[002]附属環境工学研究教育センター研究活動報告

https://doi.org/10.15017/4102258

出版情報：附属環境工学研究教育センター研究活動報告. 2, 2020-07-31. Center for Research and Education of Environmental Technology, Faculty of Engineering, Kyushu University
バージョン：
権利関係：

EFFECT OF HEAVY RAIN DUE TO CLIMATE CHANGE ON LEACHING BEHAVIOR OF LANDFILL WITH MUNICIPAL SOLID WASTE INCINERATION BOTTOM ASH

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ABSTRACT: The significant increases in bottom ash (BA) as residue of incineration methods have caused serious problems for the environment and human life. In Japan, 70% of municipal solid waste incineration (MSWI) BA is disposed into landfills. The increased frequency of heavy rain directly affects MSWI residues in landfill. To evaluate the impacts of heavy rain on leaching behaviors of total organic carbon (TOC), total nitrogen (TN), and ions from MSWI residue landfill, laboratory-scale experiments were conducted using three columns filled with BA. A sequence of experiments simultaneously simulated leaching behavior of TOC, TN, and ions under different heavy rainfall levels of 25, 50, and 100 mm/h and normal rainfall of 7.5 mm/h. Results showed an decreasing tendency of leaching behavior of targeted constituents after heavy rain and increases after normal rain. The pH value fluctuated around 11–12 after heavy rain but decreased to 7–8 after normal rain. The heavier rain caused more fluctuation in leachate concentrations. The correlation between rainfall variation and pH, TOC, TN, and Cl^- variation of leachate from BA columns was described by relationships of the form $y = -a \times \ln(x + 1)$, where x is rainfall variation and y is leachate parameter (pH, TOC, TN, and Cl^-).

1. INTRODUCTION

Incineration is the most popular method of municipal solid waste (MSW) treatment in developed countries due to advantages of reducing waste mass and volume by 70% and 90%, respectively (Ole, 1996), as well as recovering energy for electricity and heat generation (Allegrini et al., 2015). The enormous amount of bottom ash (BA), accounting for approximately 90% of the residue remaining after the combustion process (Um and Ahn, 2017)(Stegemann et al., 1995), is a serious problem for governments to cope with due to containing highly soluble salts, organic matter, and other compounds (Lam et al., 2010) (Joseph et al., 2018) (Loginova et al., 2019). Although BA is reused as alternative material for cement production (Dou et al., 2017), a large amount is disposed directly to landfill. In Japan, 70% of the total incineration ash is disposed to landfill.

After closing landfill, discharge of landfill leachate occurs during a long-term process from several months to several years or even centuries (Qiang et al., 2015). Leachate treatment processing is time consuming and costly and every landfill encounters it. Advanced constructed landfills separate rain water from the landfill site as much as possible by constructing a rainwater drainage channel or covering by

specialized materials (e.g. high-density polyethylene). Nevertheless, it is impossible to completely separate all rainwater from landfill sites. Recently, due to the rapid economic and urban development, the urgent topics of global warming and climate change are creating great concern in the research community worldwide. The greenhouse gases discharged by human daily activities have been contributing to changes in the hydrological cycle and leading to formation of heavy rain events (Min et al., 2011)(Yamamoto, 1993)(Trenberth et al., 2003). Extreme weather events are more frequent and more complex in many places around the world. According to the Japan Automated Meteorological Data Acquisition System, heavy rain of more than 50 and 80 mm/h has increased by 1.3 and 1.7 times in the last 30 years. Abnormal changes in weather, especially heavy rain, can affect the environment and public life (Linden and Office, 2007)(Pimenta, 2007)(Willems et al., 2012), and its impacts on landfill stabilization need to be clarified.

Landfill test cells or large scale lysimeters are used in landfill research; however, laboratory-scale lysimeters are mostly used in landfill simulation research with advantages such as reliable data, short duration experiments, cost savings, and simple implementation (Barlaz et al., 1989)(Qiang et al., 2015)(Aljaradin and Persson, 2016). Many researchers have investigated the effects of different factors on leaching behaviors, such as pH, liquid to solid ratio (L/S), ash characteristics, weathering and aging, contact time, and investigation scale (Quina et al., 2009)(Qiang et al., 2015)(Phoungthong et al., 2016)(Zhang et al., 2016)(Dou et al., 2017). However, little research has examined the impacts of heavy rain on leaching behavior of landfill. The infiltration of different rainfall levels into the landfill body and the leachate process remain unknown.

The objectives of this study are to:

1) Evaluate the leachate quantity under different rainfalls simulation,

2) Evaluate the impacts of heavy rain on leaching behavior of total organic carbon (TOC), total nitrogen (TN), and ions from BA packed columns,

3) Investigate the relationship between variation in rainfall and variations of pH, TOC, TN, and Cl^- of leachate from monofill containing only BA.

The results will provide information for landfill managers to predict fluctuations in leachate quality according to rainfall.

2. MATERIALS AND METHODS

2.1 Waste samples

The BA samples were collected from a continuous operating incinerator (stoker type) in Fukuoka Prefecture, Japan. First, the samples were air-dried at room temperature for 48 h prior to sieving to collect particles of less than 10 mm. The screened waste was thoroughly mixed and large particles such as metal and glass were discarded manually.

2.2 Column experiments

The schematic diagram of experimental columns is shown in Figure 1. Four Plexiglas cylinders 1000 mm in height and 150 mm in inner diameter were used. Columns were packed with fresh BA at density of 1100 kg/m^3. Column structure was organized with a 50-mm bottom layer of drainage gravel size 6–20 mm to facilitate leaching, 800-mm layer of BA, 50-mm of gravel size 2–6 mm layer in combination with paper filter to adequately distribute rainwater infiltration throughout the columns, and a final 100-mm back-up space in the case of rainwater overload. There was 1-mm metal mesh placed on the upper and lower surfaces of waste layers in order to fix the position of these layers inside columns.

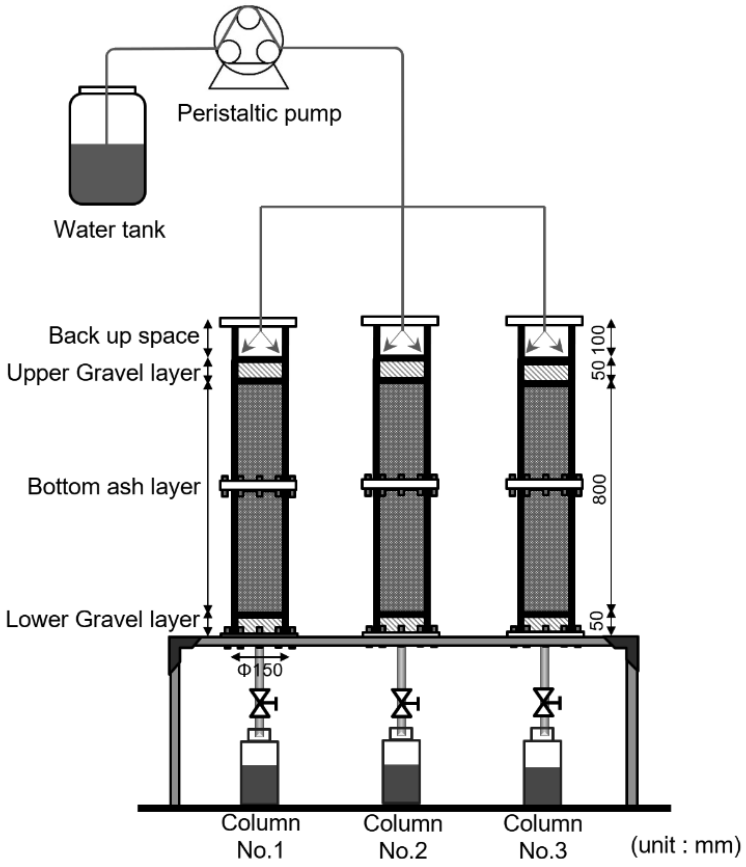

Figure 1. Schematic diagram of experimental set up.

In this study, semi-aerobic condition was created to simulate the real conditions of Japanese landfill. The pipe from which leachate samples were collected also functioned for air ventilation. The column was open at the top, and so air could infiltrate into the MSWI ash layer. Temperature inside the column was uncontrolled. To simulate different rain conditions, tap water from silicon pipes was added by peristaltic pumps on top of the columns. Prior to every experiment, flow rates of pumps were adjusted carefully to create conditions in each column as similar as possible. Column Nos.1–3 were packed with BA.

2.3 Heavy rain simulation

Table 1 and Figure 2 show details of operational conditions of the rainfall simulation experiment. The rainfall simulation had two main stages: the first was supplied normal rainfall of rain intensity 7.5 mm/h for 30 min per day, corresponding to a total of 650 ml/week within 30 days; and the second had a combination of normal and heavy rain. In the second stage, the rain intensities in columns Column Nos.1–3 were 25, 50, and 100 mm/h, respectively. For the heavy rain simulation period, columns were supplied water within 60 min. In the second stage, the heavy rain frequency was changed from one time of supplying heavy rain per week to one time per 4 weeks. The normal rain was changed from five times per week at the first stage to two times per week and then one time per week for the second stage. The leachate samples extracted from the column experiment were filtered by 0.45-μm micropore filter prior to measuring pH, electrical conductivity (EC), TOC, TN, and ions.

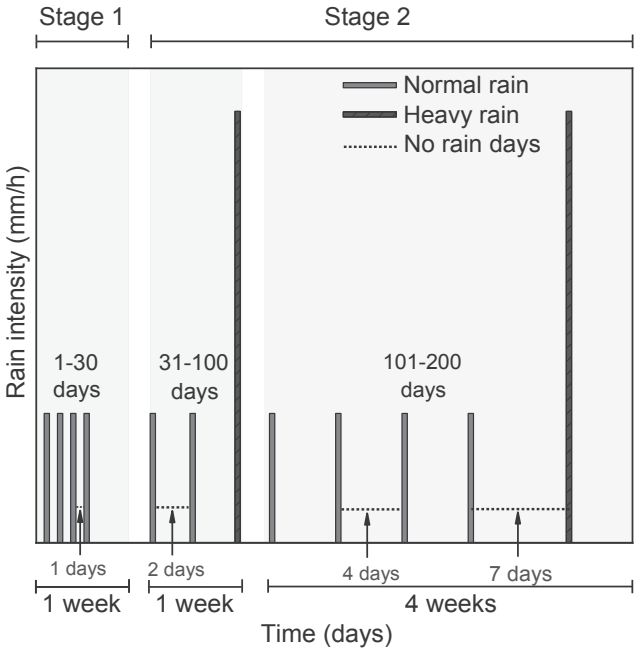

Figure 2. Details of rainfall simulation.

Table 1. Operational conditions of column experiment.

Case	Packed material	Landfill structure	Heavy rain intensity (*) (mm/h)	Normal rain intensity (**) (mm/h)
No.1	Bottom ash	Semi-aerobic	25	7.5
No.2			50	
No.3			100	

(*) At heavy rain condition, all columns was supplied water within 60 min
(**) At normal rain condition, all columns was supplied water within 30 min

2.4 Analytical methods

Before conducting the column experiment, fresh BA was analyzed for physical (moisture, dry density, and coefficient of permeability) and chemical properties (pH, EC, TOC, TN, and ion concentrations) (Table 2). Samples were agitated on a horizontal shaker during 6 h with L/S = 10:1 following by 0.45-μm micropore filtering prior to analysis. The chemical composition of MSWI BA for major elements (weight percentage) and trace elements (ppm) was determined by X-ray fluorescence (Table 3). The pH was tested using a pH meter (Model: F-53, Horiba, Japan). The EC was measured using a pH/ion/cond meter (Model: F-74BW, Horiba). The TOC and TN in leachate were analyzed using a TOC analyzer (Model: SSM-5000A, Shimadzu, Japan). Cations (Na^+, K^+, and Ca^{2+}) and anions (Cl^-, NO_3^-, and SO_4^{2-}) were analyzed using ion chromatography (Model: DX-120, Dionex, Japan).

Table 2. Fresh MSWI bottom ash characteristics.

Characteristics	Unit	Bottom ash samples
Physical characteristics		
Moisture	%	11.33
Dry density	kg/m^3	2.02
Coefficient of permeability	cm/s	0.08
Chemical characteristics		
pH		12.12
EC	mS/cm	7.38
Cl^-	mg/l	2596.51
SO_4^{2-}	mg/l	9.80
NO_3^-	mg/l	3.91
Na^+	mg/l	850.65
K^+	mg/l	288.34
Ca^{2+}	mg/l	897.44
TOC	mg/l	24.59
TN	mg/l	1.99

Table 3. Chemical composition of fresh MSWI bottom ash.

Composition	Bottom ash samples (***)
Major components (%)	
CaO	44.91
SiO_2	19.66
Al_2O_3	9.79
Fe_2O_3	5.05
Cl	3.03
MgO	2.50
P_2O_5	2.09
TiO_2	1.78
Na_2O	1.66
S	1.03
K_2O	0.58
MnO	0.13
Total (%)	*92.21*
Dominant trace components (mg/kg)	
Zn	7034
Ba	1532
Pb	1501
Cu	1203
Cr	466
Cd	114
Total (%)	*0.01*

(***) Loss on ignition of BA samples: 6.5%

3. RESULTS AND DISCUSSION

3.1 Leachate generation

Leachate generation over the 200 days of the experiment is shown in Figure 3(a). The column with heavier rainfall (No.3) created more leachate than column No.2, and column No.1 showed the smallest amount of leachate during the experiment. The total cumulative leachate amounts of column Nos.1–3 were 7542, 12 167, and 22 549 ml, respectively. Data showed that with similar amount of normal rain simulation in first 30 days, the discharge time and amount of leachate differed among the three columns. This result can be explained by the different water paths inside columns, water was retained longer in column No.3 (9 days) than Nos.1 and 2 with 5 days.

The ratio of leachate generation to rainfall increased significantly within the first 30 days (Figure 3(b)). This ratio was close to 1 after 35 days of experiment for column No.3, but not until 72 and 81 days for Nos.1 and 2, respectively. This ratio fluctuated around 1 and tended to decrease up until 200 days.

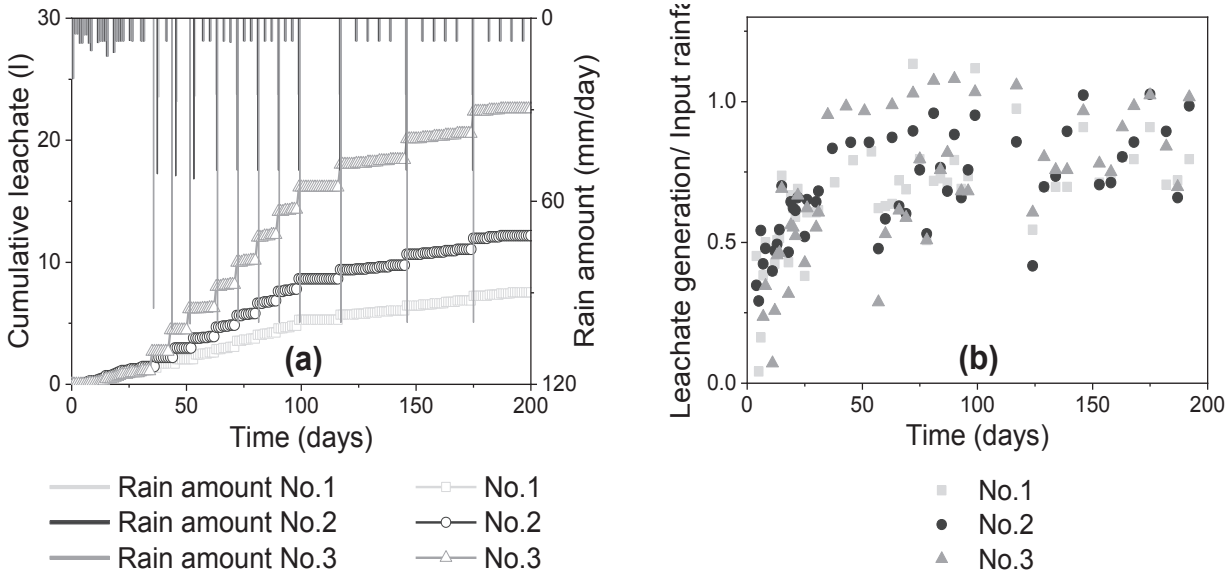

Figure 3. Leachate generation (a) and the ratio of leachate generation to input rainfall (b) for column Nos.1–3.

3.2 Impacts of heavy rain on pH and EC

During the first 30 days, pH fluctuated within 7–11 for all columns (Figure 4(a)). In the next stage, when heavy rain was supplied, the fluctuation in pH value was stronger than in the first stage. After heavy rain occurred, pH increased to approximately 12, but decreased to around 7–8 for normal rain. Obvious changes in EC value were observed during the experiment (Figure 4(b)). Maximum EC value of column No.3 was 168 mS/cm at 13 days, which was 1.7 times that of column Nos. 1 and 2 with EC around 100 mS/cm at 6 days. The EC value from No.3 leachate decreased significantly from 54 to 13 mS/cm (by approximately four times) when supplying heavy rain. A decreasing trend also occurred in column Nos. 1 and 2 with smaller magnitudes from 36 to 15 and from 24 to 13 mS/cm, respectively. All columns showed similar trends for EC change, decreasing after heavy rain and increasing after normal rain.

The high pH of leachate from experimental columns can be explained by the hydrolysis and dissolution of alkaline components, such as alkali-earth and alkali metal oxides (Dou et al., 2017)(Haberl and Schuster,

2019);

$MO + H_2O \rightarrow M_2^+ + 2OH^-$, where M = Ca

$M_2O + H_2O \rightarrow 2M^+ + 2OH^-$, where M = Na, K or others,

along with the existence of dissolved alkaline minerals such as calcite ($CaCO_3$), calcium gehlenite ($Ca_2Al[AlSiO_7]$), and calcium chloride ($CaCl_2$) (Rendek et al., 2006)(Yao et al., 2010).

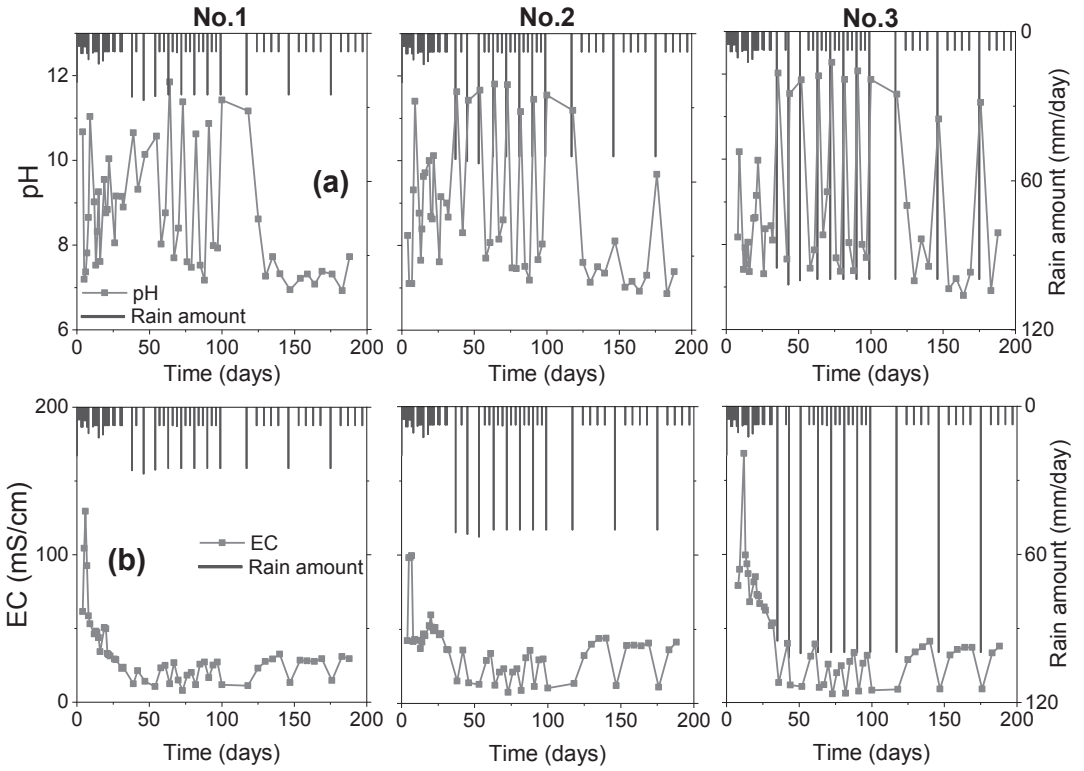

Figure 4. The pH (a) and EC (b) variation of column Nos.1–3 with heavy rain amounts.

Under the experimental conditions, pH variation was not only affected by heavy rain. The experiment was conducted under semi-aerobic condition that allowed infiltration of atmospheric CO_2 through the leachate collection pipes. Atmospheric CO_2 was absorbed into MSWI ash particles by molecular diffusion. This caused the reduction of pH value from around 11–12 to 7–8. When supplying heavy and normal rain, water particles replaced the space inside columns. Water particles reacted with CO_2 to form H^+ and HCO_3^- ions and so reduce the pH of leachate: CO_2 (aq) + H_2O (l) → HCO_3^- (aq) + H^+ (aq). When heavy rain simulation used a large amount of water, this caused the column to become waterlogged, and gas exchange between air and ash was dramatically reduced (Zevenbergen and Comans, 1994). In contrast, normal rain with less water resulted in CO_2 easily penetrating into the waste body. This phenomenon caused the difference in pH value between heavy and normal rain. Additionally, rain changed the pH due to water particles extracting alkaline compounds present on the BA surface. Heavy rain extracted large amounts of alkaline compounds and so led to high pH. The movement of atmospheric CO_2 into the BA environment and microbial respiration producing CO_2 may have slightly reduced the pH after 200 days of the experiment (Zevenbergen and Comans, 1994).

The leachate EC was measured to determine leaching behavior of salts. The change in EC was affected by dissolution of salts in BA (He et al., 2017). When rainfall increased, the EC of all columns reduced – entirely consistent with a previous study (Chen, 1996).

3.3 Impacts of heavy rain on leaching behavior of bottom ash landfill

3.3.1 TOC and TN

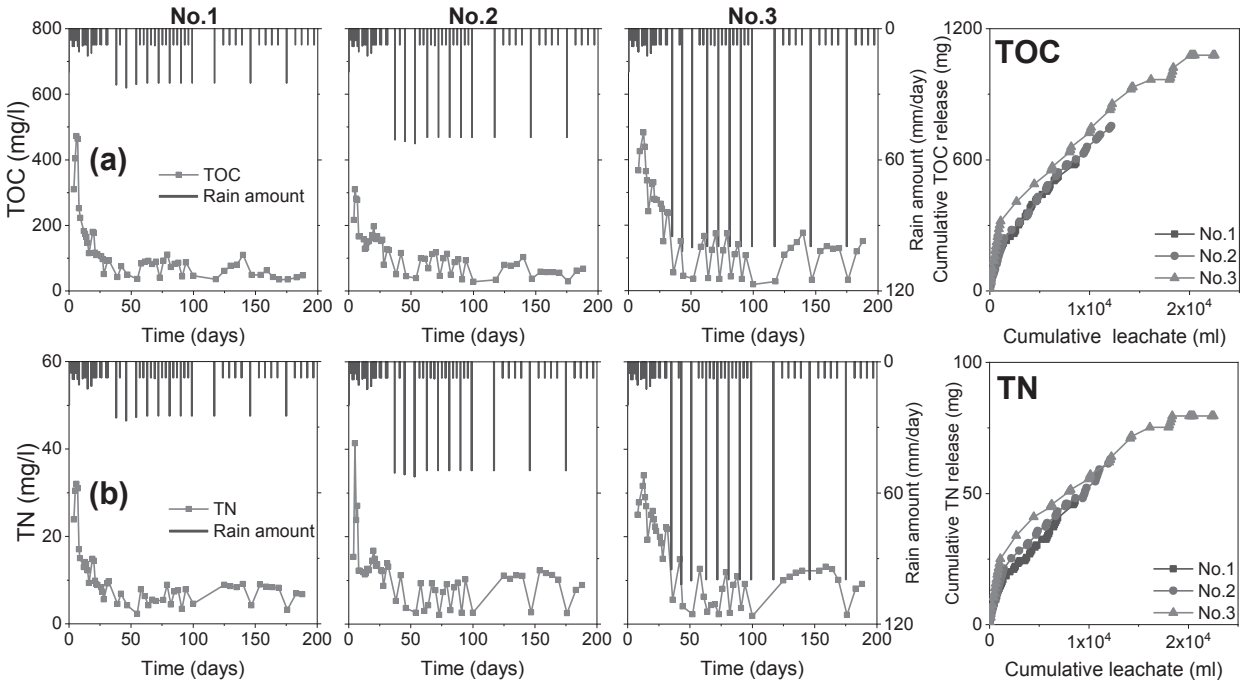

Figure 5. Concentration and cumulative release of TOC (a) and TN (b) with heavy rain for column Nos.1–3.

In general, TOC declined when supplying heavy rain and increased for normal rain (Figure 5(a)). During the first 30 days of supplying normal rain, TOC from column Nos.1–3 quickly reached maximum values of 472, 309, and 482 mg/l, respectively. The TOC fell dramatically to around 50 mg/l with the first heavy rain simulation in all three columns, with the largest reduction for column No.3, followed by No.2 and No.1.

Similarly to TOC, the TN level increased after the first week of supplying normal rain (Figure 5(b)). However, TN values varied little among columns Nos.1–3 with heavy rain simulation, with maxima of approximately 32, 41, and 34 mg/l, respectively. The TN also declined with heavy rain and again increased with normal rain.

Different dilution slopes occurred for TOC and TN among the columns, with greater slope at higher rainfall (Figure 5). Cumulative TOC and TN release were similar among columns. After 8000 ml of leachate was discharged, total TOC released from column Nos.1–3 was 550, 581, and 659 mg, respectively; and correspondingly TN released was 45, 48, and 51 mg. Water washing removed organic residues in MSWI ash (Panchangam et al., 2010). Removing organic compounds from MSWI ash was confirmed by the presence of TOC and TN (organic nitrogen) in leachate – TOC and TN are considered to indicate microbial activity. Furthermore, TOC is considered as a substrate for microbial activity. The amount of CO_2 generated by microbial respiration is usually directly dissolved in MSWI ash pore water (Rendek et al., 2006). The absorption of CO_2 due to microbial respiration, and from the surrounding atmosphere, into

MSWI ash pore water as well as alkaline compounds, plays an important role in the observed pH reductions.

3.3.2 Inorganic components

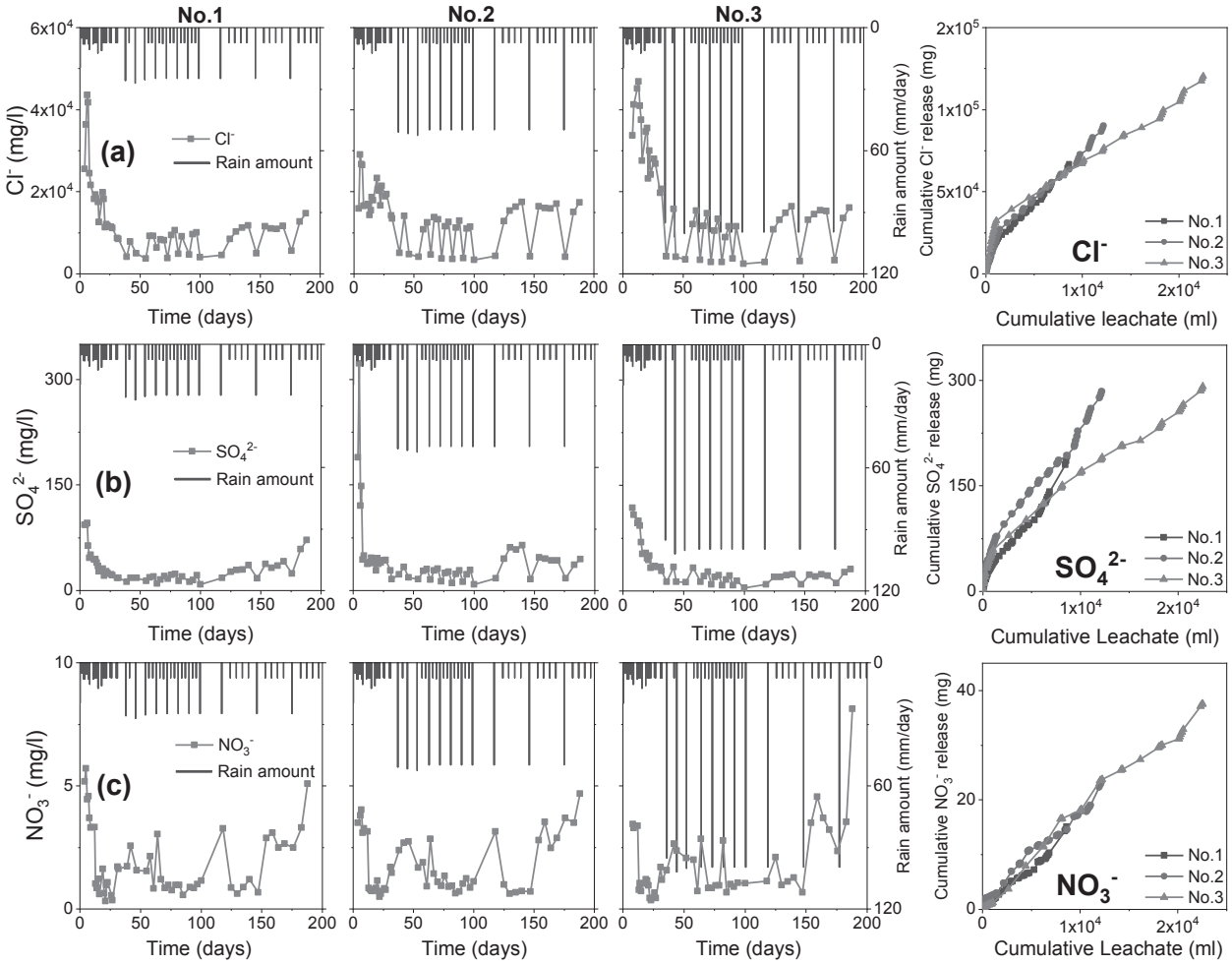

Figure 6. Concentration and cumulative release of Cl^- (a), SO_4^{2-} (b), NO_3^- (c), Na^+ (d), K^+ (e), and Ca^{2+} (f) with heavy rain for column Nos.1–3.

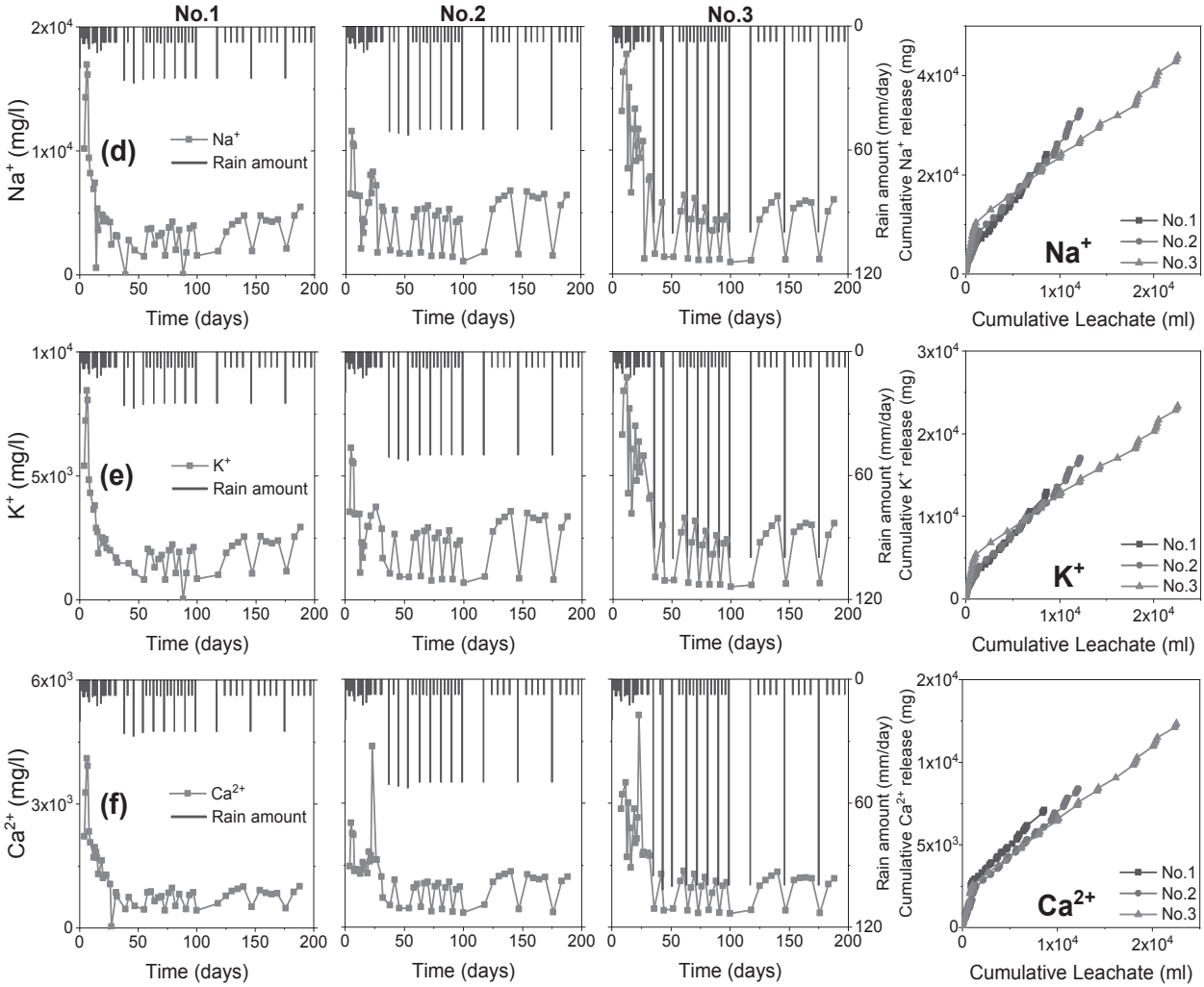

Figure 6. (continued)

The leaching behavior of ions from column Nos.1–3 was observed for almost 200 days under different rain simulations (Figure 6). Concentrations of soluble matter were very high in the first leachate fractions, with Cl^-, SO_4^{2-}, NO_3^-, Na^+, K^+, and Ca^{2+} reaching maxima before declining during the first 30 days of normal rain. The increase in leaching concentration of ions at the first time of supplying water may be caused by dissolution or desorption from surfaces of the ash matrix (Dijkstra et al., 2006)(Lokeshappa, B., Dikshit, 2012). Specifically, the surface film layer of MSWI ash particles contains a large number of readily leachable components, which in an aqueous environment can be readily washed out by water (Gunasekara et al., 2015). When reacting with MSWI ash, water particles firstly create a thin-film of hygroscopic water imbedding the waste particles. Under unsaturated waste conditions, capillary forces between waste particles affect each other. The capillary spaces between waste particles, called capillary water, are considered as the network of aeration as well as soluble and insoluble fractions in water. When supplying heavy rain, the gravitational force overcomes the interaction force between water and MSWI ash particles, and the mobile components of MSWI ash particles are removed with the movement of water particles.

Ions Cl^-, Na^+, K^+, and Ca^{2+} all showed high concentration release, consistent with previous results (Ole, 1996)(Meima and Comans, 1997). Among all columns, No.3 discharged higher total leaching levels of Cl^-,

SO_4^{2-}, Na^+, K^+, and Ca^{2+}. The major leachate components were Cl^-, Na^+, K^+, and Ca^{2+}, with maximum discharged value from column No.3 of 46 887, 17 736, 8957, and 5128 mg/l, respectively. The higher the level of rain, the lower was the concentration of ions. Considering the case of NO_3^-, the amount of NO_3^- increased after 154 days, while the rest period was fluctuated. The same trend of total ion release was apparent for Cl^-, Na^+, K^+, Ca^{2+}, and SO_4^{2-}. The three columns released similar amounts of NO_3^- after 7500 ml of leachate was discharged (Figure 6(c)). The heavy rain was supplied once per four weeks after 100 days of the experiment, and the cumulative concentration release curve of all ions (except NO_3^-) had a greater slope for column No.2 compared to the other columns.

The very high Cl^- discharged concentration showed that the experimental incineration ash samples contained major soluble chlorides, such as NaCl, KCl, and $CaCl_2$. Heavy rain with high intensity is a major factor in reducing the high value of chlorides in MSWI ash by solubility and dilution mechanisms (Chen et al., 2016). The soluble matter concentration increased after normal rain and decreased after heavy rain. This can be explained by the reaction time between water and ash particles. Large amounts of water that entered the columns during heavy rain simulation shortened the reaction time between waste and water particles, leading to reduced leachate concentration. Conversely, the lower water amount for normal rain seeped through the waste layer at lower flow rate and increased the retention time of rain water in the waste layer.

3.3.3 Variation of leachate parameters in 24-h heavy rain simulation experiment

When supplying heavy rain, leachate samples were analyzed every 10 min to determine the variation of leachate parameters for all columns. Data were collected for six times of heavy rain simulation from December 2018 until February 2019. The variation in leachate parameters in the 24-h experiment is shown in Figure 7. In the first part of this experiment, all columns had high pH values of leachate of 11–12. The lowest recorded pH was in column Nos.1 and 2 of ~12 and pH in No.3 always exceeded 12. A decreasing trend occurred for all columns after 24 h, with the greatest fall for column No.1 (from pH 12 to 9). Regarding leachate pH, heavy rain firstly dissolved and flushed the alkaline components from BA causing the high pH of leachate (Haberl and Schuster, 2019), then the pH gradually decreased. This was also affected by atmospheric CO_2 infiltrating to the inner part of column and lowering the pH in the latter period of the 24-h experiment.

The EC, TOC, TN, and ion values showed an initial decreasing trend and then increased. The sorption process also played an important role in controlling the release of elements (TOC, TN, Cl^-, SO_4^{2-}, NO_3^-, Na^+, K^+, and Ca^{2+}) that showed sorption affinity to active sites on the BA surface (Arickx et al., 2010)(Luo et al., 2019). The large amount of water from the heavy rain initially covered the MSWI ash particles, and the water particles extracted the readily leachable components on the BA surface.

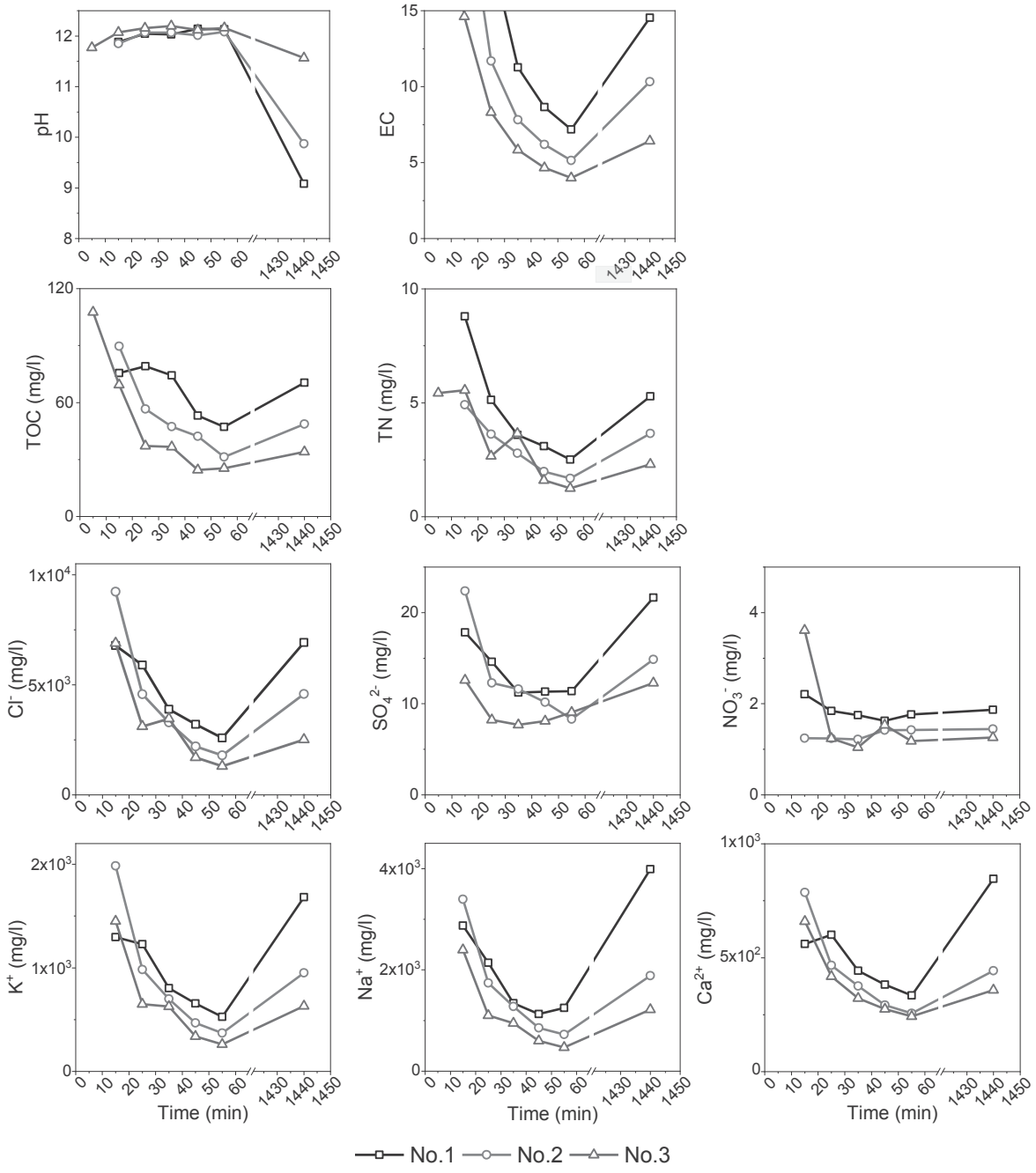

Figure 7. Leachate parameter variations for columns Nos.1–3 in the 24-h experiment: pH, EC, TOC, TN, Cl^-, SO_4^{2-}, NO_3^-, Na^+, K^+, and Ca^{2+}.

The large amount of rain water had a high gravitational force, leading to reduced retention time inside ash layers, and this explains the decreasing trend of leachate parameters (except pH) when heavy rain was initiated. When heavy rain ceased, the remaining water molecules occupied the space in the pore structure of BA materials. Then ash–water interaction forces caused the reaction that lead to the slight increase of leachate concentrations after heavy rain simulation.

3.4. Correlation between rainfall and leachate parameters

Rainfall and leachate concentration data were calculated using the following equation.

$$\Delta X = \frac{X_{n+1} - X_n}{X_n}$$

Where

ΔX is change in rainfall, pH, TOC, TN, and Cl^-.

X_{n+1} is value of rainfall, pH, TOC, TN, and Cl^- at day n + 1.

X_n is value of rainfall, pH, TOC, TN, and Cl^- at day n.

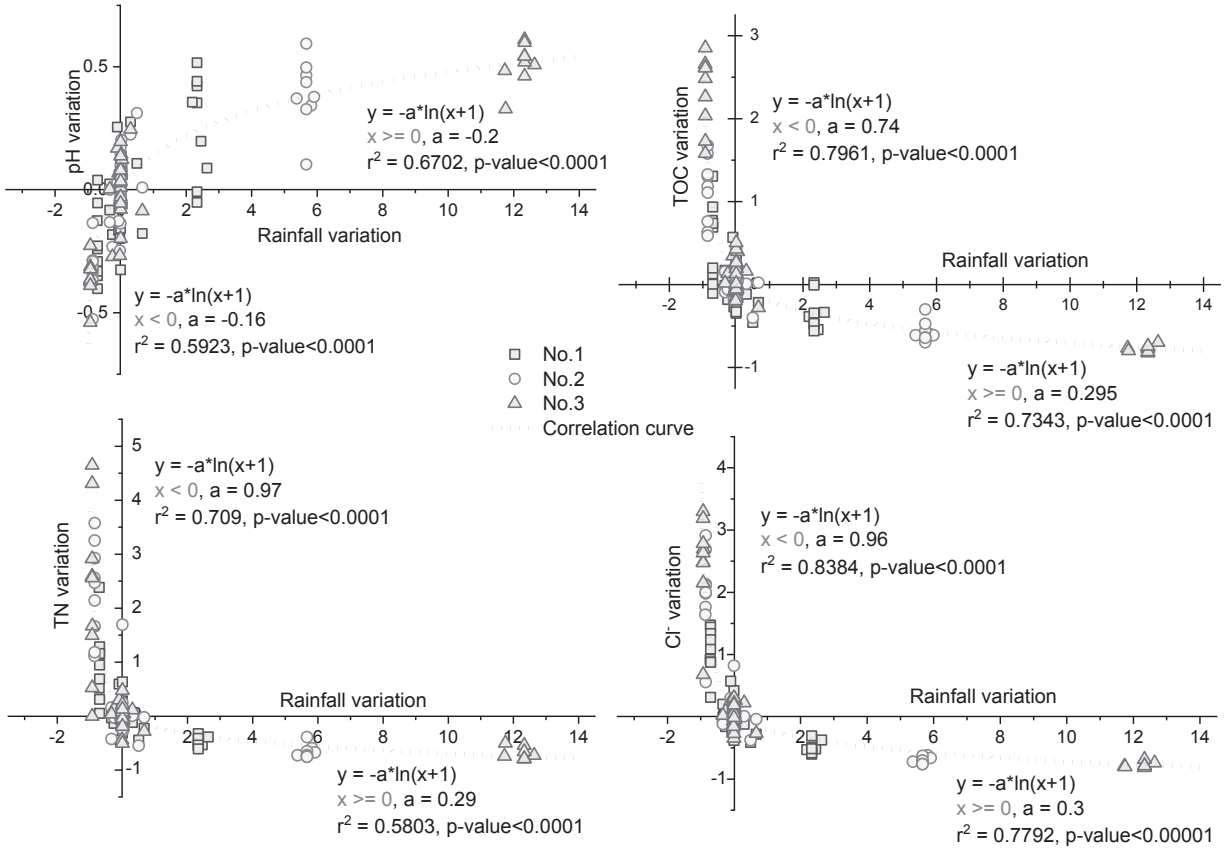

Figure 8. Relationships between rainfall and leachate parameters: pH, TOC, TN, and Cl^-.

The equation showing relationships of Cl^-, TOC, TN, and pH variation with rainfall variation is as follows.

$$y = -a \times \ln(x + 1)$$

Where

y is leachate parameter (pH, TOC, TN, and Cl^-) variation.

x is rainfall variation.

a is a coefficient with two different values corresponding to two cases of x value ($x < 0$ and $x \geq 0$).

Coefficient “a” was determined by satisfying the condition that the root-mean-square deviation had a minimum value.

Figure 8 shows the relationships between rainfall and leachate parameter variations using $y = -a \times \ln(x + 1)$. There were significant relationships between Cl^- and TOC with rainfall variation but pH and TN variation showed weaker relationships. The fitted relationships for experimental data were significant with coefficients of determination (r^2) comparatively high and p-value < 0.0001. The change of rain amount caused changes in leachate concentrations. These results may contribute to predicting leaching behavior from MSWI ash landfill sites when heavy rain occurs. This will assist landfill managers in understanding the complex changes in leachate concentration and allow better solutions to deal with these.

4. CONCLUSIONS

This research was conducted over a period of 200 days using different rainfall simulations. The results showed the impacts of heavy rain on leachate quantity and quality. Columns with heavier rainfall discharged more leachate. When heavy rain occurred, EC, TOC, TN, and ion concentrations were reduced but pH increased. In contrast, EC, TOC, TN, and ions increased after normal rain simulation, whereas pH decreased. Higher intensity and amount of rainfall showed more fluctuation in leachate concentration. In the first 30 days of supplying normal rain at 7.5 mm/h, leachate concentrations increased but declined significantly following the first heavy rain simulation. The column with heavy rain of 100 mm/h discharged leachate for 9 days later than for columns with 25 and 50 mm/h, which discharged leachate for 5 days. The flow rate of rain water and the retention time of rain water inside the waste layer affected the leaching behavior. The major mechanisms of heavy rain affecting MSWI residues in landfill were solubility and dilution. There were significant correlations between leachate parameters (Cl^- and TOC) and rainfall variation. Fitting equations of the form $y = -a \times \ln(x + 1)$ to experimental data showed significant relationships.

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