

Sediment Oxygen Demand of a Leachate Pond at an Offshore Municipal Solid Waste Disposal Site 2 Years after the Site Was Closed

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Abstract

Various water quality parameters of a leachate pond at an offshore municipal solid waste disposal site were monitored. The pH, dissolved oxygen (DO), and water temperature at the bottom of the leachate pond were measured during Sep. (the 1st period) and Nov.-Dec. (the 2nd period) of 2011. The results suggested that the stratification of water temperature in the pond had gradually broken down due to convection occurring between the end of the 1st period and the 2nd period. The pH was almost constant at 10 - 11 during the 1st period and was approximately 11.5 during the 2nd period. The DO was almost zero during both periods. An anaerobic batch experiment with sampled sediment was undertaken to elucidate the mechanism of material leaching from the sediment. DO decreased under all experimental conditions. With respect to oxidation reduction potential (ORP) and total sulfide in addition to DO, the condition most closely mimicking that of the site became the most anaerobic. The average sediment oxygen demand, SOD_{ave} , was calculated using a brief numerical model based on batch experiment data. The SOD_{ave} was 1114.7 mg/m²/d, indicating that at least 434 g/d of oxygen must be supplied to the leachate pond to maintain the DO.

Keywords

Offshore Disposal Site, Leachate Pond, Sediment, Sediment Oxygen Demand

1. Introduction

In Japan, approximately 80% of municipal solid waste (MSW) is incinerated, both to reduce its volume and for sanitary treatment [1]. Particularly in urban areas, finding new sites for final disposal of wastes has become increasingly dif-

difficult. It is thus necessary to develop strategies to manage MSW disposal sites in a more environmentally safe manner and to promote the reduction, reuse, and recycling of wastes at both the technological and legal levels.

Leachate discharged from a MSW landfill must be treated appropriately in accordance with its quality and volume [2]. Urban residents living near disposal sites may experience some unpleasantness due to odors associated with the landfill as well as potential health hazards due to gases emitted from the landfill site [3]. In addition, the color of the leachate can be worrying to residents [2]. Complaints regarding leachate color and/or hydrogen sulfide odors can be lessened by careful management of leachate pond water quality. It is therefore necessary to consider measures that will prevent deterioration of pond water quality.

For the purpose of generating data that would aid in development of appropriate methods for controlling the water quality in leachate ponds, the authors investigated a leachate pond at an offshore disposal site 2 years after the site was closed. Pond water and sediment were sampled and analyzed in 2011 (9 Aug.), and the water mass balance in the leachate pond was also examined [4]. At offshore disposal sites, most of the waste is submerged under seawater, and the velocity of water percolating through the waste layer is very slow [5]. Thus, water quality in the leachate pond may deteriorate significantly due to high nutrient and heavy metal levels, especially after disposal is completed, necessitating long-term management of sea area disposal sites [6] [7] [8].

In order to obtain detailed water quality data for the offshore MSW site leachate pond, the authors measured the pH, DO, and water temperature at the bottom of the leachate pond over an approximately 1-month period in September and November-December of 2011. Anaerobic batch experiments using sediment sampled on 9 Aug. were then conducted in order to characterize the elution of various materials from the leachate pond sediment. Based on DO data, sediment oxygen demand was calculated for use in developing a strategy to improve the water quality of the leachate pond.

2. Materials and Methods

2.1. Correlation between Various Parameters in the Water-Sediment of the Leachate Pond [4]

An overview of the targeted offshore MSW disposal site in Hiroshima Prefecture, Japan, is shown in **Table 1**. This disposal site is a semi-aerobic type with a final cover soil after closing and has a structure to prevent the percolation of precipitation water. Schematic of the pond structure, and photo taken in 2011 of the leachate pond is shown in **Figure 1**.

The leachate pond was anaerobic type without stirring. Leachate from the disposal site is collected in a leachate pond and sent to a wastewater treatment facility for treatment by coagulation-sedimentation, neutralization, and sand filtration. Particularly in summer, there is an offensive odor associated with the site, mainly due to hydrogen sulfide gas, and there are many chironomids in

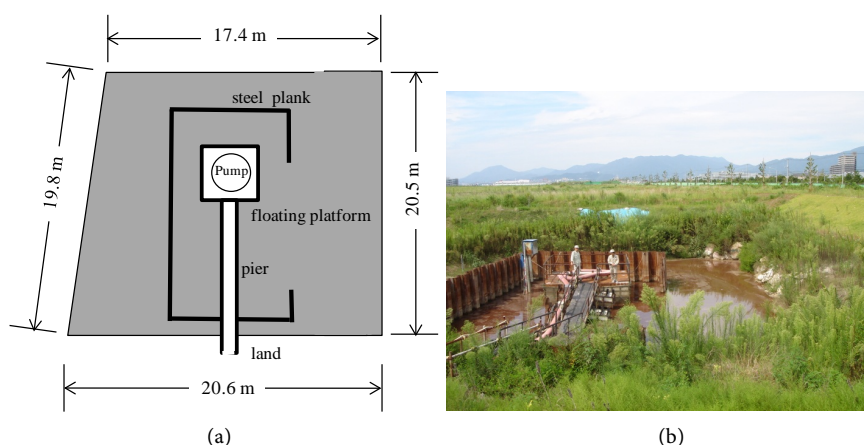


Figure 1. Condition of the leachate accumulation pond: (a) schematic of the pond structure, and (b) photo [4].

Table 1. Overview of the targeted offshore MSW disposal site [4].

	Landfilled period	Jan. 1991-Mar. 2009
	Disposal area	375,500 m ²
	Volume	4,855,000 m ³
Landfill	Mean depth	13.1 m
	Total weight of disposed waste	5,280,000 tons
	Waste types	Inorganic wastes (sludge, gypsum board, incineration residues, construction waste soil, debris, glass, and ceramic waste)
	Area	389.5 m ²
Leachate pond	Mean depth	2.9 m
	Mean retention time	37.8 days

flight in the area, resulting in complaints from neighboring residents.

Water and sediment in the leachate pond were sampled on 9 Aug., 2011. Samples were taken to the laboratory and kept frozen until analysis. They were thawed at every analysis and kept with the refrigerator until analysis. In this study, we analyzed pH (HM-50G, TOA DKK Corp.), EC (CM-30G, TOA DKK Corp.), SS, COD_{Mn} (KMnO₄ titration under acidic condition), NH₄⁺-N (Hach 8155, Hach Corp.), NO₂⁻-N (Hach 10019), NO₃⁻-N (Hach 8039), total phosphorus (Hach 8190), SO₄²⁻ (Hach 8051), Cl⁻ (AgNO₃ titration), Ca, total Cr (Total Cr), Mn, Fe, Cu, Zn, Cd, and Pb using an ICP quantometer (SPS7800, Seiko Instruments, Inc.) after passing through a 1 μm membrane filter in the pond water. Sediment samples were dried at 100°C, and ignition loss (IL) was measured by heating at 800°C for 2 h in an electric furnace (FUL230FA, Advantec Inc.). Element contents of Pb, Cd, Zn, Cu, Fe, total Cr, Mn and Ca were analyzed by ICP after digestion in nitric and perchloric acid. Total sulfide in each sediment sample was analyzed by the gas detector tube (Hedoro-tec S, Gastec Corp.).

These results have been reported [4]. It was thought that various chemical and microbial reactions would occur under conditions of high alkalinity and high salt concentrations in the water and sediment of the leachate pond [9] [10]. Thus, in this study, correlation coefficients for various parameters in both samples were calculated to examine this possibility qualitatively.

2.2. Continuous Measurement of pH, DO, and Water Temperature in the Leachate Pond

To examine the water quality of the leachate pond, continuous in situ measurements were made at the offshore landfill between 8-29 Sept., 2011 (1st period) and 20 Nov.-21 Dec., 2011 (2nd period). Data regarding pH, DO, and water temperature were collected using a pH and DO meter equipped with a data logger (DM-32P, TOA DKK Corp.). The meter was placed above the pond sediment. Measurements were taken every hour, and the data were collected after each measurement period.

2.3. Batch Experiments with Sediment to Estimate Pond Water Characteristics

For the purpose of estimating the pollution mechanism particularly in summer, sediment oxygen demand was determined in batch experiments with sediment sampled from the leachate pond. Sediment was sampled on 9 Aug, 2011 and then air dried at 50°C. Leachate pond water sampled at the same time or pure water was used as a solvent. The properties of the sediment and leachate water are shown in **Table 2**. The Fe and Ca content in the sediment were high. The water was highly alkaline (pH 9.5) and contained high concentrations of Cl⁻ and

Table 2. Properties of the sediment and leachate water sampled on 9 Aug. [4].

Sample	Parameter	Data	Parameter	Data
Sediment	Ca (mg/kg)	10,700	Cu (mg/kg)	289
	T-Cr (mg/kg)	132	Zn (mg/kg)	723
	Mn (mg/kg)	741	Pb	207
	Fe (mg/kg)	25,500	Ignition loss (%)	7.2
	pH (-)	9.5	Ca (mg/L)	3.10
	EC (S/m)	1.604	T-Cr (mg/L)	0.008
	SS (mg/L)	74	Mn (mg/L)	<0.005
Leachate water	COD _{Mn} (mg/L)	152.83	Fe (mg/L)	0.026
	NH ₄ ⁺ -N (mg/L)	0.41	Cu (mg/L)	0.008
	NO ₂ ⁻ -N (mg/L)	0.03	Zn (mg/L)	<0.005
	NO ₃ ⁻ -N (mg/L)	5.4	Cd (mg/L)	<0.005
	T-P (mg/L)	1.86	Pb (mg/L)	0.024
	SO ₄ ²⁻ (mg/L)	1100	Cl ⁻ (mg/L)	4,360

SO_4^{2-} . The particle size distribution of the sediment by sieving and a dynamic light scattering particle size analyzer (LB-550, Horiba Ltd.) is shown in **Figure 2**. The 50% cumulative passing diameter (d_{50}) was 0.024 mm, and visual inspection revealed a considerable amount of silt and slag. The number of sulfate-reducing bacteria in the sediment (7.1×10^5 cells/g-dry sediment) was determined using the MPN protocol with modified ISA medium [11].

For batch experiments, sediment and solvent were placed in a polyethylene bottle (50 mm diameter, approximately 65 mm height) with an inner cap, and the bottle was then sealed and thoroughly mixed. The experimental conditions are shown in **Table 3**. For each experimental run, some preliminary trials were first performed to decide experimental conditions. A total of 11 undisturbed sediment cores were sampled on 9 Aug., 2011 using an acrylic pipe (44 mm inside diameter, approximately 1,000 mm height). From the relationship between the water depth in the leachate pond and the dry weight of sampled sediment [4], the liquid/solid ratio [L/S] ranged from 5 to 10. The bottles were placed in an incubator maintained at either 20°C or 30°C. The conditions for run 4, which most closely matched the conditions of the site, were leachate water used as the solvent, L/S 5, and incubation at 30°C.

At the time the polyethylene bottle was removed from the incubator, the pH, EC, ORP (D-72, Horiba, Ltd.), and DO were measured immediately. Water was filtered using a glass-fiber filter with a pore size of 0.45 μm and then analyzed for

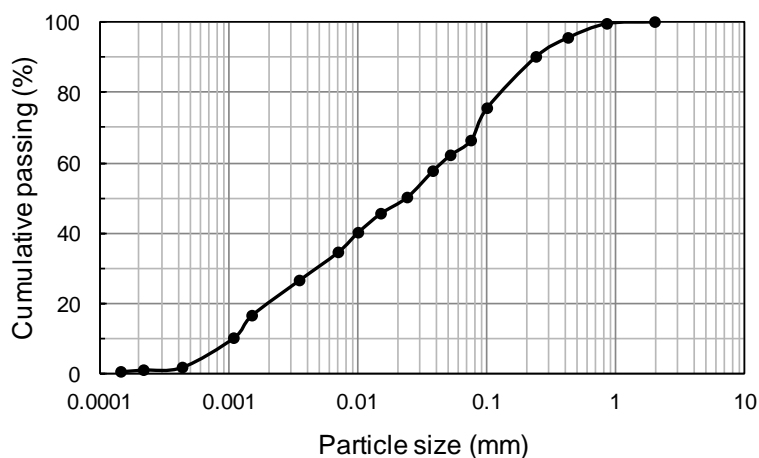


Figure 2. Sediment particle size distribution.

Table 3. Experimental conditions.

Run No.	Sediment (g-dry)	Solvent	L/S	Temperature (°C)
1	10	Pure water	10	20
2		Leachate water		
3	20	Pure water	5	30
4		Leachate water		

COD_{Mn}, NH₄⁺-N, NO₂⁻-N, NO₃⁻-N, T-P, SO₄²⁻, Ca, T-Cr, Mn, Fe, Cu, Zn, Cd, Pb, and Cl⁻. Sediment was collected from the bottle and kept frozen until analysis. After thawing, the water content of the sediment was measured, and T-S was analyzed. The experiment was conducted in the shade. Results are reported as the mean of 3 samples collected at each sampling time.

3. Results and Discussion

3.1. Correlation between Various Parameters in the Water-Sediment of the Leachate Pond

Figure 3 shows the correlation between various parameters examined in the water and sediment samples collected on 9 Aug, and only correlations satisfying are shown in the figure. Positive correlations are shown by solid lines, and negative correlations are indicated by dashed lines. In the sediment, Fe was strongly correlated with pond water parameters (pH, EC, NH₄⁺, T-P, Ca, Fe, Cl⁻). Ca was correlated with pH, COD, NH₄⁺, Ca, and Fe in pond water parameters. In the pond water, COD, Ca, and pH were correlated more with sediment parameters. The pH of the pond water was not correlated with any other water parameter, but it was correlated with IL, Ca, Fe, and Zn in the sediment. This indicates that the pH in the pond water is influenced by elution of Ca from the sediment and by the inflow of fresh leachate from the landfilled layer.

With respect to organic substances, the sediment IL, which was 7.2% - 12.1% in each sample, was correlated only with pond water pH. Pond water COD was correlated with Ca, T-Cr, Mn, Cu, and Zn but was not correlated with any other water parameter. As chlorophyll-a was not detected in any of the samples, the pond water COD was thought to be derived from landfilled waste that was not incinerated. Especially in summer, it was anticipated that the DO would be almost completely consumed and that the sulfate-reducing activity would be high,

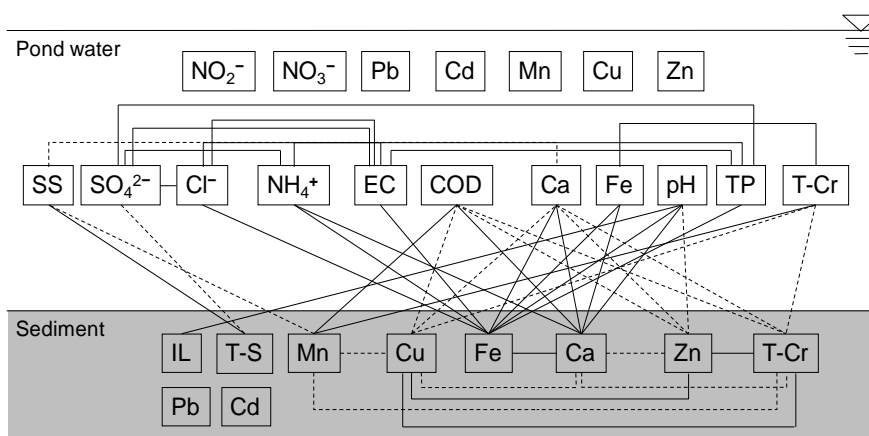


Figure 3. Correlation between various parameters in the water-sediment of the leachate pond. Only correlation coefficients between each parameter (r) satisfying $|r| \geq 0.8$ are shown. Positive and negative correlations are shown by solid lines and dashed lines, respectively.

as T-S and SO_4^{2-} were negatively correlated ($r = -0.87$). Of the heavy metals identified in the pond water (Cr(VI), Mn, Fe, Cu, Zn, Cd, and Pb), none of them exceeded the effluent standard in Japan.

3.2. Continuous Measurement of pH, DO, and Water Temperature in the Leachate Pond

Figure 4 shows the results of the continuous measurement of pH, DO, and water temperature above the sediment of the leachate pond. Lost data between 23 Nov and 13 Dec in the 2nd period are indicated by dashed lines. The daily average temperature in Hiroshima City is shown for the same periods [12]. Although the patterns of change in the pond water temperature and the temperature in Hiroshima City were similar during the 1st period, the water temperature was about 2°C - 10°C higher than the temperature during the 2nd period. These data suggest that the temperature stratification in the water in the leachate pond had gradually broken down as a result of convection beginning in November. The pH during the 1st period was approximately 10 - 11, and during the 2nd period, the pH was nearly constant at about 11.5. The pH was high because about 70% of the land filled waste was incineration residues.

The DO was almost zero during both the 1st and 2nd periods. During the 1st period, the pond water temperature was about 20-27°C, and there was an odor, due primarily to hydrogen sulfide. In addition, the Fe content of the sediment was high (2.55%) and the color of the pond water seemed to be reddish-brown due to the presence of Fe(II). The absence of DO was attributed primarily to consumption by microorganisms. During the 2nd period, the water temperature was about 13°C - 21°C, and there was no smell around the pond. The color of the pond water was blackish-brown. It was suggested that DO supplied to the bottom of the pond by convection was consumed by chemical oxidation of Fe(II) and sulfide generated in the summer rather than by microorganisms.

3.3. Batch Experiment with Sediment

DO, ORP, pH, and EC results are shown in Figure 5. The DO declined in all

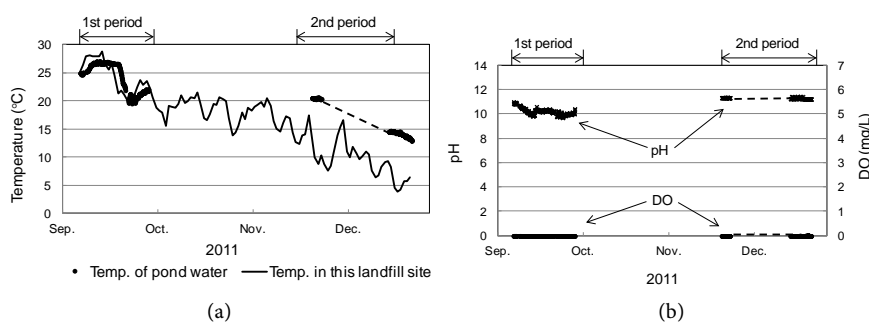


Figure 4. Results of continuous measurements in the leachate pond. (a) Water temperature in the leachate pond. The daily average of the temperature of Hiroshima City is shown for the same time period [12]; (b) pH and DO. Note that the broken line between 23 Nov. and 13 Dec. in the 2nd period indicates data loss.

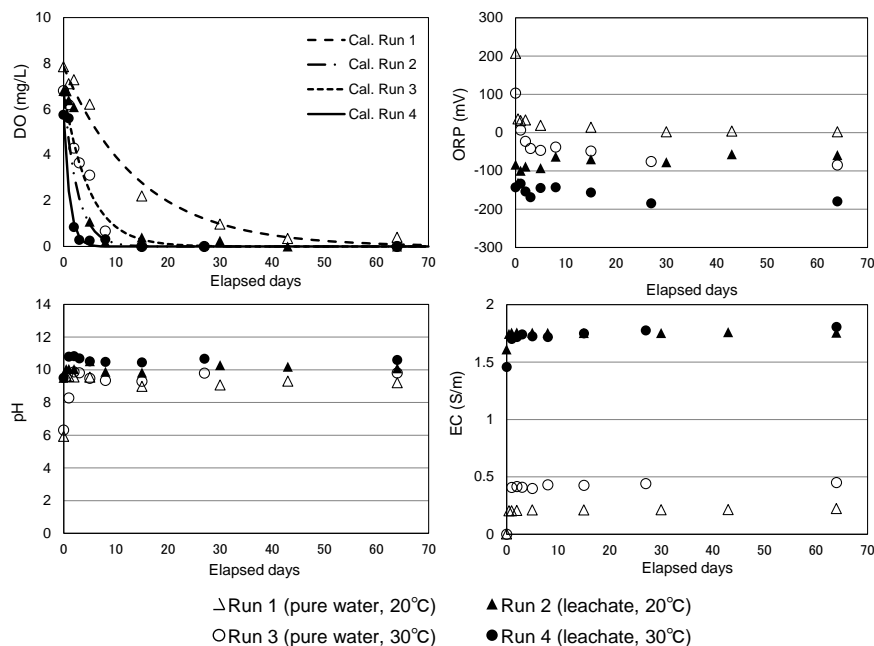


Figure 5. DO, ORP, pH, and EC results for the batch experiments. Lines in the DO graph show calculated data from curve fitting of the batch experiment DO concentration data using equation (2). The rate constant of oxygen consumption (L/kg/d), k , were 2.70, 1.01, 4.32, and 1.23 L/kg/d for runs 1, 2, 3, and 4, respectively.

experimental runs, and the rate of decline was highest with leachate used as the solvent and 30°C as the incubation temperature. Particularly in run 4 (leachate, L/S 5, 30°C), which most closely matched the conditions of the site, the DO declined to almost zero within 3 days. The lowest rate of DO consumption (0.40 mg/L after 64 days) was observed in run 1 (pure water, L/S 10°C, 20°C). The ORP also decreased with time, reaching -180 mV in run 4 and -60 mV in run 2 (leachate, L/S = 10°C, 20°C). In runs 1 and 3, which used pure water as the solvent, the pH rose immediately after the beginning of the experiment, reaching approximately pH 9 - 10 and then remaining stable. In contrast, in runs 2 and 4, which used leachate water as the solvent, the pH was approximately 10 - 10.5 and did not fluctuate considerably throughout the experiment. The EC was almost constant in each run, although it did rise immediately after the beginning of the experiment, suggesting that elution of ions from the sediment reached equilibrium very soon after the experiment began. The trends in metal concentrations were similar to the trend for EC. None of the metal concentrations exceeded the effluent standard in Japan, and almost all were present at levels at least 10 times lower than the effluent standard during the experimental period.

Figure 6 shows the results of COD_{Mn} , $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and T-S analyses. COD_{Mn} rose gradually and then became constant. As for the amount of increase in COD_{Mn} over the experimental period, it was largest in run 3, at 131.1 mg/L, followed by run 4 (in which the condition most closely mimicked those of the

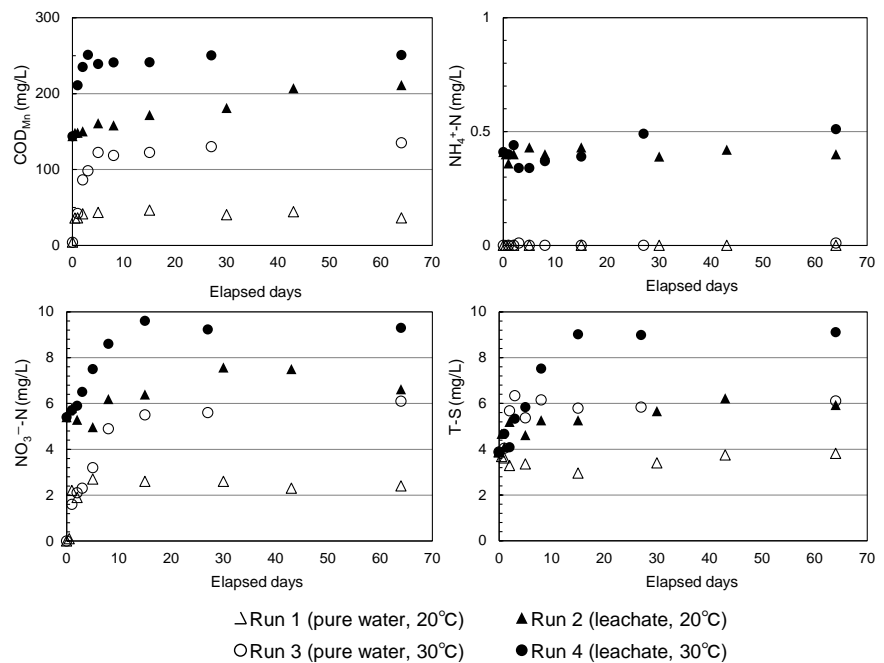


Figure 6. COD_{Mn} , $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and T-S results for the batch experiment.

site), at 107.1 mg/L. The $\text{NO}_3^-\text{-N}$ concentration rose, similar to the COD_{Mn} , whereas there was little change relative to the initial state for $\text{NH}_4^+\text{-N}$. The rise in $\text{NO}_3^-\text{-N}$ concentration could only be explained by diffusion from the sediment. With respect to the amount of increase in $\text{NO}_3^-\text{-N}$ during the experimental period, it was greatest in run 3 (pure water, L/S 5, 30°C), at 6.1 mg/L, followed by run 4 (which most closely mimicked the site conditions), at 3.9 mg/L, a trend similar to COD_{Mn} .

The amount of increase in T-S was the greatest in run 4 (5.23 mg/L). This was thought to be due to the activity of sulfate-reducing bacteria following the decreases in DO and ORP. Run 3, which used pure water as the solvent, exhibited a 2.23 mg/L increase in T-S, which was attributed to the reduction of residual SO_4^{2-} in the sediment. In run 2, which used leachate water as the solvent and incubation at 20°C, the T-S concentration rose 2.05 mg/L.

The optimum temperature for many sulfate-reducing bacteria is thought to be 25°C - 30°C [13]. Run 2 appeared to provide a micro-environment suitable for the growth of sulfate-reducing bacteria, even though some nitrate acid remained and the surrounding temperature was 20°C. Other studies have reported that sulfate-reducing activity occurs at a $\text{SO}_4^{2-}/\text{COD}$ stoichiometry of ≥ 0.67 [14], [15]. In the present study, this stoichiometry was exceeded in all runs.

From the results of the batch experiment, sediment oxygen demand was calculated using a brief numerical model [16]. When the oxygen consumption rate is in proportion to the DO concentration, the following equation holds:

$$V \frac{dC}{dt} = -kCm \quad (1)$$

where C represents the DO concentration (mg/L), t represents time from the start of the experiment (days), V represents solvent volume (0.1 L), m represents the weight of sediment used in the experiment (kg-dry), and k represents the rate constant of oxygen consumption (L/kg/d).

Solving Equation (1) under the initial condition ($C = C_0$ at $t = 0$), the following equation is obtained:

$$C = C_0 e^{-\frac{km}{V}t} \quad (2)$$

From curve fitting of the batch experiment DO concentration data using Equation (2) (see **Figure 5**), k values of 2.70, 1.01, 4.32, and 1.23 L/kg/d were obtained for runs 1, 2, 3, and 4, respectively.

The average oxygen demand rate per unit area (designated average sediment oxygen demand [SOD_{ave}] in this study) [17] was calculated using Equation (3):

$$SOD_{ave} = -\frac{1}{T} \cdot \frac{1}{S} \int_0^T \frac{dC}{dt} dt \quad (3)$$

where SOD_{ave} is expressed as mg/m²/d, T represents the period during which the DO was almost zero (days), and S represents the sectional area of the polyethylene bottle (m²).

When calculating the SOD_{ave} using the results of run 4 (the conditions of which most closely matched the conditions of the site) and $T = 3$ days, a value of 1114.7 mg/m²/d was obtained. The area of the leachate pond was 389.5 m² (see **Table 1**), and the mean water volume of the pond was 1185 m³, with a range of 800 to 2000 m³ after the disposal site was closed [4]. It was concluded that an oxygen demand of approximately 434 g/d would be required to maintain DO in the pond in the summer.

Factors influencing leachate pond water quality include the composition of the landfilled waste, the time elapsed after landfilling, the landfilling method, chemical and microbial reactions occurring in the pond, and weather conditions, such as temperature and precipitation [18]. To prevent deterioration of the pond water quality, it is necessary to consider measures such as dredging [19] in addition to supplying oxygen by aeration in relation with the total cost [20] [21] [22] [23]. These measures must lead to a decrease in the load on leachate treatment equipment.

4. Conclusions

In this study, the pH, DO, and water temperature at the bottom of a leachate pond at an offshore MSW disposal site were measured in order to better understand factors affecting pond water quality following site closure. An anaerobic batch experiment using sampled sediment was also conducted to characterize the elution of materials from the sediment. The sediment oxygen demand was calculated from the resulting data for use in developing methods to improve the quality of leachate pond water. The results of this study are as follows:

The patterns of change in the leachate pond water temperature and the daily averaged temperature were similar during the 1st period (8-29 Sept., 2011), the water temperature was about 2°C - 10°C higher than the temperature during the 2nd period (20 Nov.-21 Dec., 2011). It was thought that the water temperature stratification in the leachate pond had broken down as a result of convection developing in the winter. The pH was almost constant at 10 - 11 during the 1st period and approximately 11.5 during the 2nd period. The DO was almost zero during both the 1st and 2nd periods.

In the anaerobic batch experiments with sampled sediment, the highest rate of decrease in DO was observed when leachate was used as the solvent and samples were incubated at 30°C. With respect to ORP and T-S, the condition in which leachate was used as the solvent, the L/S was 5, and samples were incubated at 30°C (conditions most closely mimicking those of the site) became the most anaerobic. T-S was also generated under the condition in which leachate was used as the solvent, the incubation temperature was 20°C, and the L/S was 10. The data from these experiments suggested the existence of a micro-environment suitable for the growth of sulfate-reducing bacteria, even though the surrounding temperature was lower than the optimum temperature.

The SOD_{ave} was calculated from batch experiment DO data using a brief numerical model. The SOD_{ave} was 1114.7 mg/m²/d, suggesting that in summer, it is necessary to supply approximately 434 g of oxygen per day to the leachate pond.

To satisfy Japanese effluent water standards, it is necessary to continue to manage for more than 20 years in general. It is necessary to shorten the management period, and control measures for the leachate pond are critical to the appropriate management of landfills.

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