration 24.0 gL^{-1} is achieved when acid concentration is 2 M and temperature is 20 °C.

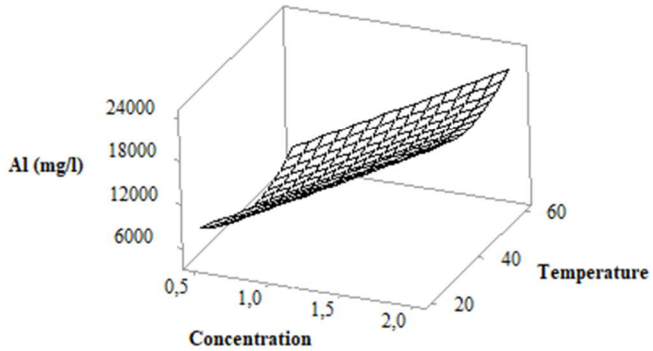
According to ANOVA analysis acid concentration is the dominant process variable, but due the significance of quadratic and interaction terms of the other two factors they must be taken into account when considering optimal conditions for leaching process. Leaching time proved to be insignificant factor which means that diffusion rates are not affected by the other factors when leaching pulverized sludge.



a)



b)



c)

Figure 2. Changes in aluminium concentration (Al mg/l) with respect to: a) concentration (M) and volume of acid (mL), b) volume of acid (mL) and temperature (ºC), c) concentration (M) and temperature (ºC) when leaching dried sludge (m= 1g)

3.4 Optimization and verification of the models

In both models no clear maximum could be found within the range of variable values. However, moving further from factor levels displayed in this study would mean increasing the costs of leaching. In addition, an increase in phosphorous acid concentration would mean higher phosphate content in leaching solution. This would lead to re-precipitation of aluminum phosphate. For that reason optimization of leaching with both sludge types was conducted within the limits of factor values. For the factors that were removed from the reduced second-order models, the lowest factorial level was chosen. Predicted values for moist sludge and dried sludge leaching were calculated according to equations (2) and (3), respectively. Table 5 presents the optimized conditions for leaching, predicted and obtained results for these settings and the percentage errors between the actual and predicted values. With both sludge types the values obtained are smaller than predicted values, resulting in error percentages of -7.9 % for moist sludge and -14.7 % for dried sludge. The analyzed aluminium content of the sludge (table 2) estimates that in two grams of moist sludge the aluminium amount is 13 mg and for dry sludge one gram contains 188 mg. If all of those amounts would be leached in optimized conditions, the concentration of aluminium should be 2.6 and 22.4 gL^{-1} with moist and dry sludge, respectively. If the obtained results and these values are compared the difference between values is -15 % for moist sludge and -4.8 % for dry sludge. Considering the heterogeneous nature of sewage sludge, it can be concluded that the regression models acquired are realistically accurate for predicting the amount of aluminium in leach solution after leaching under optimized conditions.

	Factors						
	Acid concentration (M)	Volume of acid (mL)	Temperature (°C)	Time of leaching (h)	Predicted (Al mg L^{-1})	Obtained (Al mg L^{-1})	Difference (%)
Moist[a] sludge	0.5*	5	60	6	2400	2210 ± 130	-7.9
Dry[b] sludge	2	8.4	20	2*	24650	21030 ± 800	-14.7

[a] solid content 3-4 % [b] dewatered in 105 °C for 16 h
*Not significant factor

Table 5. Optimized leaching conditions and obtained values (n=3 (mean ± std. error)) from verifications runs

4 Discussion

In optimized conditions, the liquid to solid ratio L/S (mL/g) is 2.5 and 8.4 for moist and dry sludge, respectively. This is significantly lower ratio than what Donatello et al 2010 and Fang et al., 2018 reached when optimizing ISSA leaching with sulfuric acid (L/S ratio 20) although Donatello suggests that as low as 2 could be achieved with right stirring equipment. Franz 2008 also concludes that L/S ratio 2 to be optimal, however the acid concentration in that study was high (2.5 M) while Donatello concluded that 0.19 M sulfuric acid to be sufficient. Levlin and Hultman 2004 managed to efficiently leach P from sewage sludge with just 0.25 M hydrochloric acid. Higher acid concentration is needed with H_3PO_4 due it being a weak acid. Even at concentration of 2 M of H_3PO_4 no precipitation problems were observed as opposed to Ottesen et al 2013 study with ISSA where usage of sulfuric acid lead to a formation of gypsum crystals in ash residue.

In their two-step leaching process of ISSA Levlin and Hultman 2004 managed to leach 836 mg L^{-1} aluminium with hydrochloric acid (0.25 M) while the optimized conditions in this process manages to leach 2.6 times more even with moist sludge. If we assume that equation 1 goes to full length obtained P concentration would be 2.5 g L^{-1} which is 2.2 times higher than what Levlin & Hultmann (1.134 g L^{-1}) achieved. The leaching time for

dry sludge is similar to Donatello results while Franz reported that as short as 10 minutes lead to 90 % recovery of phosphorous. Biswas et al., 2009 also investigated the effect of time and temperature on H_2SO_4 leaching of ISSA and reported 4 hour and 30 °C as optimal, which is comparable to 60 °C and 6 hours that are needed for moist sludge. High temperature in this case offers the required energy to break the chemical bonds of the metal compounds in the sludge (Naoum et al., 2001).

5 Conclusions

The optimized leaching conditions with low liquid to solid ratio (2.5) and (8.4) for moist and dried post-precipitated sludge were established, respectively. With optimized leaching conditions, phosphorus and aluminium can effectively be recovered (85% for aluminium) from post-precipitated sewage sludge. For moist sludge optimal leaching conditions are: S/L ratio 400 g L^{-1}, temperature 60 °C and leaching time of 6 hours; while for dried sludge optimal leaching conditions are: acid concentration 2 M, S/L ratio 119 g L^{-1}, temperature 20 °C and leaching time of 2 hours. The method presented has significant potential to be used in wastewater treatment for the recovery of phosphorus in industrial applications. The possibility to recycle aluminum back to the precipitation process is pioneering feature that has not yet been presented. In the future work, the separation process of aluminium from the phosphoric acid solution is developed.

6 Acknowledgements

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7 Declaration of interest

The funder Helsinki Region Environmental Services Authority has applied for a patent (Patent application no. 17162458.8-1371) based on the results presented in this paper and the corresponding author S.R. has been nominated as one of the inventors.

8 Role of the funding source

The aforementioned funders had no role in research design or in the collection, analysis and interpretation of the data or in the writing of the report. Nor did they have any role in the decision to submit the paper for publication.

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Supplementary Data

Table S1. The design layout and experimental results for CCF design

Standard order	Factors				Response variable (Y)	
	Acid concentration (mol L^{-1})	Volume of acid (mL)	Temperature (°C)	Time of leaching (h)	Concentration of aluminium (mg L^{-1})	
	(*A*)	(*B*)	(*C*)	(*D*)	moist sludge[a]	dry sludge[b]
1	2.0	5	20	2	2380	23690
2	0.5	15	20	2	990	3270
3	0.5	5	60	2	2120	9
4	2.0	15	60	2	830	11730
5	0.5	5	20	6	2240	19
6	2.0	15	20	6	840	11860
7	2.0	5	60	6	2470	17820
8	0.5	15	60	6	810	1570
9	1.25	10	40	4	1150	8460
10	1.25	10	40	4	1500	15330
11	0.5	5	20	2	2190	32
12	2.0	15	20	2	920	12150
13	2.0	5	60	2	2000	20470
14	0.5	15	60	2	940	2110
15	2.0	5	20	6	2160	23650
16	0.5	15	20	6	860	3180
17	0.5	5	60	6	2510	10
18	2.0	15	60	6	850	12240
19	1.25	10	40	4	1130	13770
20	1.25	10	40	4	1110	15602
21	0.5	10	40	4	1210	500
22	2.0	10	40	4	1160	17640
23	1.25	5	40	4	2220	1290
24	1.25	15	40	4	860	7230
25	1.25	10	20	4	1190	15700
26	1.25	10	60	4	1220	12500
27	1.25	10	40	2	1340	10220
28	1.25	10	40	6	1200	13150
29	1.25	10	40	4	1160	14040
30	1.25	10	40	4	1220	13600

[a] solid content 3-4 %
[b] dried in 105 °C for 16 h

Table S2. Method detection limits (MDLs), wavelengths, viewing, calibration ranges and R^2-values for analyzed elements

Element	MDL ($\mu g\ L^{-1}$)	Wavelength (nm)	Plasma viewing	Calibration range ($mg\ L^{-1}$)	R^2-value
Al	30	396.153	Radial	5-50	0.9999
P	265	213.617	Axial	10-100	0.9998
Fe	144	259.939	Radial	0.25-5	0.9999
Ca	288	317.933	Radial	1-20	0.9999
K	1060	766.490	Axial	0.5-10	0.9997
Mg	29	279.077	Radial	0.25-5	0.9999
Na	255	589.592	Radial	0.8-16	0.9999
S	390	180.669	Axial	0.8-16	0.9999
Si	50	251.611	Axial	0.5-10	0.9999
Cr[a]	n/d	267.716	Radial	10	1
Cu[a]	n/d	327.393	Radial	10	1
Ni[a]	n/d	231.604	Radial	10	1
Zn[a]	n/d	206.200	Radial	10	1

[a] semi-quantitative measurement (calibration: blank + 1 standard)

n/d not determined

Table S3. Determined element concentrations (mg kg^{-1}) of post-precipitated phosphorus sludge (n=3)

	moist sludge [a]	dry sludge [b]
Fe (mg kg^{-1})	340 ± 20	5660 ± 90
Ca (mg kg^{-1})	200 ± 30	6010 ± 80
K (mg kg^{-1})	20.7 ± 1,6	1700 ± 300
Mg (mg kg^{-1})	9.1 ± 0,5	780 ± 20
Na (mg kg^{-1})	56 ± 5	5400 ± 200
S (mg kg^{-1})	201 ± 30	6700 ± 110
Si (mg kg^{-1})	129 ± 11	3800 ± 70
Cr (mg kg^{-1})[c]	—	36.9
Cu (mg kg^{-1})[c]	—	66.7
Ni (mg kg^{-1})[c]	—	30.6
Zn (mg kg^{-1})[c]	—	326.4

[a] solid content 3-4 %
[b] dried in 105 °C for 16 h
[c] semi-quantitative measurement (calibration: blank + 1 standard)

Model assumption checks

The normal probability plot of residuals, the residuals versus fitted-values plot and the predicted response versus measured values for both models are presented in figures S1-S3. According to plots in fig. S1 residuals follow the straight line confirming that the residuals are normally distributed. In the residual plot of moist sludge leaching a possible outlier can be detected, but a re-run produced the same result. Figure S2 displays random scatter, which indicates that the residuals have constant variance, while figure S3 shows that predicted values in both models match the actual values.

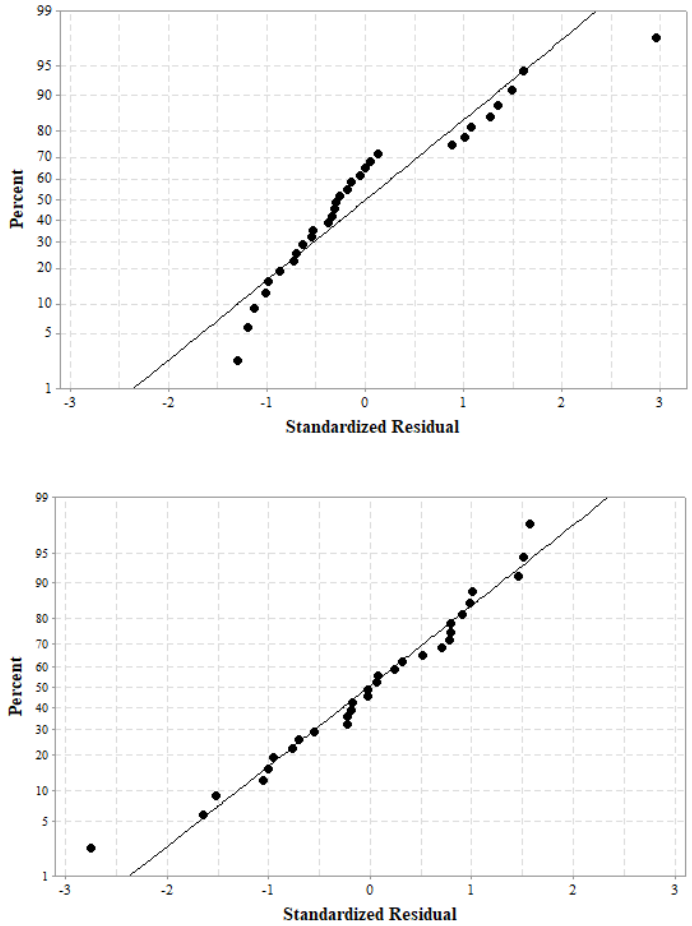

Figure S2. Normal probability plot of residuals for moist sludge (top) and dewatered sludge (bottom)

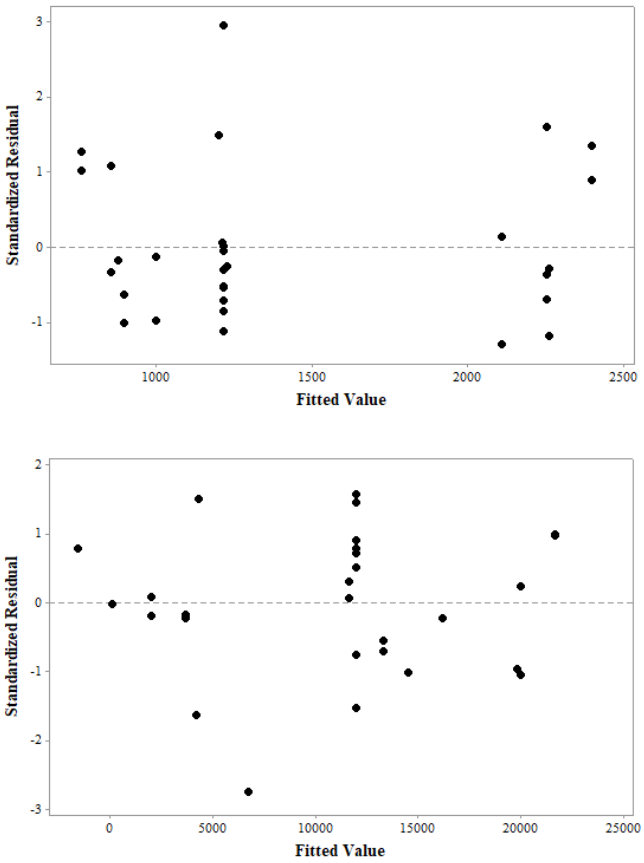

Figure S3. Plot of residuals vs fitted values for moist sludge (top) and dewatered sludge (bottom)

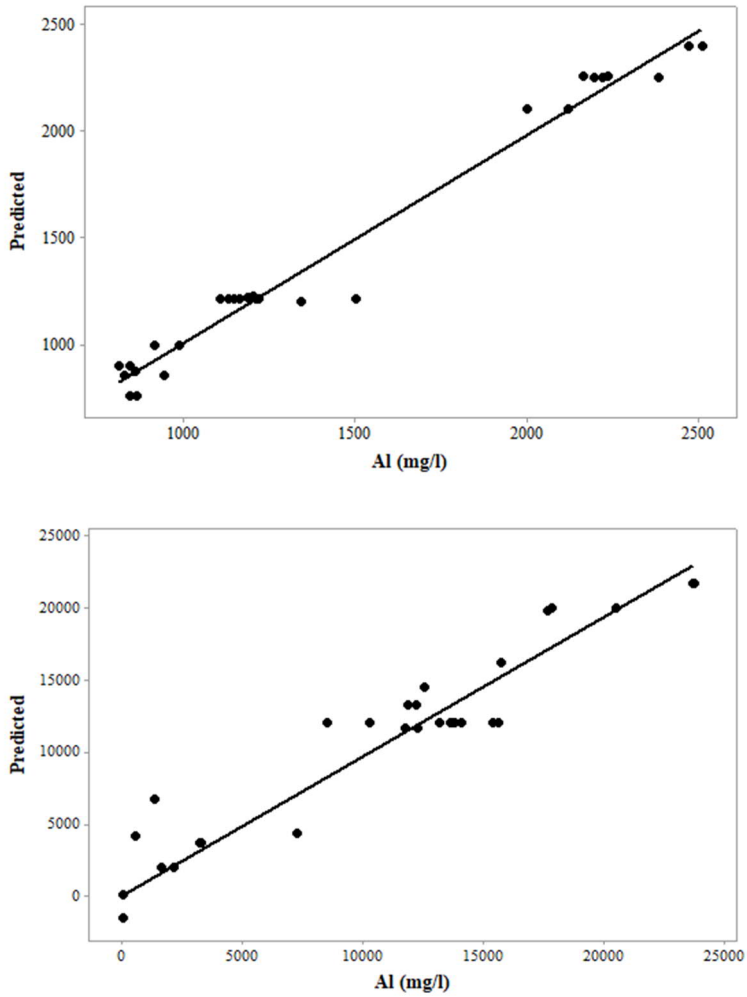

Figure S4. Plot of predicted aluminum concentration vs actual value for moist sludge (top) and dewatered sludge (bottom)

Main effect plots

Main effect plots for ANOVA analysis are presented in figures S4 and S5.

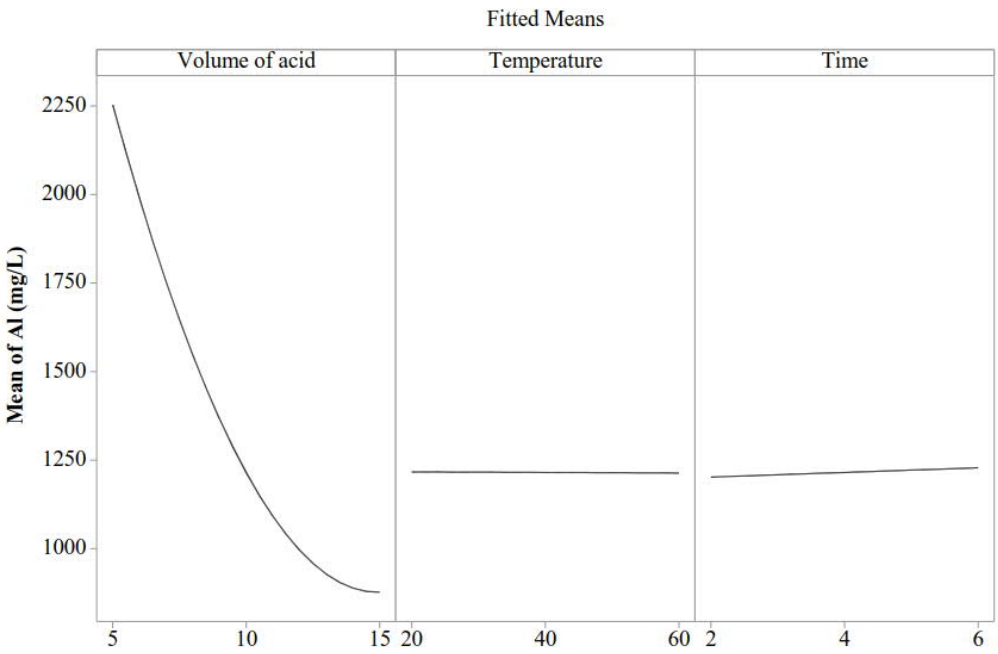

Figure S5. Main effect plot for moist sludge

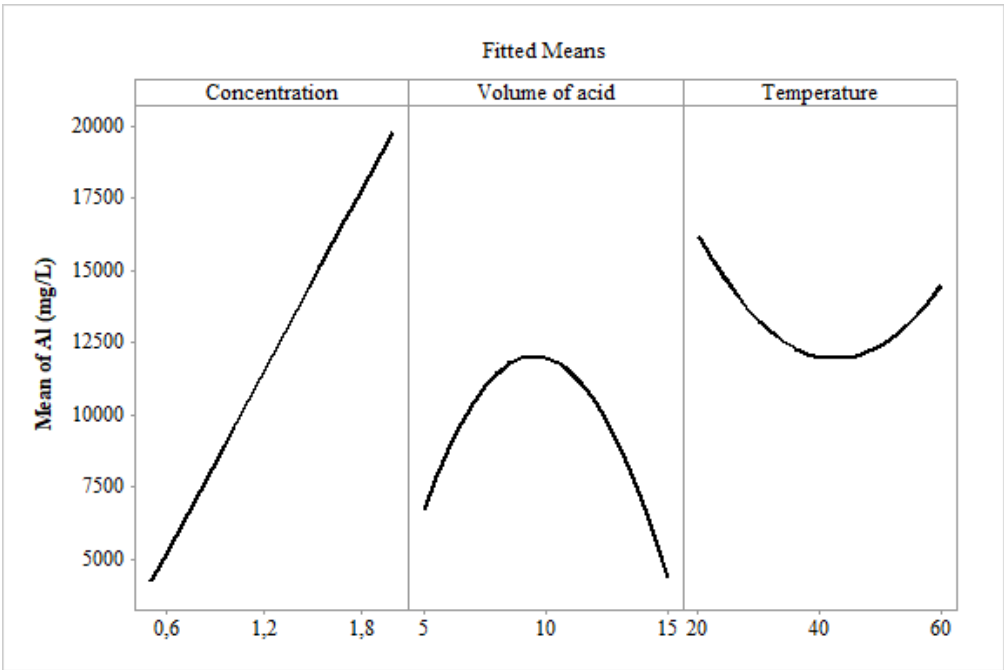

Figure S6. Main effect plot for dry sludge