

Soluble substrate concentrations in leachate from field scale MSW test cells

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Received 2 September 2005; received in revised form 26 October 2005; accepted 1 November 2005

Available online 4 January 2006

Abstract

We have monitored non-biodegradable soluble COD of leachates derived from two different landfill test cells, which were constructed at Odayeri Sanitary Landfill and operated with (C2) and without (C1) leachate recirculation for 1080 days. Refuse height and the placement area of test cells were 5 m and 1250 m² (25 m × 50 m), respectively. For leachates of both cells, initial inert soluble COD fraction (f_{non}) increased from its initial value of 0.01 to around 0.1 after 300 days of operation. Due to the development of anaerobic conditions, the value showed an increasing trend and the maximum value of 0.4 was reached on day 600. Several suitable models were also fitted to the experimental data on the basis of statistical reasoning. So as to evaluate the goodness of obtained fits, the calculated values of the sum of squares due to error (SSE), *R*-square, the residual degrees of freedom (DFE), adjusted *R*-square, and root mean square errors (RMSE) associated with the model results were compared. Logistic model for C1 test cell and Gompertz model for C2 test cell gave the best fits to the experimental data. Moreover, using the fitted model parameters, pollution loads, and BOD/COD ratios in leachates from C1 (control) and C2 (recirculation) cells were estimated and deeply discussed. The results of the study can be satisfactorily used to predict change in the composition of leachate over time, which may help to obtain better effluent quality in biological treatment of leachate.

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Keywords: Inert fraction; Leachate; Modeling; MSW; Recirculation; Test cell

1. Introduction

Municipal solid waste (MSW) is a common name for a very heterogeneous mixture of wastes of residential, commercial, sometimes industrial, and urban origin. MSWs consist of different organic and inorganic fractions such as food, vegetables, paper, wood, plastic, glass, metals, and other inert materials [1,2]. Despite the variability in its composition, organic content constitutes the highest percentage of solid waste. Sanitary landfilling is a controlled method of MSW disposal. Economical considerations continue to maintain landfills as the most attractive route for MSW disposal [3,4]. Disposal of domestic and commercial solid wastes at landfills leads to generation of leachate due to the inherent moisture of the wastes and to the external infiltrating liquids. Landfill leachate is one of highly contaminated and heterogeneous wastewaters. Its composition

and characteristics, in particular, strictly depend upon factors such as waste types, climate, and contents of organic matter, hydrogeological structure of the landfill, operational condition, and age of landfill [5–8]. Leachate often contains high concentrations of easily biodegradable and non-biodegradable organic matters as well as inorganic ions. At the same time, the characteristic of the leachate vary with regard to its composition and volume, and biodegradable matter present in the leachate with time [9–11]. Because, as a landfill becomes older, there is a shift from a relatively short aerobic period to a longer time anaerobic decomposition period which has two distinct sub-phases: an acidic phase followed by methanogenic phase. All these factors make leachate treatment difficult and they need to be taken into account when different treatment processes are considered. Since information and research surrounding the operation of landfill sites became available in a better form leachate management and treatment has gained importance. At present, collection and treatment of landfill leachates are issues surrounding the operation of landfill sites [12,13]. One of the available options is the biological leachate treatment by either aerobic or anaerobic

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processes [14]. However, anaerobic treatment methods are more suitable for concentrated leachate streams [13–17]. Treated water out of a biological wastewater treatment plant contains a number of soluble organic compounds such as residual rapidly, slowly and non-biodegradable substrate, inter and final products, and soluble microbial products [18]. Likewise, leachates from landfill also contain same compounds. Hence, the capabilities of the present leachate treatment processes are quite limited due to the high contents of both initially present inert COD in influent and the inert COD produced by microbial activities.

The objective of this study is to investigate change of present non-biodegradable soluble inert COD (S_{non}) in leachate with refuse age. For this purpose, soluble inert COD production of leachates from test cells are evaluated using sigmoid shape type (Gompertz, Richards, Schnute, and the Logistic models) by depending on refuse age.

2. Materials and methods

2.1. Test cells

Test cells (C1 and C2) used in the study are constructed at Odayeri Sanitary Landfill, which is one of the MSW landfills in Istanbul city. This site is designed for 25 years use with the area of 73 ha, and up to now only 37 ha has been used. Solid

wastes at the European side of Istanbul are collected at three transfer stations (Baruthane, Halkali, and Yenibosna) and then transported to Odayeri Sanitary Landfill. Approximately, daily 7000 tonnes of MSW are disposed in this site.

Test cell was constructed according to technical standards of a sanitary landfill. Approximately 5500 tonnes of MSW are filled to cell at equal quantities between 3rd and 9th October 2001. Refuse height is 5 m and placement area of cell is 1250 m² (25 m × 50 m). The average density and porosity of MSW in the test cells are around 0.85 tonnes/m³ and 60%, respectively. Although no part of the core study presented and discussed in this paper, a waste characterization study was carried out at the test cells. For this purpose, weight percent of each component of MSWs were determined by sample symbolizing wastes placed to cells. Characterization studies were repeated three times during the active filling of the cells. The MSW composition is determined as 44% organic, 8% paper, 6% glass, 6% metals, 5% plastic, 5% textile, 9% nylon, 17% ash, and others. These values are average percentages of waste components obtained from characterization study carried out by 3 m³ waste symbolizing wastes filling to cells.

The details of cell are shown in Fig. 1. Leachate samples are taken from the collection tanks, which were connected to the leachate drainage pipelines installed in the bottom of the test cells for the analysis. C1 and C2 test cells were constructed according to the technical standards of a sanitary landfill.

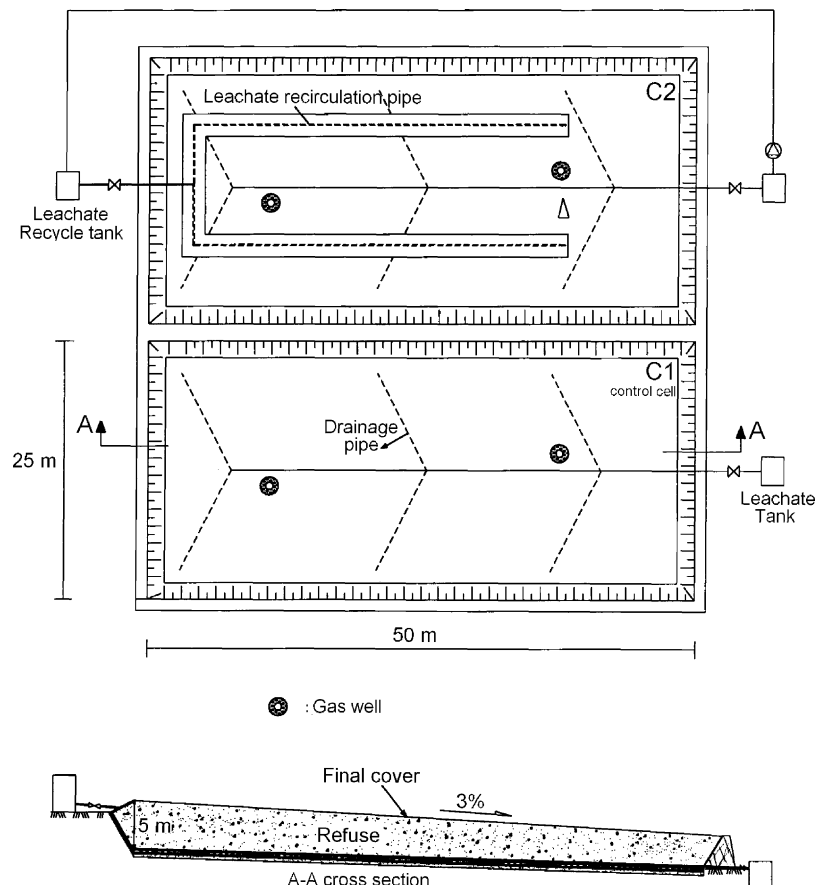


Fig. 1. Construction details of the cells.

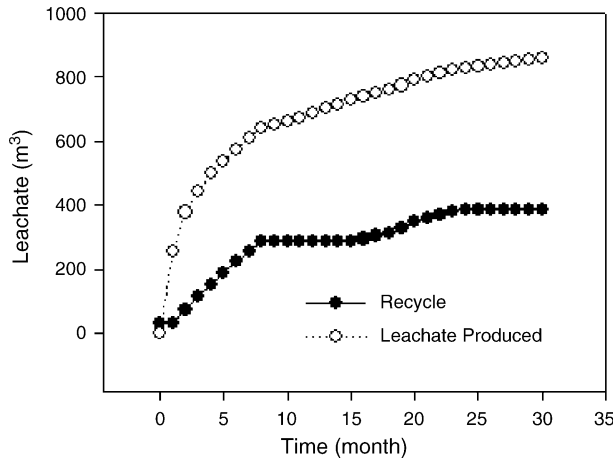


Fig. 2. Cumulative leachate production and recirculation.

Landfill bioreactors were operated as leachate recirculation (C2) and control cell (C1).

Effect of leachate recirculation on the soluble inert COD formation and its modeling are investigated on leachate samples from C2. The produced cumulative leachate and recirculation amounts are given in Fig. 2. About 44.8% of total leachate production is recirculated to the system in order to increase moisture content, which led to increase in substrate degradation and methane production rate.

2.2. Determination of soluble inert CODs

In this study, non-biodegradable soluble fractions of leachates from test cells are evaluated. The total soluble COD concentration of leachate (S_{T0}) is equal to the sum of the initially present inert soluble COD (S_{non}) which by-passes the treatment system without any change and the readily biodegradable soluble COD [7,19]. S_{non} content of leachate gradually increases as a landfill stabilizes, therefore, S_{non} should be considered carefully to design the leachate treatment plant with respect to landfill ages. The soluble COD in the effluent from a bioprocess treating leachate includes not only biodegradable and non-biodegradable compounds from the raw leachate, but also biodegradable and non-biodegradable compounds produced by the microbial activities in the treatment system itself. Experimental approaches, both comparison method and incremental method, are proposed to separately assess S_{non} and soluble microbial product (SMP) by Germirli et al. [20] and Ince et al. [19]. Leachate may include soluble microbial products if it is considered as an anaerobic bioreactor where organics in solid waste degrade by anaerobic process. Several authors have stated that SMPs results from intermediate or final products of substrate degradation and endogenous cell decomposition [21], originates from pool of organic compounds that result from substrate metabolism (usually with biomass growth), and biomass decay during the complete mineralization of simple substrates [22].

The non-biodegradable soluble COD fraction of raw leachate is determined by comparison method in present study. For this purpose, two anaerobic batch reactors having the same initial COD are operated in parallel. One is fed with the leachate as

a sole substrate, and the other with glucose. When the soluble COD in each reactor have declined to constant plateau value after the biodegradable substrate is almost entirely consumed, at this point the measured COD value is considered as a residual soluble COD (S_R) including both S_{non} and SMP [7]. SMP is obtained by glucose fed batch reactors seeded with acclimated (50% glucose and 50% leachate) anaerobic biomass. The value of SMP is determined with measuring the residual COD of glucose reactor, because glucose itself has no inert organic material. Since the seed biomass is acclimated to glucose and leachate, SMP is accepted to equal for each reactor. Then S_{non} content of the leachate is calculated given as follows:

$$S_{non} = S_{ul} - SMP \quad (1)$$

where S_{non} is the soluble non-biodegradable COD (mg/l); S_{ul} is the ultimate COD of soluble leachate reactor (mg/l); finally SMP is the residual COD of glucose fed reactor in mg/l.

By the definitions, fraction of initial soluble inert COD fraction (f_{non}),

$$f_{non} = \frac{S_{non}}{S_{T0}} \quad (2)$$

where S_{T0} is the total soluble COD concentration in raw leachate.

2.3. Operation of anaerobic batch reactors

Non-biodegradable soluble COD in leachates was carried out at anaerobic batch reactors having 500 ml volumetric capacity. Two parallel reactors are used to perform the experiments as in Fig. 3. The reactors are fed by N_2 gas for 5 min to supply anaerobic conditions before sealing with glass stoppers. Subsequently, nutrient solution, acclimated biomass (150 mg/l), and samples are filled by 50 ml glass syringe from sampling septum. Anaerobic granular bacteria is used for acclimation of seed. These bacteria with median bioparticle diameter of 2 mm are obtained from anaerobic sludge digester of the wastewater treatment plant and acclimated with 5000 mg/l soluble COD concentration (50% glucose and 50% leachate). Batch reactors were seeded with acclimated biomass in a 150 mg VSS/l concentration. The reactors are maintained at a constant temperature of 35 °C in a water bath and mixed with the magnetic stirrer at a rate of 10 min/h. Change of soluble COD in reactors is monitored at samples taken by 5 ml glass syringe. Batch reactors started with the same initial COD, one with the soluble leachate (diluted to 5000 mg COD/l) filtrated with 0.45 μ m membrane filters (S&SCHUELL, 47 mm), and the other with glucose solution being COD equivalent 5000 mg/l. At the end of the anaerobic studies, non-biodegradable soluble COD fractions of leachates are determined through Eqs. (1) and (2).

For the nutrient solution, the composition of the basal medium containing all micro and macronutrients are required in order to obtain optimum anaerobic microbial growth as given in Table 1 [23]. Nutrient solution also acts as a pH buffer is added to the glucose reactor.

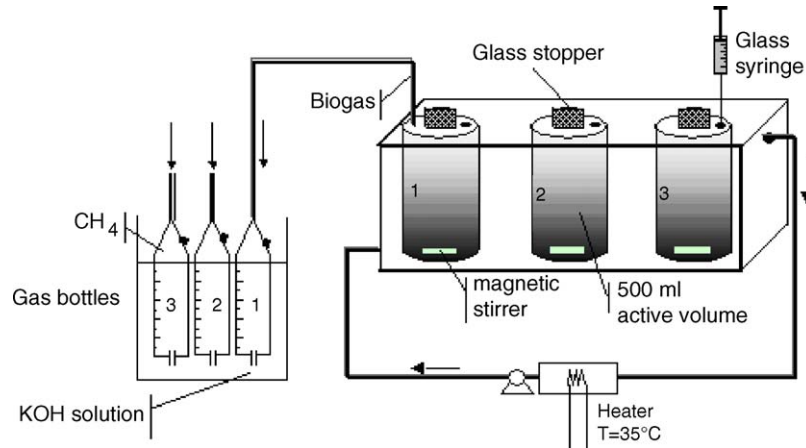


Fig. 3. Anaerobic batch reactors for soluble inert COD.

2.4. Model study

The Gompertz and Logistic models have a similar sigmoid shape with a clear inflection point. Selecting the most appropriate growth model is often a matter of trial and error. Moreover, different criteria for determining the suitability of one particular model over another vary: some authors have relied on mathematical measures of goodness of fit [24], while others have focused on direct comparisons of particular growth parameters as predicted by the various models [25]. One is based on the inflection of the slope of the growth curve in the exponential phase [26], while the other is taken to be the growth rate at infinite dilution [27]. These type models are commonly used as microbial growth kinetic [24,25,28]. There are a number of modified growth models which are Gompertz, Richards, Schnute, and the Logistic models, and others. Addition to these models it is possible to use some traditional statistical curve fitting models such as exponential, linear, quadratic, etc. Lay et al. [29], in a study, developed a model from the Gompertz equation for estimating the methane production rate and the lag-phase time under various conditions in landfill bioreactors. Similarly, in the other study of Lay et al. [30], Gompertz equation was applied to estimate hydrogen production potential and rate. Soluble microbial products can accumulate as landfill become older as a result of enhancing microbial activity in landfill bioreactors. They are

an inert material being resistance to biological treatment and can accumulate by depends upon microbial growth. Considering these factors, in this work, we applied sigmoid shape type (Gompertz, Richards, Schnute, and the Logistic models) to soluble inert COD formation by depending on refuse age.

One of the objectives of this work is to obtain a model, on the basis of statistical reasoning. To describe such a curve and to reduce measured data to a limited number of interesting parameters, investigators need adequate models. Several curve fitting models are tested and compared in such a way that they contain parameters that are microbiologically relevant. MATLAB® 7 (R14) package program was used for computations. The nonlinear equations which are given in Table 2, were fitted to growth data by nonlinear regression with a Trust-Region Reflective Newton algorithm. This is a search method to minimize the sum of the squares of the differences between the predicted and measured values. The main disadvantage of least squares fitting is its sensitivity to outliers. Outliers have a large influence on the fit because squaring the residuals magnifies the effects of these extreme data points. To minimize the influence of outliers, robust least squares regression was used. Least absolute residuals (LAR) scheme finds a curve that minimizes the absolute difference of the residuals, rather than the squared differences. Therefore, extreme values have a lesser influence on the fit. Then

Table 1
Composition of basal medium

Compounds	Concentration (mg/l)	Compounds	Concentration (mg/l)
NH ₄ Cl	1200	CuCl ₂ ·2H ₂ O	0.5
MgSO ₄ ·7H ₂ O	400	ZnCl ₂	0.5
KCl	400	AlCl ₃ ·6H ₂ O	0.5
Na ₂ S·9H ₂ O	300	NaMoO ₄ ·2H ₂ O	0.5
CaCl ₂ ·2H ₂ O	50	H ₃ BO ₃	0.5
(NH ₄) ₂ HPO ₄	80	NiCl ₂ ·6H ₂ O	0.5
FeCl ₂ ·4H ₂ O	40	NaWO ₄ ·2H ₂ O	0.5
CoCl ₂ ·6H ₂ O	10	Na ₂ SeO ₃	0.5
KI	10	Cysteine	10
MnCl ₂ ·4H ₂ O	0.5	NaHCO ₃	6000

Table 2
Used models and their equations

Name of model	Model
Gompertz	$f(x) = A_0 + A_{\max} \exp(-\exp(((m \times \exp(1))/A_{\max}) \times (L - x) + 1))$
Logistic	$f(x) = A_0 + A_{\max}/(1 + \exp((4 \times m/A_{\max}) \times (L - x) + 2))$
Richards	$f(x) = A_0 + A_{\max} \times (1 + v \times \exp(1 + v) \times \exp((m/A_{\max}) \times (1 + v) \times (x + (1/v)) \times (L - x)))^{(-1/v)}$
Cubic	$f(x) = p_1 \times x^3 + p_2 \times x^2 + p_3 \times x + p_4$
Polynomial	$f(x) = p_1 \times x^4 + p_2 \times x^3 + p_3 \times x^2 + p_4 \times x + p_5$
Linear	$f(x) = p_1 \times x + p_2$
Power	$f(x) = a \times x^b + c$
Quadratic	$f(x) = p_1 \times x^2 + p_2 \times x + p_3$
Exponential 1	$f(x) = a \exp(b \times x)$
Exponential 2	$f(x) = a \exp(b \times x) + c \exp(d \times x)$

the model results and coefficients (with 95% confidence interval) were calculated.

Finally the best fitting model for our data set was defined. One way to select the best among models is to compare them statistically. With the aim of evaluating the goodness of obtained fits, the sum of squares due to error (SSE), *R*-square, the residual degrees of freedom (DFE), adjusted *R*-square, and root mean square errors (RMSE) associated with the output model result were calculated and compared.

3. Results and discussion

3.1. Experimental results

Landfilling, composting, and co-digestion of solid wastes with sewage sludge represent the most economical methods for the disposal of MSW [31]. Anaerobic decomposition of wastes' organic contents in a landfill is the most commonly used alternative for MSW. Municipal landfill leachates consist of constituents in different molecular weight fractions. In particular, the old leachate constituents distribute in a wide range of molecular weight fractions which for old leachates are found as complex structures formed by functional groups containing N, S, and O atoms. The young leachate fractions have actually low molecular weights. The low molecular weight fractions of leachates are characterized by linear chains which are substituted through oxygenated functional groups such as carboxyl and/or alcoholic [32].

Sometimes, pollutant concentrations of biologically treated leachate exceed discharge standards due to inappropriate estimation or consideration of S_{non} and SMP. Since S_{non} is by-passes the treatment system without any change, careful consideration of S_{non} and SMP is very important structure and operating parameters and to estimate effluent residual COD [7]. Furthermore, S_{non} of leachate gradually increases as a landfill stabilizes, therefore, it should be considered carefully to design the leachate treatment plant with respect to refuse age. Therefore, the objective of this work is to determine and model the change of present S_{non} in leachate from test cells (C1 and C2) with refuse age.

The raw soluble COD (S_{T0}) contents of leachate from C1 and C2 are shown in Fig. 4. Initial values of S_{T0} are very high due to the volatile fatty acid formation at initial phase of anaerobic digestion of organic wastes in test cells. Maximum S_{T0} values are 50,000 and 43,000 mg/l for C1 and C2 leachate, respectively. After 1080 days, the S_{T0} reaches to 1250 and 460 mg/l by decreasing as refuse age increases for C1 and C2, in respective order.

Fig. 5 illustrates soluble inert COD content (S_{non}) of leachate from C1 and C2. According to the results, at the beginning of operation, when biodegradable organic portion is high in leachate, the S_{non} are 250 and 350 mg/l for C1 and C2 leachate. While the S_{T0} content of leachate decreases, it is observed that the S_{non} contents reach to 450–650 mg/l values for both test cell by increasing. This increasing value originate from including SMP and complex organics of leachate from test cells being bioreactor degrading organic waste anaerobically. It is concluded that the S_{non} increases as test cells stabilizes. After 1080

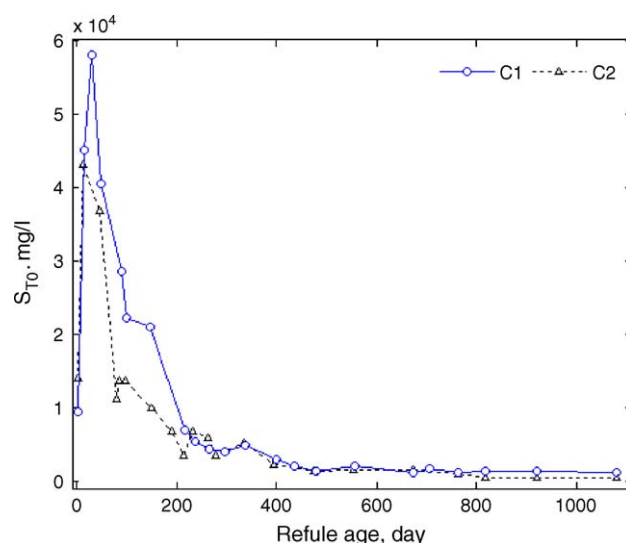


Fig. 4. Total soluble COD (S_{T0}) contents of leachate from C1 and C2.

days, S_{non} are determined to be 500 and 190 mg/l for C1 and C2 leachate, respectively. Having lower concentration in C2 than C1 is results from fastly decreasing of S_{T0} by depending upon accelerating waste stabilization by leachate recirculation operation. The practice of leachate recirculation accelerates waste degradation through provision of moisture, dilution of potential inhibitors to methanogenesis and encouragement of water flux for microbial, substrate, and nutrient transfer. This in turn facilitates more rapid waste stabilization, as well as increased levels of chemical and physical leaching of contaminants from waste. This has the associated benefits of improving overall leachate quality [33]. The value of f_{non} is 0.01 at the beginning of operation. The value increased relatively fast in C2 leachate then reached a maximum value of 0.4 for C1 and C2 leachates. This observation may originate from soluble microbial product formation as a result of microbial activities of methanogenic bacteria during the waste degradation in test cells (C1 and C2). Accordingly, it is determined that the value of f_{non} increased as landfill getting older.

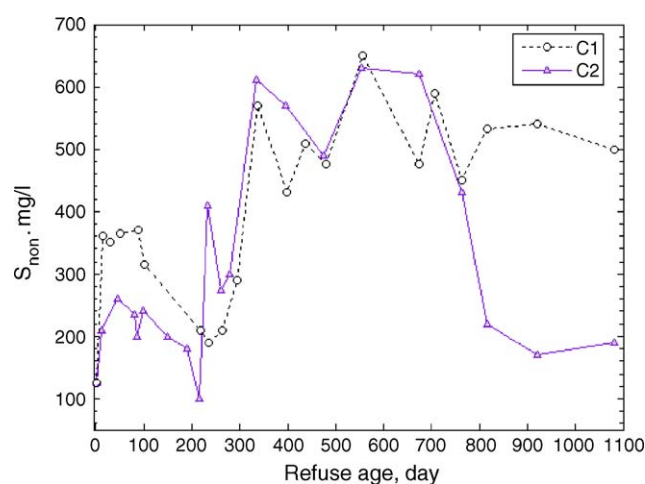


Fig. 5. The change of S_{non} contents in leachate from C1 and C2 with refuse age.

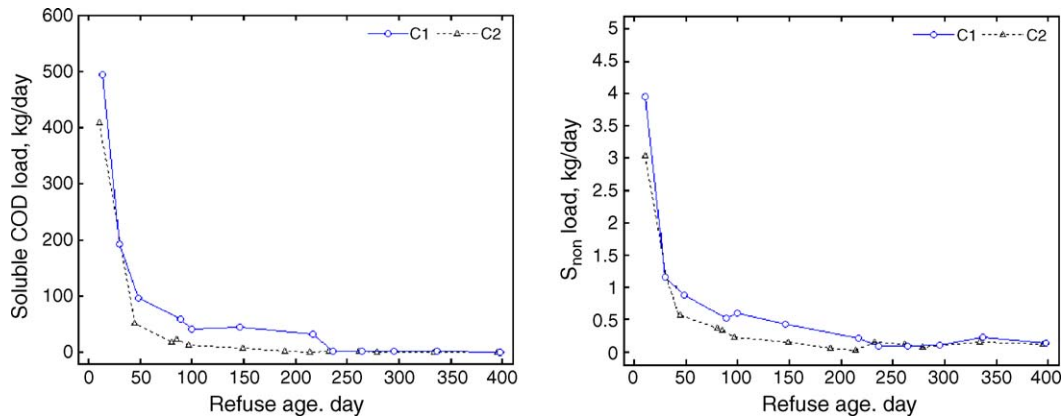


Fig. 6. Soluble COD (S_{T0}) and soluble inert COD (S_{non}) of leachates from C1 and C2 test cells.

3.2. Pollution loads and BOD/COD ratios

It was reported that inert compounds could be produced by microorganisms and accumulated with time in an anaerobic membrane bioreactor [34]. Hasar et al. [35,36] investigated the relationship between inert CODs in the influent and the effluent in aerobic bioreactor treating domestic wastewater. Similarly, sanitary landfills can be considered as an anaerobic bioreactor where organics in solid waste degrade by anaerobic process. Hence, inert compounds could be produced by anaerobic microorganisms and accumulated with time in an anaerobic landfill bioreactor. In this section of the study, COD and inert COD loads were evaluated by depending upon refuse age. Soluble COD (S_{T0}) and soluble inert COD (S_{non}) of leachates from C1 and C2 test cells are shown in Fig. 6. As can be seen from this figure, COD loads were more early reduced in C2 test cell than C1 control cell by depending upon decreasing of discharged leachate quantity and accelerating waste stabilization by leachate recirculation operation. Hence, it is observed that inert COD load is decrease passing through unchanged from external leachate biological treatment plants.

BOD parameter in leachate can be evaluated as an indicator of the anaerobic decomposition rates of organics in municipal solid waste. The BOD values in C1 and C2 leachates reached to the maximum values nearly after one month of landfilling. Then this value decrease rapidly depending upon refuse age.

Fig. 7 shows change of BOD/COD ratios of leachates derived from C1 and C2 test cells. The maximum values of measured BOD are 52 and 63 g/l for C1 and C2 cells, respectively. BOD to COD ratios are around 0.8 for both test cell during the acidogenic phase. This ratio indicates the biological activity of the leachate. The high ratio is result from the acid phase of the anaerobic degradation of organic matter. Then this ratio reached to 0.06 value by decreasing.

3.3. Results of curve fitting

When visually checked, Gomperts, Logistic, Richards, polynomial, and cubic models gave reasonably better fits which can explain the characteristics of the data. On the other hand, some of the models visually did not give reasonably good fits of the

data. According to R -square values (cf. Table 3) all the models seem reasonable for fitting such a data except Exponential 1 model. It was concluded that the best models are Logistic model for C1 test cell and Gompertz model for C2 test cell.

Present S_{non} content in leachate is very important in the process design of biological leachate treatment to optimize process structure and to estimate effluent residual COD. For this purpose, the change of f_{non} (S_{non}/S_{T0}) is modeled by modified Logistic and Gompertz equations for C1 and C2 test cell, correspondingly.

The obtained Logistic model for C1 test cell and Gompertz model for C2 test cell for change of f_{non} with refuse age are obtained as:

$$f(x)_{C1} = 0.007431 + \frac{0.3772}{1 + \exp \left[\frac{((4 \times 0.4818)/0.3772) \times (-0.4157 - x) + 2}{1} \right]}$$

$$f(x)_{C2} = 0.405 + 0.3901 \times \exp \left[- \exp \left(\frac{0.7158 \exp(1)}{0.3901} \right) (0.4955 - x) + 1 \right],$$

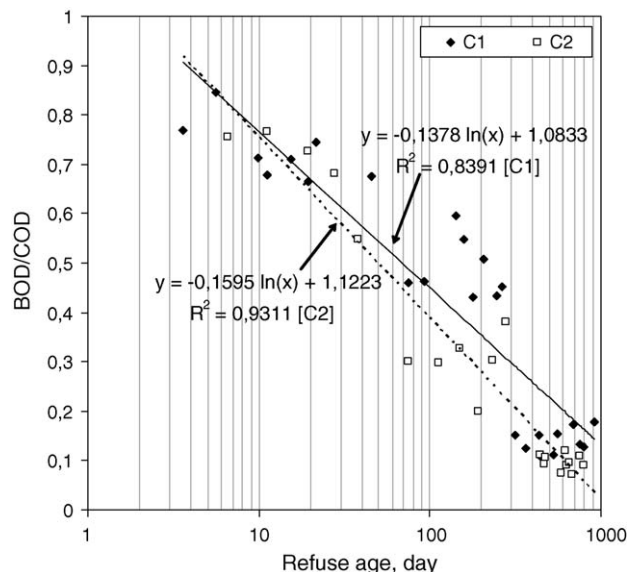


Fig. 7. BOD/COD ratios of leachates from C1 and C2 test cells.

Table 3
Results of evaluating the goodness of obtained fits

Name of model	SSE	R-square	DFE	Adj. R-square	RMSE
C1					
Logistic	0.00318	0.99432	17	0.99331	0.01368
Gompertz	0.00346	0.99382	17	0.99273	0.01426
Richards	0.00442	0.99209	16	0.99012	0.01663
Polynomial	0.00818	0.98537	16	0.98172	0.02262
Cubic	0.01094	0.98040	17	0.97700	0.02537
Linear	0.01745	0.96880	19	0.96720	0.03031
Power	0.05814	0.89610	18	0.88450	0.05683
Exponential 2	0.07908	0.85870	17	0.83370	0.06820
Quadratic	0.10910	0.80500	18	0.78330	0.07785
Exponential 1	0.28470	0.49100	19	0.46420	0.12240
C2					
Gompertz	0.00144	0.99772	17	0.99732	0.00919
Logistic	0.00180	0.99714	17	0.99663	0.01029
Richards	0.00391	0.99380	16	0.99224	0.01563
Cubic	0.01337	0.97880	17	0.97500	0.02805
Polynomial	0.01621	0.97425	16	0.96781	0.03183
Linear	0.04625	0.92650	19	0.92270	0.04934
Power	0.08525	0.86460	18	0.84960	0.06882
Quadratic	0.10910	0.80500	18	0.78330	0.07785
Exponential 1	0.15930	0.74690	19	0.73360	0.09157
Exponential 2	0.23190	0.63160	17	0.56660	0.11680

Table 4
Coefficients of obtained models (with 95% confidence intervals) for change of f_{non} with refuse age

	Logistic model for C1 Coefficients (with 95% confidence intervals)	Gompertz model for C2 Coefficients (with 95% confidence intervals)
L	-0.4157 (-0.4964, -0.335)	0.4955 (0.453, 0.538)
A_0	0.007431 (-0.004717, 0.01958)	0.405 (0.3972, 0.4129)
A_{max}	0.3772 (0.3592, 0.3952)	0.3901 (-0.4007, -0.3794)
M	0.4818 (0.4072, 0.5565)	0.7158 (0.6279, 0.8037)

and their coefficients (with 95% confidence intervals) are given in Table 4.

Fig. 8 shows f_{non} values and fitted curves for C1 and C2 test cells. Good fits are obtained between the calculated data and the model results.

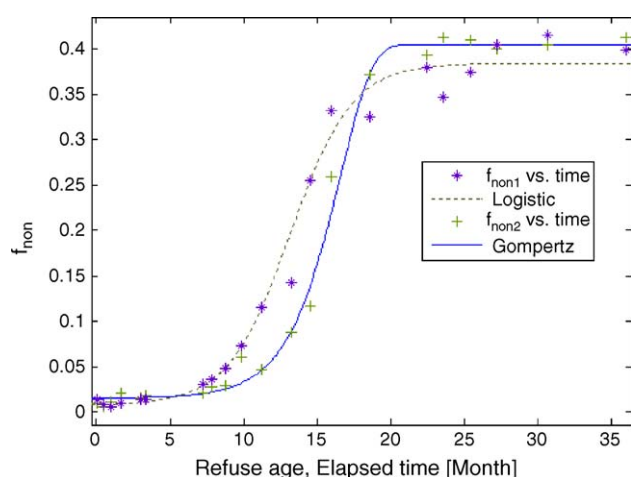


Fig. 8. f_{non} Values and fitted curves for C1 and C2 test cells.

4. Conclusion

Pollutant concentrations of biologically treated leachate may exceed discharge standards due to inappropriate estimation of S_{non} and soluble microbial product. The biodegradable fraction of substrate can be effectively removed in biological treatment system, but its non-biodegradable fraction passes through the system unchanged. Besides this fraction, the soluble microbial products, which are resistant to biological degradation, may be produced by microorganisms within the treatment systems. Furthermore, non-biodegradable compounds of leachate gradually increases as a landfill stabilizes, therefore, the change of leachate composition with respect to refuse age should be carefully considered in the design of leachate treatment plant. Hence, the knowledge on the change of non-biodegradable soluble inert COD (S_{non}) with refuse age may help operation engineer to optimize operational conditions to cope with the changed leachate composition over time. In this context, this study presents the change of non-biodegradable COD with refuse age in leachates originated from field scale test cells with (C2) and without (C1) recirculation. For leachates of both cells, f_{non} increased from its initial value of 0.01 to around 0.1 after 300 days of operation. Due to the development of anaerobic conditions, the value

showed an increasing trend and the maximum value of 0.4 was reached on day 600. Also, experimental data were fitted to several models in the literature and it was observed that Logistic model for C1 test cell and Gompertz model for C2 test cell gave the best fits to the experimental data.

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