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# The influences on leachate from landfill of incineration residuals by acid precipitation

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#### **Abstract**

Incineration of municipal solid wastes (MSW) is the main method of waste management in Taiwan. Although the incineration of MSW processes the solid wastes at 850–950 °C and destroys most of the organics, the content of incineration ashes is still a problem for landfill. Moreover, acid precipitation is much worse than before in Taiwan, especially in the northern areas. For instance, the occurrence probabilities of acid precipitation measured from 1991 to 1998 in Taipei increase from 73% to 85%. Therefore, it is more important to get a series of data that will help explore the influence of acid precipitation during disposal on characterization of pollutants than to analyze the ash properties after the incinerators have been constructed and regularly used.

In this investigation, the disposal site of incineration ashes is simulated in laboratory by test columns. An irrigation experiment is taken to simulate the acid precipitation at room temperature. In order to explore the exact influence on leachate quality of the main chemical composition of acid precipitation, columns are migrated with different concentrations of sulfate in acid precipitation. This investigation showed that the sulfate concentration of acid precipitation has an increasing effect on the accumulative release of heavy metals, such as Zn, Pb and Cu, from leachate. The sulfate concentration of acid precipitation, however, will not influence the trend of chemical oxygen demand (COD), biochemical oxygen demand (BOD<sub>5</sub>) and total organic carbon (TOC) in the leachate release.

Keywords: Municipal solid wastes; Incineration ashes; Leachate; Acid precipitation

## 1. Introduction

Incineration of municipal solid waste is the principal waste management policy in areas such as Taiwan, which faces severe space constraints. People have been ignoring the influence of municipal solid waste incinerator and the problem of ash disposal. Consequently, misunderstanding the uses of the incinerator, residents in the neighborhoods where incinerators are to be built always fight against the government's policy. That is the subject we are concerned here, for municipal solid waste incinerators are being built and operated every year. In the neighboring Japan, most of municipal solid waste has been treated by way of incineration and construction of incineration ash landfills has been a common solution to the problem of incineration ash treat-

ment. But the treatment process of wastewater from incineration ash landfills has always been based on data obtained from operating landfills other than those of any given ash landfill itself. Obviously, the above approach is inadequate when it comes to assessing the environmental problems involved with the impact of incineration ashes on landfills.

The major purpose of this investigation is to explore the possible influences of chemical composition of acid precipitation in landfills on leachate quality. According to data taken from the longitudinal analysis monitor station of Environmental Protection Administration in Taiwan, and owing to the facts of industrialization and long-range transport of acidic species from offshore areas, the occurrence probability of acid precipitation is still high in Taiwan. The occurrence probabilities of acid precipitation measured from 1991 to 1998 in Taipei increase from 73% to 85% [1,2]. Since a survey of the results from researches reveals that acid precipitation could affect the composition of soil, we infer that acid precipitation could probably also affect the release of pollutants from incineration ashes. We, therefore,

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must consider the impact of acid precipitation on the incineration ash landfill leachate when an incineration ash landfill is being constructed.

Owing to different viewpoints of researches, the information on the possible influences of chemical composition of acid precipitation in landfills on leachate quality is still inadequate. What has always been on our minds is that if this investigation can obtain the related data by lysimeters test and batch experiment, it will be the first serial systemized observations on the influences of acid precipitation in landfills on pollutant leaching. The above process is aimed to obtain the leaching quantity of both the organic materials and the inorganic materials in the ashes; it is also designed to explore the characteristics of migration distribution applicable in describing the leaching potential of pollutants in landfills containing incineration residuals.

Aside from the general properties of incineration ashes exploring the relation between air pollution and the pH of rainfall and investigating the pH and composition of rainfall constantly for the weather station of Taipei (Lu et al. [3]), this investigation tries to establish a local research on the influence of pollutants from landfill of incineration ashes by acid precipitation. Although the reaction mechanism of the pollutant has not been explored, the data of acid precipitation can also be served as reference. Calvert et al. [4] points out that the reaction mechanism of sulfate and nitrate includes homogeneous reactions, the phenomena of which change from gas to liquid. Meanwhile, heterogeneous reaction occurs on particulate matter such as dust or water drops. This investigation evaluates the reaction mechanism of sulfate and nitrate from acid precipitation and tries to describe the phenomena of acid precipitation in the industrialized regions. Owing to the fact that the pH value always responds to the solubility of CO<sub>2</sub>, Seinfeld [5] solves the pH value of rain at natural temperature that derives from the solubility of CO<sub>2</sub> at saturation. This research provides a simple index to determine the occurrence probability of acid precipitation based on the pH value of rainfall. But the characteristic of composition of acid precipitation is not completed. The formation of acid precipitation is relevant to chemical and physical reactions of pollutants in the atmosphere. Chen and Qiu [6] describes the process of acid precipitation generated by pollutants, such as transport, diffusion, transformation, adsorption, deposition and removal in the atmosphere. Chen [7] notes that the composition of acid precipitation is influenced by changes of the seasons. Meanwhile, the trend of acidic and alkaline materials like  $SO_4^{2-}$ ,  $H^+$ ,  $Ca^{2+}$ , NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, Mg<sup>2+</sup> and Cl<sup>-</sup> fits the trend of changes in the seasons. Lin and Chen [8] mentions that the rainfall in spring and the frontal rainfall in winter are the most acidic. Chang and Lin [9] employs the ISCST3 (Industrial Source Complex Short-Term, 1995) model to simulate the process involved in transport, diffusion, transformation, adsorption and deposition of pollutants in Taoyuan area to assess the relation between the pollutant and the acceptor.

Researches [10–13] on the characteristics of incineration ashes have been widely disseminated, but most of the analyzes have been limited to the inorganic properties of incineration ashes. Furthermore, difficulties in controlling experimental con-

ditions hamper the accuracy of the results. Consequently, the approach adopted above is inadequate when it comes to assessing the environmental problems concerning the impact of incineration ashes on landfills.

The characteristics of landfill leachate have been widely modeled. A survey of the results from the papers listed below has inspired the development of models to describe the phenomena of landfill leachate. Examining the characteristics of leachate from a landfill containing incinerator ashes, Kenneth et al. [14] simulates the leachate quality of a 25-year-old landfill in laboratory and compares it with that of a sanitary municipal solid waste landfill. The pH value of leachate from the municipal solid waste incineration ash landfill is 10.2-10.6, higher than that from the municipal solid waste landfill. Since the leachate quality of the high alkaline ash landfill cannot be neutralized by acid precipitation, it remains highly alkaline. Moreover, the values of TOC, COD and BOD<sub>5</sub> of the municipal solid waste incineration ash landfill are lower than those of the samples from the municipal solid waste landfill. Therefore, the organic content of the leachate from municipal solid waste incineration ashes is pretty low. The leachate from the municipal solid waste incineration ash landfill contains high levels of salts, such as K<sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, and Cl<sup>-</sup>, just like what are found in the municipal solid waste landfill. Chen and Yang [15] employs ten lysimeters to simulate the leachate from the landfill. The quantity of leachate decreases with increased addition of rainfall, while it increases with greater burial depth. The pH is around 6–8 and remains uninfluenced by rainfall intensity. The chloride level of the leachate, nonetheless, increases with greater burial depth, while the COD level of the leachate decreases with lower burial depth. Chow and Gau [16] notes that bottom ash of municipal solid waste is highly alkaline. Unlike the change of chloride, the pH and COD are stable at burial depths between 120 cm and 180 cm. In other words, given the burial depth of 180 cm, incineration ashes display adsorba-

The fact that the presence of acid precipitation has become more frequent, the leaching characteristics of leachate in landfill containing incineration residuals related to acid precipitation is now available to be observed. In this sense, this investigation also serves to describe the influences of acid precipitation at a landfill site.

#### 2. Experimental methods

The test materials, that is, the bottom ashes, are sampled from the municipal solid waste incinerators. Five laboratory scale lysimeters are designed to simulate the disposal site of incineration ashes in the laboratory. Before the test materials are placed into the lysimeters, the moisture of test materials is modified with distilled water to the original value, 23 wt.%, and the top of the lysimeter is covered with a 15 cm layer of soil sampled from landfills site of municipal solid waste.

The experiment consists of lysimeters test and extraction experiment. The lysimeters test focuses on explorations of the influence of acid precipitation and burial condition. The lysimeters are irrigated with simulated acid precipitation. Given the fact that the composition of acid precipitation in Taiwan (Chang

Table 1
The chemical properties of rainfall

Station	pH value	Cl-	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>
1	4.63	84.7	46.0	111.1	70.6	35.9	20.6
2	4.62	45.7	29.0	96.9	37.2	34.2	21.7
3	4.52	107.8	34.8	129.1	89.1	39.9	38.0
4	4.61	35.2	52.5	91.7	34.4	29.1	9.9
5	4.67	265.6	33.1	109.4	244.3	40.7	56.4

Data are presented as µeq/l.

and Lin [9]) and the acidic substances from the atmosphere are roughly 70% in form of  $SO_X$  and 30% in form of  $NO_X$  [17], we, therefore, decide to study on the influence of the different strengths of sodium sulfate and sodium nitrate of acid precipitation at a landfill site.

Acid precipitation is prepared using average chemical properties of rainfall (see Table 1) and adjusted to pH value 4.6 by way of adding hydrochloric acid.

An irrigation experiment is conducted to simulate acid precipitation at room temperature, ranging from 10 °C to 22 °C, and leachate quality is then analyzed over serial, systemized observations. Items to be analyzed include organic matter like COD, BOD<sub>5</sub>, TOC, inorganic matter like Cl<sup>-</sup>, SO<sub>4</sub><sup>-</sup>, and heavy metals like Cu, Pb, Zn are analyzed. The 1st lysimeter is irrigated daily with distilled water, while the 2nd lysimeter is with acid solution 1, the 3rd with acid solution 2, the 4th with acid solution 3, and the 5th with acid solution 4. Table 2 summarizes the experimental conditions of acid precipitation. Therefore, the leaching potential of pollutants in landfills containing incineration residuals, such as the solubility of the organic as well as the inorganic matter, is thus influenced by different concentration of acid precipitation and is to be determined by the analysis of the leachate.

Since the incineration ashes still contain a few organisms, the adsorption and the desorption parameters are more influential on the leachate quality than on the biological reaction. But, in our previous study [18], acid precipitation decreases the pH value of landfill site in the beginning stage, which is suitable for the growth of microorganism and would degrade the organic matter. Thus,  $BOD_5$  becomes an important parameter in this research to reveal the effect of microorganism in landfill.

#### 2.1. Lysimeter test

All the lysimeters are held at room temperature and based on the average of annual precipitation, and a fixed quantity of

Table 2
The condition of simulative acid precipitation experiment

Lysimeter	Precipitation simulation
1 (blank)	Distilled water pH 5.6
2 (acid solution 1)	$1 \times [SO_4^{2-}] + 1 \times [NO_3^-] \text{ pH } 4.6$
3 (acid solution 2)	$2 \times [SO_4^{2-}] \text{ pH } 4.6$
4 (acid solution 3)	$2 \times [NO_3^{-}] \text{ pH } 4.6$
5 (acid solution 4)	$2 \times [SO_4^{2-}] + 2 \times [NO_3^-] \text{ pH } 4.6$

 $[SO_4^{2-}]$ : 5 mg/l;  $[NO_3^{-}]$ : 2.3 mg/l.

liquid is added to the lysimeters on the daily basis. Leachate from the lysimeters is collected daily, and heavy metal and organic components of each sample are measured. The lysimeter test consists of three main parts, and they are (1) the lysimeters, (2) the packing, and (3) daily irrigating.

#### 2.2. The lysimeters

Five laboratory scale lysimeters are set up to simulate landfills containing incineration residuals in an indoor laboratory. The diameter (D) of lysimeters is 20 cm and the height (H) is 90 cm. All of the lysimeters are made of PVC. Before the test materials are placed into the lysimeters, the test materials must be shredded into pieces of the dimensions of less than 1 cm so that they are generally homogeneous.

### 2.3. The packing

The burial density  $(\rho)$  of incineration ashes in any given actual landfill in Taiwan is designed around 1200 kg/m<sup>3</sup>. Therefore, the weight of landfill material in each column is:

$$\left(\frac{\pi}{4}\right) \times D^2 \times H \times \rho = 33.92 \,\mathrm{kg} \tag{1}$$

The thermal probe is placed in the window of each column in order to measure the landfills temperature. Meanwhile, the equipment for leachate collection is set at the bottom of each column. After the packing of the lysimeters with incineration ashes, the top of the lysimeter is covered with a 15 cm layer of soil sampled from landfills site of municipal solid waste.

#### 2.4. Daily irrigating

According to the inquisitional results of actual landfill site obtained by Chow [19], the intensity of rainfall would not cause evident change of the leachate quality but the quantity of leachate would be increased after heavy rainfall. Hence, for the fundamental research, we adopt the mean yearly precipitation (*I*), which is 2500 mm in Taiwan, to evaluate the leachability of major element from landfill of incineration residuals by general precipitation. Consequently, a fixed quantity of simulated acid precipitation is added daily to the lysimeters to simulate the landfill site. To guarantee normal irrigation and to avoid the acid precipitation accumulating in the surface and causing short flow, the simulated acid precipitation is dropped into the lysimeter slowly through the needle of a syringe in a crisscross manner in order to simulate the actual infiltration and irrigation situation.

The 1st lysimeter is irrigated daily with distilled water, the 2nd is irrigated with acid solution 1, the 3rd lysimeter with acid solution 2, the 4th with acid solution 3, and the 5th lysimeter with acid solution 4. Irrigation is conducted daily to simulate the mean precipitation, with quantity based on recorded precipitation levels. According to the actual burial situation in Taiwan, the quantity of irrigation is 0.3–0.7 times that of precipitation. The investigation adopts the top ratio of it, namely 0.7, so the

quantity of irrigation is:

$$\left(\frac{\pi}{4}\right) \times D^2 \times \left(\frac{I}{365}\right) \times 0.7 = 150.62 \text{ ml/day}$$
 (2)

An irrigation quantity of 150 ml/day is thus conducted. Given different pH values and concentration of composition of rainfall, this step is supposed to compare the solubility of the organic and the inorganic matter. The acid precipitation is added to five lysimeters from the top, and leachates are collected and analyzed thereafter.

#### 2.5. Batch extraction experiment

The toxicity characteristic leaching procedure (TCLP) is developed for the evaluation of the leaching quantity potential of both the organic and the inorganic analytes present in liquid, solid, and multiphasic wastes, as this is an effective method to obtain the quantity for leaching in a short period of time. However, extract from a bottle extractor still cannot be used to demonstrate that is the maximum concentration of heavy metals. The results of the batch extraction experiment can offer the reference leaching quantity of heavy metals.

According to the TCLP testing guidelines specified in NIEA R201.13C of the Environmental Protection Administration (EPA) of Taiwan, the extracts are filtered through glass fiber paper and analyzed for the concentration of Pb, Zn and Cu by using atomic absorption spectrometer (AAS). This procedure is designed to compare the soluble tendency of heavy metals with the results of lysimeter test.

#### 2.6. Analytical methods

# 2.6.1. Organic component analyzes of leachate chemical oxygen demand (COD)

According to the chemical oxygen demand (COD) testing guidelines specified in NIEA W516.53A of the EPA of Taiwan, the dichromate reflux is one of many a standard method used to measure the COD of the sample. The above laboratory procedure, in fact, makes reference to the United States Environmental Protection Agency (EPA) methods for analysis of waters and wastes, method 410.3 (1983).

The 50 ml leachate, or an aliquot diluted to 50 ml, is refluxed in strongly acid solution with a known excess of potassium dichromate ( $K_2Cr_2O_7$ ). The blank is prepared by using 50 ml distilled water. After digestion, the remaining unreduced  $K_2Cr_2O_7$  is titrated with ferrous ammonium sulfate to determine the amount of  $K_2Cr_2O_7$  consumed and the oxidizable matter is calculated in terms of oxygen equivalent.

2.6.1.1. Biochemical oxygen demand 5 days (BOD<sub>5</sub>). According to the biochemical oxygen demand (BOD<sub>5</sub>) testing guidelines specified in NIEA W510.54B of the EPA of Taiwan, the Winkler titration is one of many a standard method used to measure the BOD<sub>5</sub> of the sample. The laboratory procedure also makes reference to the United States Environmental Protection Agency (EPA) methods for analysis of waters and wastes, method 405.1 (1974).

The leachate sample is then introduced to an environment suitable for bacterial growth at reproducible temperatures, nutrient sources and light within a 20 °C incubator so that oxygen will be consumed. Quality controls, standards and dilutions are also run to test for accuracy and precision. Determination of the dissolved oxygen within the sample is done through Winkler titration methods. The difference in initial DO readings (prior to incubation) and final DO readings (after 5 days of incubation) predicts the BOD of the sample.

2.6.1.2. Total organic carbon (TOC). Total organic carbon (TOC) provides a speedy and convenient way of determining the degree of organic contamination. A carbon analyzer using an infrared detection system is used to measure total organic carbon. Organic carbon is oxidized into carbon dioxide.

According to the total organic carbon testing guidelines specified in NIEA W530.51C of the EPA of Taiwan, the Combustion-Infrared spectrometer method is one of many a standard means adopted to measure the TOC of the sample. The leachate sample is measured for the carbon dioxide by using an OIA-1020 total organic carbon analyzer. However, TOC provides a more direct expression of the organic chemical content of leachate than BOD $_5$  and COD.

#### 2.6.2. Heavy metal analyzes on the leachate

According to the heavy metal testing guidelines specified in NIEA R305.20T, NIEA R306.20T, and NIEA R307.20T of the EPA of Taiwan, leachate samples from every lysimeter are filtered through glass fiber paper and analyzed for the concentration of Pb, Zn and Cu by using atomic absorption spectrometer. A Hitachi Z-5000 AAS is used to obtain the quantity of Pb, Zn and Cu of the leachate.

#### 3. Results and discussion

It was when the releasing of the compositions of ashes apparent had declined to a stable stage we decided to present this article. At that time, the concentration not only had become stabilized but had also become much lower than it was in the beginning stage of the burial. This is supposed to compare with the batch extract experiment in order to have a better understanding of the serial changes of the pollutants in the landfills. Compared with the concentration of the main chemical composition of acid precipitation caused by the development of industry, nitrate is much lower than sulfate. Thus, the following discussions are devoted to investigating the exact influence on leachate quality of sulfate of acid precipitation.

# 3.1. Influence of $[SO_4^{2-}]$ on the solubility of organic matter

# 3.1.1. Influence of $[SO_4^{2-}]$ on the solubility of COD

Following irrigation at the same pH values and different concentrations of main chemical composition in acid precipitation such as sulfate and nitrate, Fig. 1 displays changes of COD in all lysimeters. The figure reveals that the maximum value of COD in the leachate is at around 8000 mg/l and the variety is unstable after the landfill has been created, and then declines gradually.

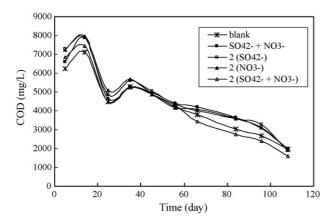


Fig. 1. COD analysis results of leachate from lysimeters.

The tendencies of COD release of the leachate from all lysimeters are similar. Comparing the concentration of the irrigated  $[SO_4{}^{2-}]$  in all lysimeters with COD in leachate from all lysimeters, it shows the acid precipitation does not increase the release of COD after the creation of the landfill. A comparison of the changes of  $[SO_4{}^{2-}]$  in Fig. 2 reveals that the trend of  $[SO_4{}^{2-}]$  does not correspond to the trend of COD release. Based on the research results of Kida et al. [20], the content of  $[SO_4{}^{2-}]$  is more than 1800 mg/kg in bottom ashes. Owing to the releasing of sulfide in incineration ashes,  $[SO_4{}^{2-}]$  of the leachate declines from 70 mg/l to 5 mg/l 10–108 days after the lysimeter has been created but the concentration of leachate remains higher than the concentrations in the simulative acid precipitation. Therefore, the concentration of sulfate in leachate of lysimeter1 and lysimeter 4 are lower than the other lysimeters.

Fig. 3 presents the amount of accumulative releasing of COD in leachate from all lysimeters. Since the ignition loss is defined as the organics index of the residuals, the total quantity of organics could be calculated through the mass balance of ignition loss. The exact value of ignition loss is 2.9% in our test materials. It is estimated that the amount of net accumulative release of COD by ignition loss, rating from 3% to 6%, could probably be shown in total accumulative release of COD. Table 3 indicates only a part of the easily degradable organism in ashes is released in the short period of time. It is evident that acid precipitation does not increase the release of COD after the creation of the landfill.

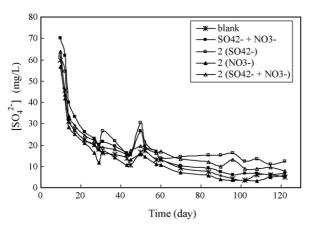


Fig. 2. [SO<sub>4</sub><sup>2-</sup>] analysis results of leachate from lysimeters.

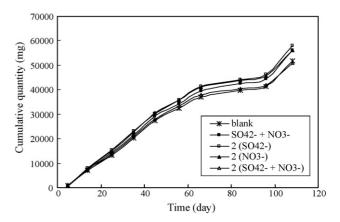


Fig. 3. Amount of accumulative releasing quantity of COD.

Table 3
The cumulative releasing quantity of COD

Number of lysimeter	Quantity of cumulative releasing (after 122 days) (mg)	Cumulative release rate (%) <sup>a</sup>	
1 (blank)	51,600	5.2	
2 (acid solution 1)	55,700	5.7	
3 (acid solution 2)	57,500	5.8	
4 (acid solution 3)	56,200	5.7	
5 (acid solution 4)	50,800	5.2	

<sup>&</sup>lt;sup>a</sup> Cumulative release rate = cumulative releasing quantity of COD/theoretic quantity of organic matter.

From the fact that the level of  $[SO_4^{2-}]$  contained in this ashes is 4 wt.% we can infer that the concentration of  $SO_4^{2-}$  in leachate releases slowly after the landfill has been created. Fig. 4 shows that the amount of accumulative releasing of  $[SO_4^{2-}]$  is only 0.01% of the ashes 108 days after the lysimeter has been created.

### 3.1.2. Influence of $[SO_4^{2-}]$ on the solubility of $BOD_5$

Our previous researches [18,19] indicate that bottom ashes still contain high level of organic matter. In fact, BOD<sub>5</sub> of leachate is affected by biological, physical, and chemical reactions after the landfill has been created. However, the biological reactions are neglected when organic matter has been almost

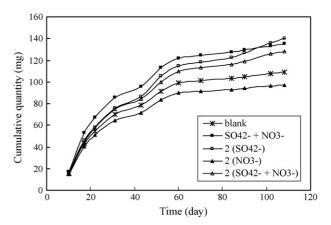


Fig. 4. Amount of accumulative releasing quantity of  $[SO_4^{2-}]$ .

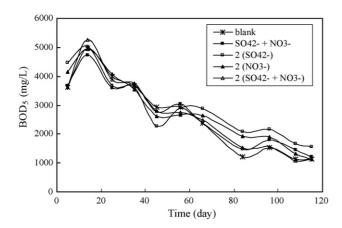


Fig. 5. BOD<sub>5</sub> analysis results of leachate from lysimeters.

all released from bottom ashes in the last stage of the landfill, and only the physical and chemical reaction will be taken into consideration. According to the ignition loss of the incineration ashes, we presume that most organic matter has been dissolved and little has been decomposition. Therefore, analysis of  $BOD_5$  in leachate is conducted to understand the release state of organic matter.

Fig. 5 refers to the varied values of BOD<sub>5</sub> in all lysimeters and reveals that the tendency of BOD<sub>5</sub> release of the leachate from all lysimeters remains similar even after the creation of the landfill.

The figure reveals that the peak  $BOD_5$  of the leachate is at around  $5200 \, \text{mg/l}$  14 days after the lysimeter has been created. The tendency of  $BOD_5$  release of the leachate from all lysimeters is similar after the creation of the landfill. Due to the same opinion about COD, therefore, the major reason that causes  $BOD_5$  release is irrigation. A comparison of the changes of  $[SO_4{}^{2-}]$  in Fig. 2 shows that the trend of  $[SO_4{}^{2-}]$  does not correspond to the trend of  $BOD_5$ .

# 3.1.3. Influence of $[SO_4^{2-}]$ on the solubility of TOC

The varied values of TOC in the leachate from all the lysimeters were shown in Fig. 6. In Fig. 6, we find out the peak TOC of the leachate is at around 3300 mg/l and the variety is also unstable after the creation of the landfill. It then declines from

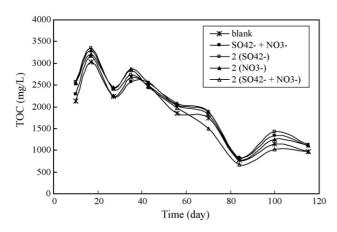


Fig. 6. TOC analysis results of leachate from lysimeters.

Table 4
The heavy metals concentration in incineration bottom ashes

Elements	Ashes
Cu	5185
Cu Pb	3249
Zn	3223

Data are presented as mg/kg.

2600 mg/l to 880 mg/l 35–122 days after the lysimeter has been created. Fig. 6 displays that the TOC release of the leachate from all lysimeters is similar after the creation of the landfill.

Comparing the varied values of TOC, COD and BOD<sub>5</sub> from all the lysimeters, we find that the releasing tendency is similar. The TOC/COD ratios, ranging from 0.25 to 0.5, show that dissoluble matter in the organic matter of the leachate has already dissolved on a great scale. Results of leaching tests have indicated that simulated acid precipitation does not increase the release of organic matter, from that we infer that acid precipitation will not increase the complex fate of heavy metals and organic matter of the leachate.

A comparison of the changes of  $[SO_4^{2-}]$  in Fig. 2 reveals that the trend of  $[SO_4^{2-}]$  also does not correspond to the trend of TOC release.

# 3.2. Influence of $[SO_4^{2-}]$ on the solubility of inorganic matter

Table 4 provides the concentration of heavy metals in the incineration ashes by acid digestion (NIEA R353.00C). Table 5 shows the inorganic matter concentration of incineration ashes extracted by extraction fluid 2 (pH  $2.88 \pm 0.05$ ) in TCLP test. The concentrations of Cu and Pb in the leachate are lower than the regulatory standards for effluent in Taiwan, which are 15 mg/l and 5 mg/l. Moreover, the varieties of Pb, Zn and Cu concentrations of leachate in all lysimeters are shown in Figs. 7–12.

Pb: Fig. 7 shows that the concentration of Pb release from lysimeters irrigated with distilled water is 9–17 mg/l, while that in lysimeters irrigated with solution of simulated acid precipitation 1 is 8–21 mg/l; 15–22 mg/l in those with solution of simulated acid precipitation 2; 15–22 mg/l in those with

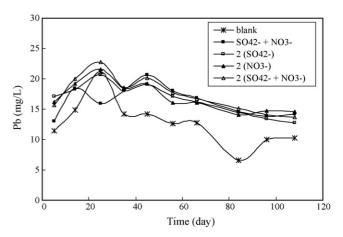


Fig. 7. Pb analysis results of leachate from lysimeters.

Table 5
Inorganic matter concentration of ashes in TCLP tests

Elements	Cu		Pb Zn		Zn	n		pH of the extract		
	mg/l	mg/kg	Soluble rate (%)	mg/l	mg/kg	Soluble rate (%)	mg/l	mg/kg	Soluble rate (%)	
Extraction										
Distilled water	1.6	32	0.62	21.7	434	13.36	1.89	37.8	1.17	12.26
TCLP solution	2.8	56	1.08	1.1	22	0.68	0.76	15.2	0.47	7.60

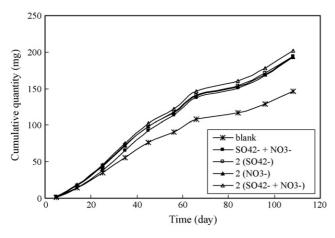


Fig. 8. Amount of accumulative releasing quantity of Pb.

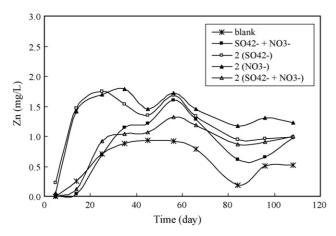


Fig. 9. Zn analysis results of leachate from lysimeters.

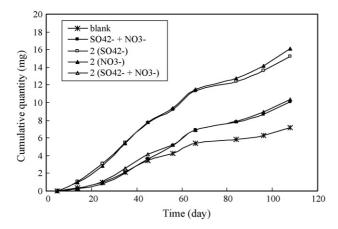


Fig. 10. Amount of accumulative releasing quantity of Zn.

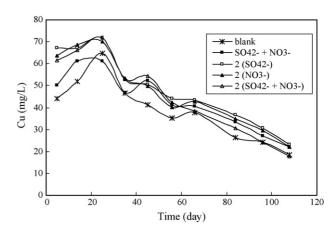


Fig. 11. Cu analysis results of leachate from lysimeters.

solution of simulated acid precipitation 3; 13–23 mg/l in those with solution of simulated acid precipitation 4, respectively.

The tendencies of Pb released from the leachate collected from all lysimeters go slowly soon after the landfill has been created. From the comparison of the tendencies of Pb released from all lysimeters, it is important to note that the presence of the difference to the lysimeter irrigated with distilled water and other acid solution 30 days after the landfill has been created. The results of the lysimeters test show that simulated acid precipitation has an increasing effect on the Pb released.

Not only the concentration of Pb shows the effected phenomenon, but the accumulative releasing of Pb also does. Accumulative releasing of Pb from lysimeter 1 to lysimeter 5 in Fig. 8 reveals that the quantities are 162.7 mg, 215.4 mg, 214.0 mg, 215.2 mg and 224.2 mg. The accumulative releasing of Pb in

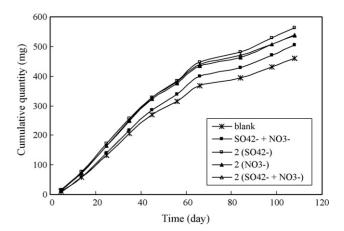


Fig. 12. Amount of accumulative releasing quantity of Cu.

Table 6
Amount of cumulative releasing quantity and soluble rate of Pb

Number of lysimeter	Release rate (%) (compare with lysimeter 1)	Quantity of cumulative releasing after 108 days (mg)	Soluble rate (%) (compare with Table 5)
1	100	162.7	1.11
2	132	215.4	1.46
3	132	214.0	1.45
4	132	215.2	1.46
5	138	224.2	1.52

the lysimeters irrigated with acid solution exceeds that of the lysimeters irrigated with distilled water for more than 30%, thus acid precipitation has an increasing effect on the release of Pb from incineration ashes. Table 6 indicates the quantity of accumulative releasing and the soluble rate of Pb. The results show that the impacts of sulfate and nitrate are nearly the same. The soluble rate of Pb for all lysimeters is under 2%. Comparing the soluble rate of Cu and Zn in all lysimeters, we find that the soluble rate of Pb is lower than Cu and Zn. It reveals that the release of Pb is quite slow and the potentiality of the release of Pb is still high over a prolonged period of time under the acidic conditions.

Zn: Fig. 9 indicates that the concentration of Zn release from lysimeters irrigated with distilled water is 0-0.93 mg/l, while that in lysimeters irrigated with solution of simulated acid precipitation 1 is 0–1.22 mg/l; 0.23–1.72 mg/l in those with solution of simulated acid precipitation 2; 0.06–1.88 mg/l in those with solution of simulated acid precipitation 3; 0–1.21 mg/l in those with solution of simulated acid precipitation 4, respectively. Compared with the concentration of Cu and Pb release in lysimeters, the concentration of Zn is the lowest. Fig. 10 reveals the accumulative releasing of Zn from leachate of all lysimeters. From the results we find that the quantity of accumulative releasing from lysimeter 1 is the lowest, the quantity of accumulative releasing from lysimeter 3 and lysimeter 4 is similar, while the quantity of accumulative releasing from lysimeter 2 and lysimeter 5 is similar too. Therefore, we infer that individual sulfate and nitrate have a similar effect on the release of Zn. When the primary anions of rainfall such as sulfate and nitrate exist alone, the effect on Zn release is more than when they are mixed.

Fig. 10 and Table 7 reveal that the quantities of accumulative releasing of Zn from lysimeter 3 and lysimeter 4 are similar, the quantities of accumulative releasing of Zn from lysimeter 2 and lysimeter 5 are also similar, but the quantity of accumulative releasing of Zn from lysimeter 1 (distilled water) is the lowest. Therefore, we may infer that not only  $[SO_4^{2-}]$  has an increasing

effect on Zn released, but also does  $[NO_3^-]$ . Since the soluble rate of Zn for all lysimeters is under 2%, it reveals that the release of Zn is very slow and the potentiality of the release of Zn is still high over a prolonged period of time under the acidic conditions.

Cu: Fig. 11 illustrates the concentration of Cu release from lysimeters irrigated with distilled water is 0–65 mg/l, while that in lysimeters irrigated with solution of simulated acid precipitation 1 is 0–62 mg/l; 0–72 mg/l in those with solution of simulated acid precipitation 2; 0-70 mg/l in those with solution of simulated acid precipitation 3; 0–72 mg/l in those with solution of simulated acid precipitation 4. The tendencies of the decline of the concentration of Cu in lysimeters go rapidly soon after the landfill has been created. Fig. 12 reveals the accumulative releasing of Cu from leachate from all lysimeters. From the results, we find that the quantity of accumulative releasing from lysimeter 1 is the lowest and the quantity of accumulative releasing from lysimeter 3 is the highest. The difference between the quantities of accumulative releasing from lysimeter 1 and lysimeter 3 is 110.1 mg, more than 20% apart. Therefore, we infer that the simulated acid precipitation has some increasing effect on the release of Cu.

Table 8 indicates the quantity of accumulative releasing and the soluble rate of Cu. The results show that the simulated acid precipitation has some increasing effect on the release of Cu. Meanwhile, the tendency of the decline of the concentration of Cu in lysimeters is rapid soon after the landfill has been created; and from this fact we infer that Cu will be released rapidly from lysimeters too.

Camparing the soluble rate of Pb, Zn and Cu from all lysimeters in Tables 6–8, we find out that the soluble rate of Cu is higher than Pb and Zn. Acid precipitation will increase the leaching quantity of Pb, Zn and Cu from 10% to several times as much. Meanwhile, since the soluble rate of those heavy metals is lower than the potential they have, the concentration of heavy metals from leachate is still a problem for wastewater treatment. Due to the analysis of leachate quality, the concentration of Cu and

Table 7
Amount of cumulative releasing quantity and soluble rate of Zn

Number of lysimeter	Release rate (%) (compare with lysimeter 1)	Quantity of cumulative releasing after 108 days (mg)	Soluble rate (%) (compare with Table 5)  0.61	
1	100	7.87		
2	145	11.38	0.89	
3	210	16.50	1.29	
4	225	17.71	1.38	
5	148	11.68	0.91	

Table 8

Amount of cumulative releasing quantity and soluble rate of Cu

Number of lysimeter	Release rate (%) (compare with lysimeter 1)	Quantity of cumulative releasing after 108 days (mg)	Soluble rate (%) (compare with Table 5)
1	100	490.9	45
2	110	539.6	50
3	122	601.0	55
4	117	575.9	53
5	116	568.3	52

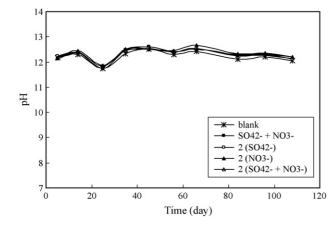


Fig. 13. pH of leachate.

Pb will not meet the regulatory standards, 3 mg/l and 1 mg/l, for effluent in Taiwan. And according to the estimation by mass balance with results of TCLP testing, the accumulative quantity released after the landfill has been created will be almost equal to the total release of Cu from TCLP testing in more than 10 years, while that of Pb is 2.4–4.4% and Zn is 1.4–3.2%. It is evident that the landfill site of incineration ashes must face up to the treatment problem of the leachate.

Because of the broad neutralization capacity of incineration ashes, the effect of acid precipitation (pH 4.6) upon pH of all lysimeters is slight (see Fig. 13). Thus, the results of the measure of Pb, Zn and Cu release under influences of different irrigation liquids reveal that lysimeters irrigated with solution of simulated acid precipitation release higher quantity of the above said heavy metals than those irrigated with distilled water. The concentration of sulfate and nitrate contained in irrigation acid solution reveals an evident influence on lysimeters irrigated with acid rather than those irrigated with distilled water. From the measured results of the concentration of heavy metals in the leacheate released from lysimeters, we infer that the concentration of the primary anions of acid precipitation, such as sulfate and nitrate, has an increasing effect on the release of heavy metals.

# 4. Conclusions

Observing all the lysimeters irrigated at the same pH values and different concentrations of main chemical composition in acid precipitation, such as sulfate and nitrate, COD of the leachate is at around 8000 mg/l after the landfill has been created,

whereas  $BOD_5$  is  $5200 \, \text{mg/l}$  and TOC is  $3300 \, \text{mg/l}$ . We have demonstrated that only a part of the easily degradable organism in ashes is released in a short time span. It is evident that acid precipitation does not increase the release of organic matter such as COD,  $BOD_5$  and TOC. From the results of the concentration of heavy metals in the leacheate released from lysimeters we infer that acid precipitation has an increasing effect on heavy metals released after the landfill has been created.

Furthermore, the leaching concentration of those heavy metals is all related to the final pH of the extracting reagent. The pH values of leachate remain stable while irrigated with acid precipitation (pH 4.6). Since the soluble rate of those heavy metals is lower than the potential they have, the concentration of heavy metals from leachate is still a problem for wastewater treatment.

The concentration of Cu and Pb will not meet the regulated standards for effluent in Taiwan. It is evident that landfill of incineration ashes must face up to the treatment problem of the leachate due to the influences of acid precipitation on it.

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