LONG-TERM MONITORING AND PREDICTION FOR LEACHATE CONCENTRATIONS IN SHANGHAI REFUSE LANDFILL

ZHAO YOUCAI[1*], LIU JIANGGYING[1], HUANG RENHUA[2] and GU GUOWEI[1]
[1] *National Laboratory of Pollution Control and Resource Reuse, School of Environmental Engineering, Tongji University, Shanghai 200092, China;* [2] *Shanghai Waste Treatment Co.*
(author for correspondence, e-mail: zyclmk@online, sh.cn)*

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Abstract. Parameters such as pH, concentrations of Cl^-, turbidity, NH_3-N, COD_{Cr} and BOD_5 (simplified as COD and BOD respectively in the following text) in leachate generated in the large-scale testing landfill unit and closed landfill units at Shanghai Refuse Landfill were monitored from April 1995 to October 1998. The mathematical simulation formula between these parameters and refuse age were established based on the data obtained from the testing landfill unit and justified by the data obtained at the closed landfill units from 1989 to 1993. The long-term predictions for the leachate concentrations for the Landfill were made using the mathematical simulation formula established. It was predicted that the COD and BOD may be reached to the strictest standard for pollution control on municipal solid wastes landfill in China, i.e., COD < 100 mg L^{-1} and BOD < 30 mg L^{-1}, after 15 yr natural attenuation. The time predicted for NH_3-N concentrations to reach the discharging standard, 15 mg L^{-1}, was found to be at least 23–26 yr or even longer. The predictions for concentrations of Cl^-, turbidity, and pH values in leachate are also given. The natural attenuation of Cl^- is the slowest and might be decreased to 200 mg Cl^- L^{-1}, the agricultural irritation standard, after at least 58 yr.

Keywords: landfill, leachate, monitoring, MSW, prediction, refuse

1. Introduction

It is well known that refuse (municipal solid waste) landfill could be viewed as a large-scale bioreactor as the refuse filled would undergo complex and slow physical, chemical and biological changes in a landfill. As a result, the refuse will be degraded, landfill gases generated, refuse surface settled down, and concentrations in leachate attenuated slowly and gradually (Bookter *et al*., 1982; Dean *et al*., Ham *et al*., 1982; Tounsend *et al*., 1996).

The treatment of leachate is one of the most important issues in the management of a landfill. Conventionally, the leachate is pumped out from the landfill units and led into the anaerobic or aerobic treatment plants. The processes for leachate treatment are always very complex and costs are usually quite high (Zhao *et al*., 1999).

In fact, it has been proved that the concentrations and biodegradability of leachate are always decreased, as the refuse age is increased (Wang Luochun, 1999; Zhu

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et al., 1996). In the most landfills in the world, leachate is collected and mixed together from all the landfill units, regardless of the refuse age and leachate concentrations. Hence, the leachate with lower concentrations and biodegradability from the landfill units with longer refuse age may be mixed with the leachate from the landfill units with shorter refuse age. This practice is not considered to be economically and technically reasonable though it may guarantee the stability of leachate quality so that the treatment plants can be operated smoothly as designed.

According to our experiences, the leachate from landfill units with different refuse age should be collected and treated separately in order to guarantee the leachate concentrations to reach the discharging standard with the minimum costs. In addition, the concentrations of leachate may be reduced naturally to the discharging standard after long-term biodegradation in the closed landfill. Such leachate should not be led to the treatment plants but directly discharged into water receivers (Zhao *et al*., 1999).

Therefore, the dependence of leachate concentrations on refuse age for the given landfill should be explored so that the reliable and accurate data can be provided for the optimization of leachate trreatment processes. In this work, a large scale testing landfill unit was constructed at Shanghai Refuse Landfill in May 1995. The leachate was monitored until October 1998. Meanwhile, the leachate generated in the landfill units closed in 1989 to 1994 was also collected and analyzed. The experimental results were reported in this paper.

2. Experimental

2.1. Construction of the Testing Landfill Unit

A large testing landfill units with double layers was constructed at Shanghai Refuse Lanfill. A total weight of 10180 tones of refuse was placed during a period from 4 to 18 April 1995. The refuse height was 4 m, the same height as all the other landfill units at the Landfill, and the filing area was 3000 m^2 (50×60 m). The collection pipes for leachate were installed. As a result, the average density of refuse in the testing landfill was around 0.85 ton/m^3. The contents of refuse were determined to be 40% moisture, 12% plastic films, bags, bottles and other products and 24% organic matters such as trees, woods, cooking wastes, and 24% of inorganic matters such as stone, sand, coal ashes, glass bottles. Plastic film with high strength, which had been previously tested and was proved to be effective, was installed as liners for the bottom and walls. The results of long-term monitoring showed that the liners worked well. The leachate was drained out of the underlying pipes which was integrated into one pipe directed to the collection well. The leachate samples were taken from this well. The concentrations of leachate were analyzed by conventional methods.

2.2. Collection and analysis of leachate in the closed landfill units

The leachate generated in the closed landfill units of 1989–1993 was collected from the collection wells and analysed in October 1998. The refuse at Shanghai Refuse Landfill is placed by area method and the units are separated by clay wall. The leachate from different units is not likely to be mixed together. The area of each unit is around 10 hectares.

All the leachate samples determinations were triplicated and the data given in the tables and figures were the average values. In general, all the analysis in this work are very common and the accuracies are rather high with an acceptable error.

3. Mathematical Simulation Method

For a function

$$y = f(t) \tag{1}$$

it can be established by M numbers of known values (t_i, y_i), i = 1, 2 ... M. Assume the function contains R numbers of unknown constants a_1, a_2, ..., a_R. When the M numbers of known values are used for the solutions of R numbers of unknown constants, i.e.,

$$y_i = f(t_i) \tag{2}$$

if M = R, then all the unknown constants can be solved; if M < R, then the unknown constants are unavailable, and the function can not be established. In the most cases, M is more than R (M > R). The problem is how to get the most probable value of y using all the known values.

Assume the function can be expressed as:

$$y = a\,(b^t) \tag{3}$$

which will be used in this paper, then it can be solved by the least square method using the known values avaialable (t_1, y_1), (t_2, y_2), $\cdots$, (t_M, y_M). The values of a and b can be obtained by the following formulas:

$$\sum_{i=1}^{M} \log(y_i) = M\log(a) + \sum_{i=1}^{M} t_i \log(b)$$

$$\sum_{i=1}^{M} t_i \log(y_i) = \sum_{i=1}^{M} t_i \log(a) + \sum_{i=1}^{M} t_i^2 \log(b)$$

$$a = \exp\left[\frac{\sum_{i=1}^{M}\log(y_i)\sum_{i=1}^{M}t_i^2 - \sum_{i=1}^{M}t_i\sum_{i=1}^{M}t_i\log(y_i)}{M\sum_{i=1}^{M}t_i^2 - \left(\sum_{i=1}^{M}t_i\right)^2}\right]$$

$$b = \exp\left[\frac{M\sum_{i=1}^{M}t_i\log(y_i) - \sum_{i=1}^{M}t_i\sum_{i=1}^{M}\log(y_i)}{M\sum_{i=1}^{M}t_i^2 - \left(\sum_{i=1}^{M}t_i\right)^2}\right]$$

and the correlation coefficients, r, can be derived as follows:

$$r = \frac{lty}{\sqrt{ltt - lyy}}$$

$$lty = \sum_{i=1}^{M}(t_i - \bar{t})(y_i - \bar{y}) = \sum_{i=1}^{M}t_i y_i - \frac{1}{M}\left(\sum_{i=1}^{M}t_i\right)\left(\sum_{i=1}^{M}y_i\right)$$

$$ltt = \sum_{i=1}^{M}t_i^2 - \frac{\left(\sum_{i=1}^{M}t_i\right)^2}{M}$$

$$lyy = \sum_{i=1}^{M}y_i^2 - \frac{\left(\sum_{i=1}^{M}y_i\right)^2}{M}$$

4. Results

4.1. Monitoring of concentrations in the leachate in the testing landfill unit

The leachate was collected from the collection well of the testing landfill unit for every one to two weeks from May 1995 to October 1998 and the parameters characterizing the leachate such as pH, Cl^-, turbidity, NH_3-N, COD and BOD concentrations were determined. Totally, 64 samples were collected and analysed in this period. The relationships between these parameters and refuse age (days) are shown in Figures 1–7 respectively.

The pH values in the monitoring period of 3.5 yr were found to be at the range of 6.2 to 8.5 and fluctuated with the refuse age (Figure 1). The relationships between pH values and the refuse age may be roughly categorized into three phases. In the first phase (0–1.5 months), pH values were decreased from 7.3 to the lowest value of 6.2, which may indicate that the refuse was undergoing aerobic biodegration;

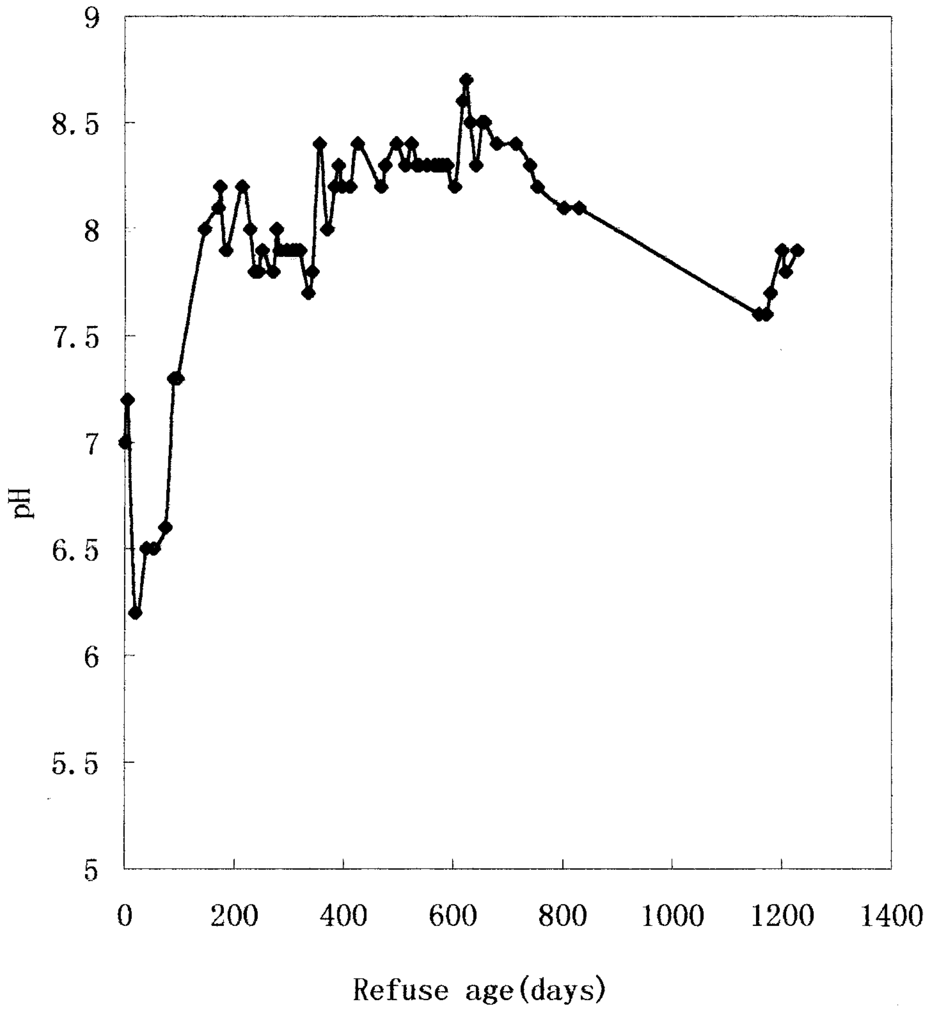

Figure 1. Relationship between the pH value of leachate and the refuse age (days).

and then the pH values were increased to 8.1–8.4 in the following 4 months, which may be classified as the second phase, where the facultative biodegradation may be predominated. In the initial stage of the third phase (5–23 months), the pH values were kept at around 8, and the refuse just fell into the anaerobic biodegradation and methane was generated. Finally, the refuse entered into the second stage of the third phase with strict anaerobic biodegradation (after 23 months), in which the pH values were found to be lower than 8.

The concentrations of chloride ions in the leachate was quite high when the refuse was filled, and then decreased rapidly to around 2700 mg L^{-1} in 70 days.

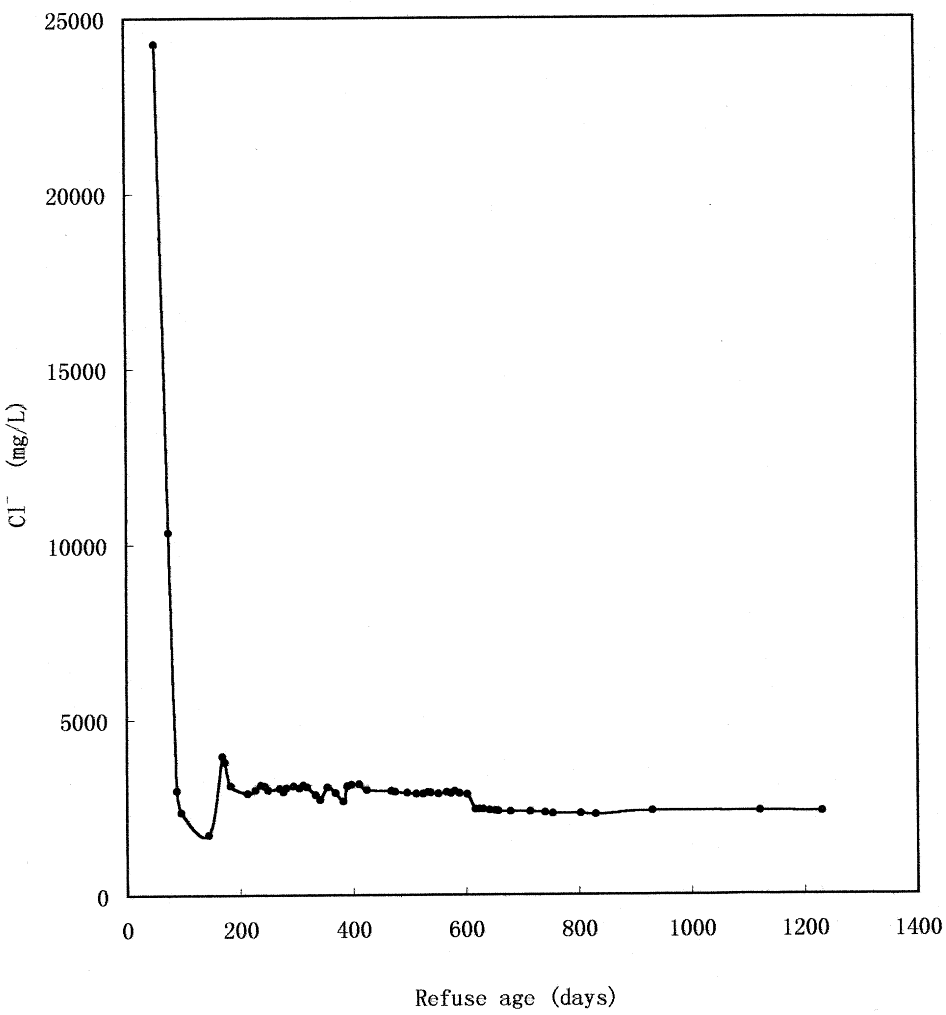

Figure 2. Relationship between the Cl^- concentrations (mg L^{-1}) in leachate and the refuse age (days).

The further decrease of Cl^- was found to be very slow, and the value was around 2400 mg L^{-1} at 3.5 yr (Figure 2).

The relationships between the turbidity of leachate and refuse age may be also categorized into three phases (Figure 3). In the first phase, it increased rapidly to the maximum value of 2153.06 units (around at 90th days), and then decreased also sharply at the second phase (100–400 days, with the value of 483.38 at 400 days) as the refuse age increased. After around 400 days, the decrease trends of turbidity became slowly, with a value of 390 units at 1200 days.

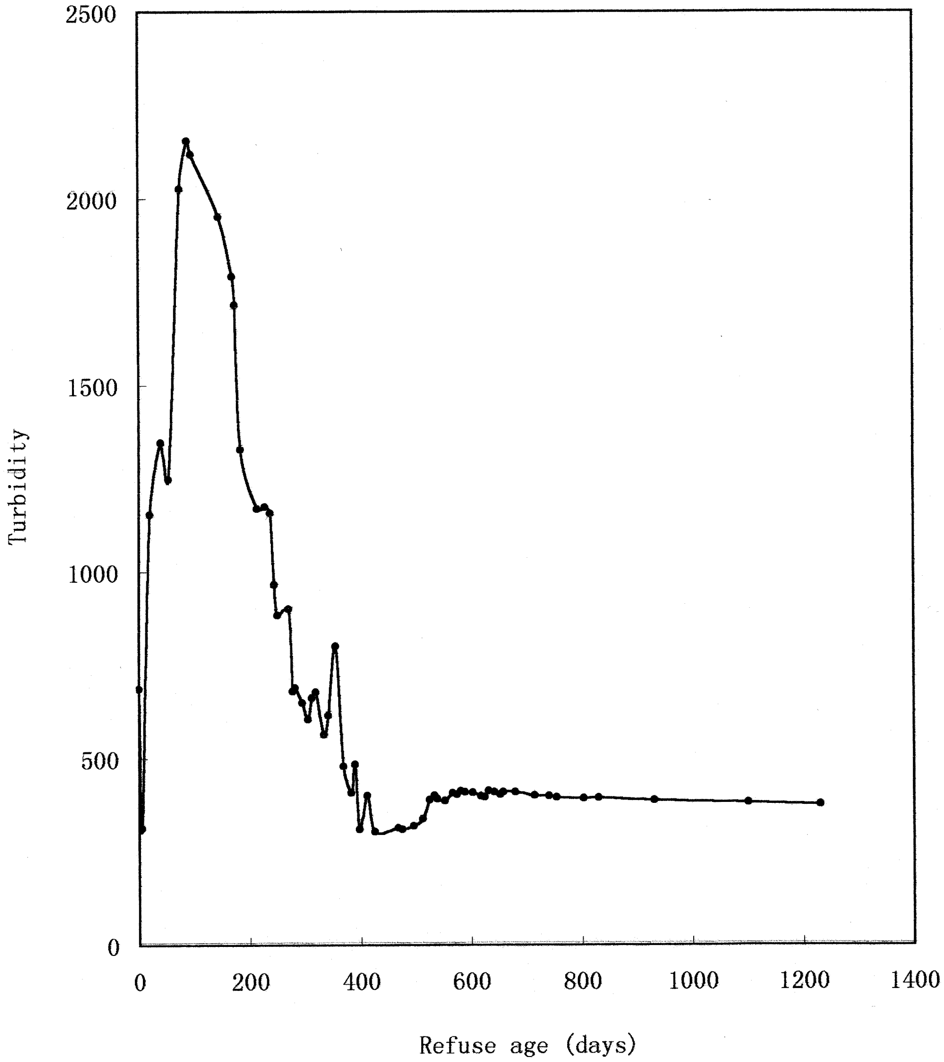

Figure 3. Relationship between the turbidity of leachate and the refuse age (days).

The 'three phases' categorization method can be also used for the cases of NH_3-N concentrations in the leachate (Figure 4). At the first phase (0–100 days), the NH_3-N concentrations increased rapidly to the highest value of 3900 mg L^{-1}. In the second phase (100-450 days), the concentration fluctuated with the refuse age, with an average value of around 3,000–3,500 mg L^{-1} (the highest value of 4,4457.98 mg L^{-1}). The concentrations were decreased sharply from 4,400 mg L^{-1} to lower than 500 mg L^{-1} at the initial stage of the third phase (450–550 days), and then decreased gradually after 450 days to 457.63 mg L^{-1} in 1220 days when the degradation is entered into the second stage of the third phase.

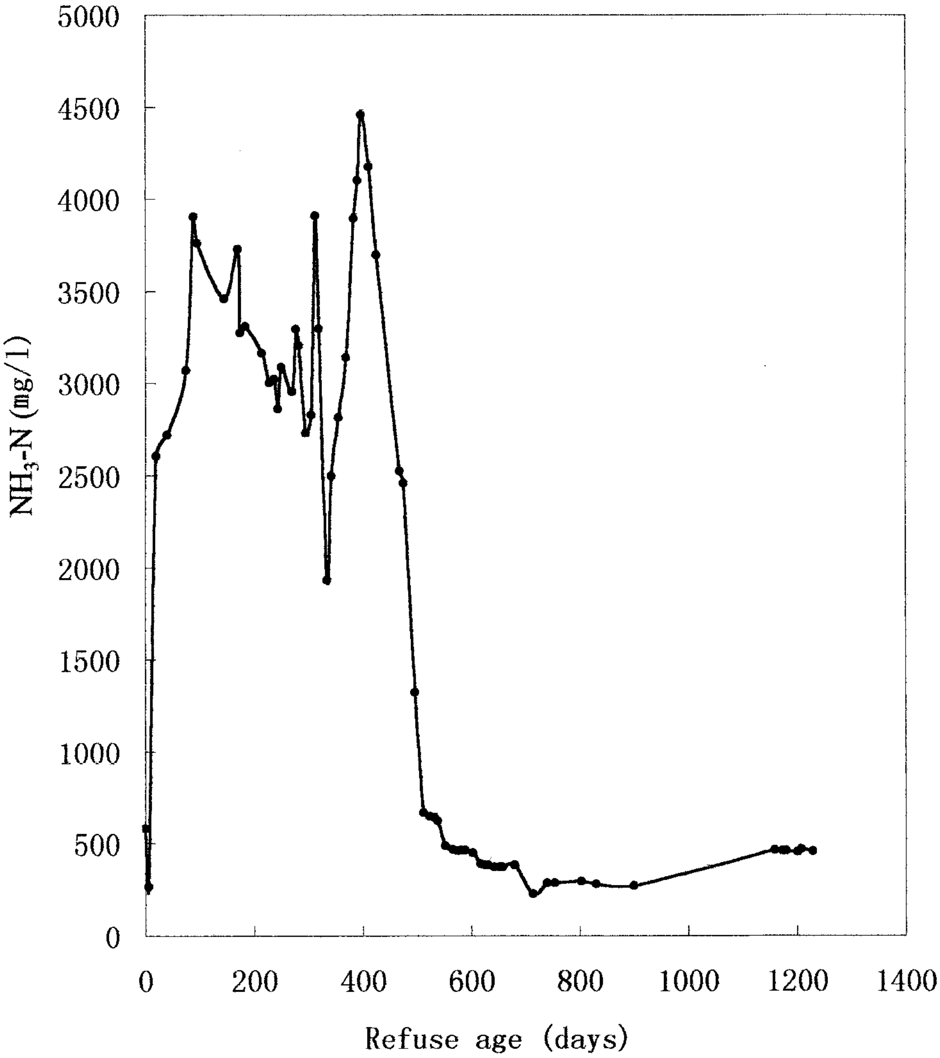

Figure 4. Relationship between NH_3-N concentrations (mg L^{-1}) in leachate and the refuse age (days).

The dependencies of COD and BOD on the refuse age are shown in Figures 5 and 6. It can be seen from Figure 5 that COD concentrations in the leachate is rapidly reached to the maximum value of 54889.6 mg L^{-1} in 45 days, and then decreased sharply to around 8200 mg L^{-1} in 90 days. Since then, the COD concentration decreased gradually as the increase of refuse age. COD was found to be around 1350 mg L^{-1} after 40 months when the monitoring was terminated.

The similar phenomenon can be found for the relationships between BOD concentrations and refuse age as shown in Figure 6. The BOD was rapidly increased to

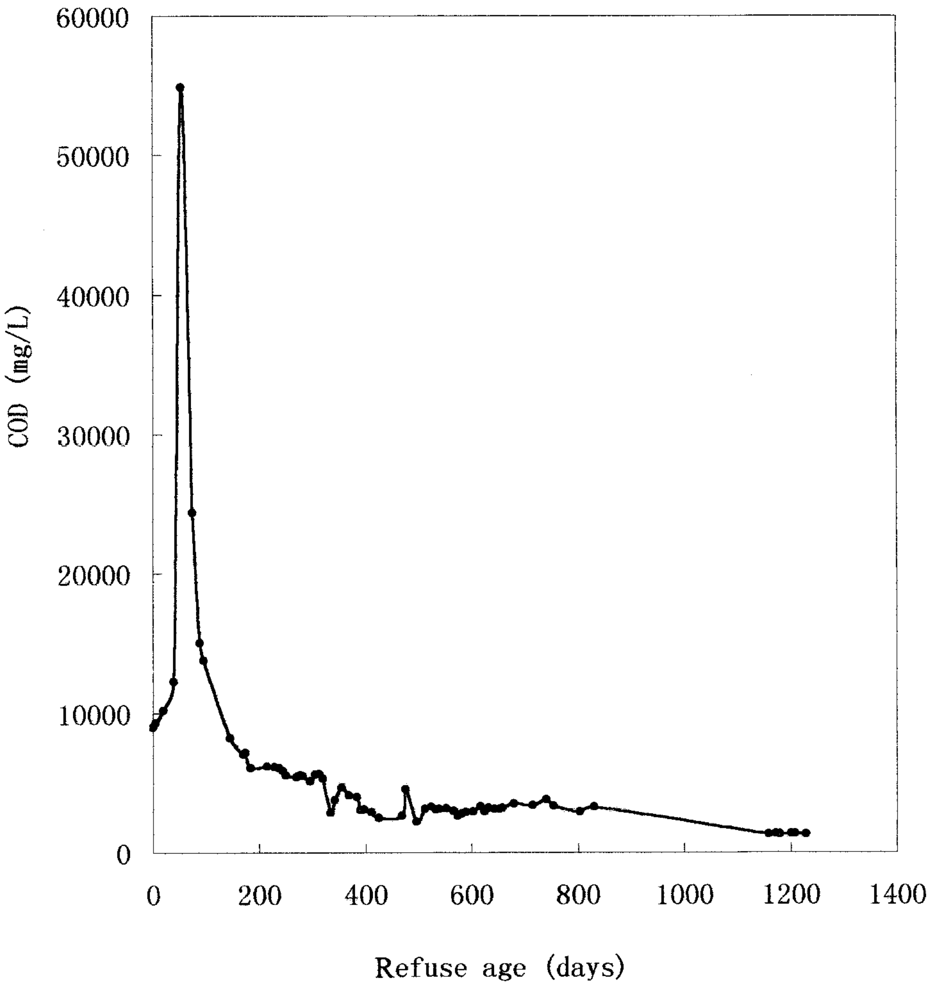

Figure 5. Relationship between the COD concentrations of leachate (mg L^{-1}) and the refuse age (days).

the maximum value of 15949.12 mg L^{-1} in 45 days, and then decreased to around 2500 mg L^{-1} in 90 days and to 335.8 mg L^{-1} in 40 months.

The ratios of BOD and COD are shown in Figure 7. It can be seen that the ratios fluctuated with the refuse age. The highest value may be over 0.55 and the lowest may be below 0.14. In general, the ratios fell into the level of good biodegradability before 400 days with ratios over 0.45, and then decreased sharply and fell into the level of poor biodegradability with the ratios below 0.45.

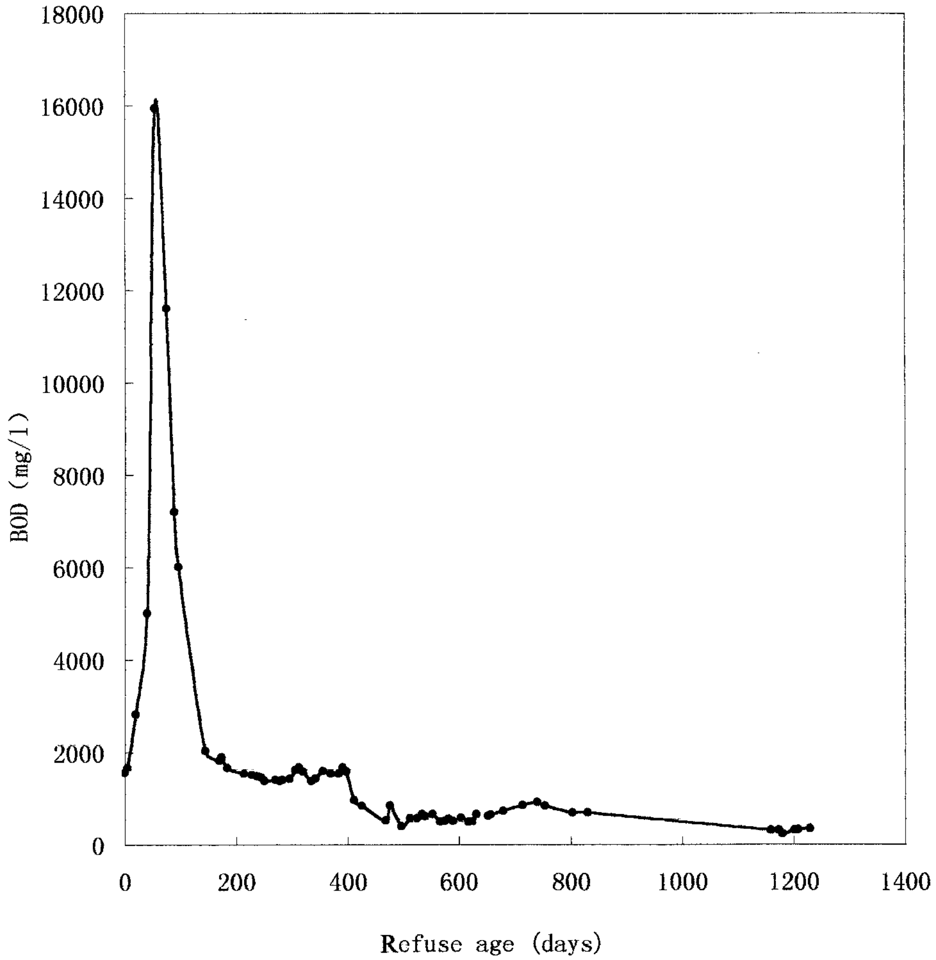

Figure 6. Relationship between the BOD concentrations (mg L^{-1}) of leachate and the refuse age (days).

4.2. MATHEMATICAL SIMULATION FOR THE RELATIONSHIPS BETWEEN LEACHATE CONCENTRATIONS AND REFUSE AGE IN THE TESTING LANDFILL

It is attempted to understand the long-term attenuation effects of all the parameters in the leachate except for pH at Shanghai Refuse Landfill so that the treatment of leachate can be conducted more effectively. Therefore, the data of Figures 2–6 should be simulated and used for the long-term prediction of the corresponding parameters.

The physical, chemical and biological changes taken place within the landfill is very complex. It seems not easy to propose a reasonable mathematical model to

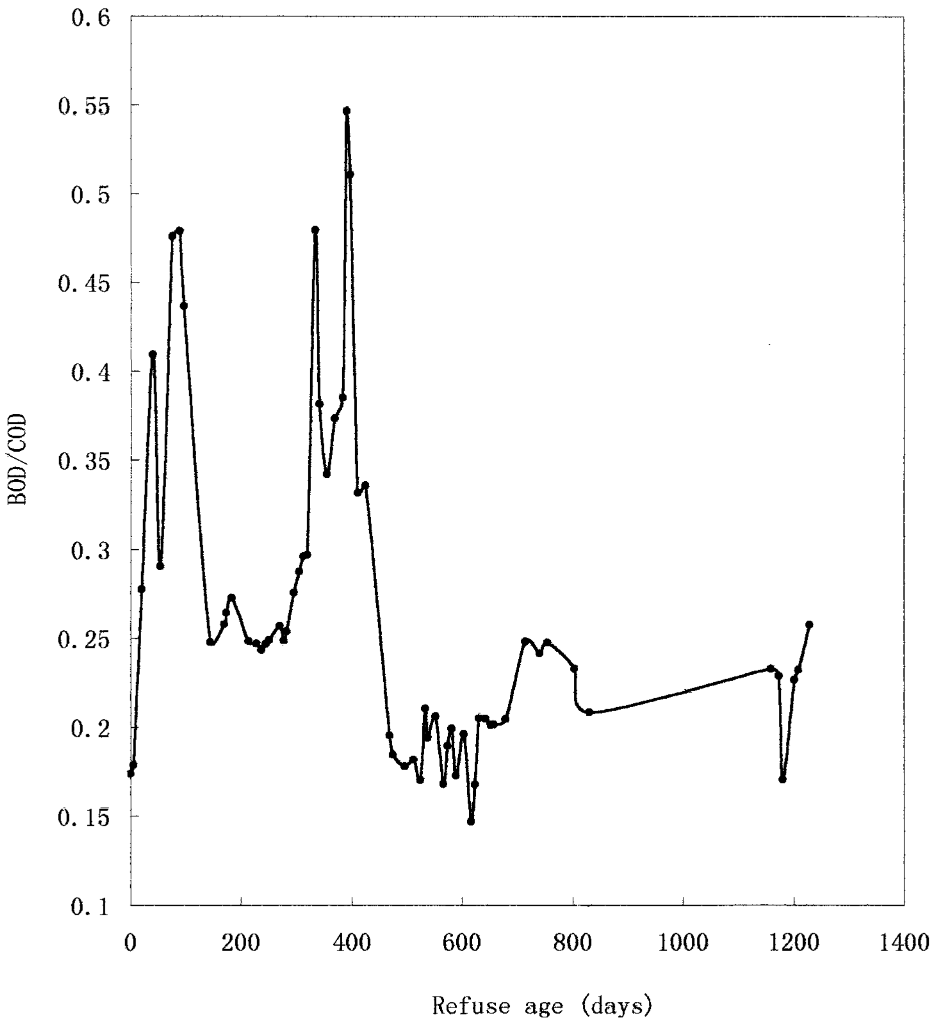

Figure 7. Ratios of BOD/COD in the leachate.

match the real process of degradation in the landfill. Nevertheless, it can be considered that the biological processes should be predominant after the degradation fall into the initial stage of the third phase. As a result, first-rate reaction may be proposed for the dissolution of pollutants from the refuse in landfill, i.e.

$$C = C_0 \times A^t \tag{4}$$

Where C is Cl^-, turbidity, NH_3-N, COD or BOD in the leachate (units for turbidity and mg L^{-1} for the other parameters) as the time of t (refuse age, day), and A the attenuation coefficient, C_0 the starting concentration at t_0. Equation (4) was

TABLE I

Summary of mathematical simulation for Cl^-, turbidity, NH_3-N, COD, BOD

Parameters	Mathematical simulation formula	Range of refuse age, t (days)	Correlation coefficients, |r|
Cl^- (mg L^{-1})	$Cl^- = 3132.90 \times 0.99987^t$	t > 184	0.8328
Turbidity	$T = 490.87 \times 0.999358^t$	t > 410	0.9995
NH_3-N (mg L^{-1})	$NH_3\text{-N} = 824.03 \times 0.999528^t$	t > 500	0.8886
COD (mg L^{-1})	$COD = 9104.88 \times 0.99844^t$	145 < t > 1227	0.9665
	$COD = 1340.00 \times 0.99936^{t-A}$	A = 1168, t > 1227	0.9712
BOD (mg L^{-1})	$BOD = 2441.46 \times 0.998479^t$	145 < t > 1227	0.9521
	$BOD = 490.00 \times 0.99913^{t-A}$	A = 1168, t > 1227	0.9348

solved using the mathematical simulation method above-mentioned by taking the expression

$$y = a(b^t)$$

as

$$C = C_0 \times A^t$$

while the value of a was assumed to be the constant as C_0 selected from Figures 2–6.

Take the relationships between the chloride ion concentrations in the leachate and refuse age as an example (Figure 2). It can be seen that the turbidity decreased sharply before the refuse age was less than around 184 days; nevertheless, such a decrease becomes very slow after 184 days. Hence, the mathematical simulation formula can be established easily between chloride concentrations and refuse age after 184 days. Take t_0 = 184 days, where the chloride ion concentration, Cl^- (C_0), is 3132.90 mg L^{-1}, then the mathematical simulation formula can be expressed as:

$$Cl^- = 3132.90 \times 0.99987^t, \; r = 0.8328.$$

Similarly, the mathematical simulation formulas of other parameter can be established based on the data shown in Figures 3–6, and the results are summarized in Table I.

TABLE II

NH_3-N concentrations in the leachate collected from the closed landfill units of 1989–1993 and the comparisons between the simulated and monitoring results

Refuse age (years)	1.5	2	3.3	5	6	7	8	9
Simulated (mg L^{-1})	636.33	583.80	461.27	348.14	293.03	246.65	207.61	175.00
Monitoring (mg L^{-1})	621.03	383.27	457.63	437.24	284.56	206.81	180.22	160.09
Error (%)	2.4	52.3	0.8	–20.4	3.0	19.2	15.2	9.3

Note: The data of 1–3.3 yr are taken from the monitoring results of the testing landfill unit, and the data of 5–9 yr taken from the closed landfill units of 1989–1993. The errors are calculated based on the monitoring data.

4.3. Monitoring of leachate in the closed landfill units from 1989 to 1993 and justification of the validity of the mathematical simulation formula

The leachate in the closed landfill units of 1989, 1990, 1991, 1992, and 1993 were collected and analysed from October to December 1998. These units were used and closed in April or May of the corresponding years, the same month as the testing landfill units. The refuse composition in these closed units could be also proposed to be the same as that in the testing landfill unit according to the record of the Landfill Office. More than 5 holes were dug for each unit and the leachate were collected from these holes. At least 3 samples were collected and analysed for each hole for every two weeks in the monitoring duration of 3 months, and the data were averaged. Tables II–IV showed the results of NH_3-N, COD and BOD and their comparisons with the simulated values calculated from formula shown in Table I.

Obviously, the simulated and monitoring data were in good agreement, and the errors seem to be acceptable. Hence, the mathematical simulation formula established in this work can be considered to be reliable and accurate, and may be used for the long-term prediction of corresponding parameters in the leachate of Shanghai Refuse Landfill. This conclusion can be extended to the cases of chloride ion and tubidity.

4.4. Long-term predictions of the natural attunuation of concentrations in the leachate at Shanghai Refuse Landfill

Based on Equations given in Table I, it can be easily calculated the values of every parameters at any refuse age (in terms of years). The results are shown in Tables V–IX. The natural attenuation of Cl^- seems to be very slow (Table V). It at least needs

TABLE III

COD concentrations in the leachate collected from the closed landfill units of 1989–1993 and the comparisons between the simulated and monitoring results

Refuse age (years)	1	2	2.2	3.3	5	6	7	8	9
Simulated (mg L^{-1})	5149.8	2912.8	2599.1	1388.0	943.0	646.9	525.2	468.0	330.0
Monitoring (mg L^{-1})	4106.8	3405.6	2939.1	1358.3	1012.7	1007.9	316.9	390.8	300.0
Error (%)	25.4	–14.5	11.6	2.2	–6.9	–25.9	66.0	19.8	10.0

Note: The data of 1–3.3 yr are taken from the monitoring results of the testing landfill unit, and the data of 5–9 yr taken from the closed landfill units of 1989–1993. The errors are calculated based on the monitoring data.

TABLE IV

BOD concentrations in the leachate collected from the closed landfill units of 1989–1993 and the comparisons between the simulated and monitoring results

Refuse age (years)	1	2	2.2	3.3	5	6	7	8	9
Simulated (mg L^{-1})	1401.33	803.99	719.44	454.95	292.83	225.93	174.33	134.52	103
Monitoring (mg L^{-1})	1534.08	844.25	684.50	335.90	244.64	240.64	139.44	94.24	85.00
Error (%)	–8.6	–4.7	5.1	35.5	19.7	–6.1	25.0	24.7	21.2

Note: The data of 1–3.3 yr are taken from the monitoring results of the testing landfill unit, and the data of 5–9 yr taken from the closed landfill units of 1989–1993. The errors are calculated based on the monitoring data.

58 yr for the Cl^{-} concentration in leachate to reach 200 mg L^{-1}, the concentration regulated for the agricultural irrigation water.

The turbidity is found to be decreased slowly and the values may be reduced naturally to 18 after 15 yr (Table VI). The water would be quite good at this level of turbidity.

The prediction of NH_3-N decrease with the refuse age is given in Table VII. It can be seen that the NH_3-N concentration in the leachate at Shanghai Refuse Landfill would be decreased to the discharging standard of 15 mg L^{-1} after at least 24 yr, which is much longer than that needed for the COD and BOD concentration attenuation.

The current strictest discharging standards for the leachate in China is COD < 100 mg L^{-1} and BOD < 30 mg L^{-1}, which seems to be also the universal

TABLE V

The prediction of natural attenuation of Cl^- concentration in the leachate

t (years)	4	5	6	7	8	9	10	11	12
Cl^- (mg L^{-1})	2591	2471	2357	2247	2143	2043	1949	1859	1773
t (years)	13	14	15	16	17	18	19	20	21
Cl^- (mg L^{-1})	1691	1612	1537	1466	1398	1333	1272	1213	1157

TABLE VI

The prediction of natural attenuation of turbidity in the leachate

t (years)	4	5	6	7	8	9	10	11	12	13	14	15
Turbidity	243	192	152	120	95	75	60	47	37	29	23	18

TABLE VII

The prediction of natural attenuation of NH_3-N in the leachate

t (year)	4	5	6	7	8	9	10	11	12	13
NH_3-N (mg L^{-1})	414	348	293	269	208	175	147	124	104	88
t (year)	14	15	16	17	18	19	20	21	23	24
NH_3-N (mg L^{-1})	74	62	52	44	37	31	26	22	16	13

TABLE VIII

The prediction of the natural attenuation of COD in the leachate

t (years)	4	5	6	7	8	9	10	11	12
COD (mg L^{-1})	1112	943	747	526	468	330	261	207	164
t (years)	13	14	15	16	17	18	19	20	
COD (mg L^{-1})	129	103	81	64	51	40	32	25	

standard for all the wastewaters discharging in the most of countries worldwide. From Tables VIII and IX, it can be seen that such a discharging standard may be possible to reach after around 15 yr of natural attenuation. It is interesting to find that the attenuation time for COD to reach 100 mg L^{-1} is almost the same time for BOD to reach 30 mg L^{-1}. Hence, it is universally considered that, when the COD concentration in the leachate could reach the discharging standard of 100 mg L^{-1}, then the corresponding BOD concentration could also reach the standard of 30 mg L^{-1}, and vice versa.

TABLE IX
The prediction of the natural attenuation of BOD in the leachate

t (years)	4	5	6	7	8	9	10	11	12	13	14	15
BOD (mg L^{-1})	379	293	226	174	135	103	80	62	48	37	28	22

5. Discussions

According to our experiences, NH_3-N in the leachate is the pollutant that is very difficult to be removed. COD, BOD and SS may be readily reduced to the discharging standard after treatment. However, in most cases NH_3-N concentration may usually be quite high and further measures should be taken in order to reduce it to the acceptable level. In this work, it was observed that the concentration of NH_3-N decreased to the lowest in about 700 days and then increased slightly. The mathematical simulation formula of NH_3-N has been in fact averaged within the testing period. Hence, it may be predicted that the NH_3-N concentration attenuation in leachate may be lower than that given in Table VII. Nevertheless, it is sure that the NH_3-N concentration will be decreased as the refuse age is increased. More accurate prediction for NH_3-N concentration attenuation in leachate is likely when a longer period of testing is conducted and more data are available.

Based on the predictions mentioned above, COD and BOD concentrations in leachate at Shanghai Refuse Landfill would be reduced naturally to the discharging standard and no treatment process seems to be necessary for these two parameters after around 15 yr. However, NH_3-N concentration would be still very high and should be removed before discharging into the water receivers. Hence, it may be proposed that only processes for the removal of NH_3-N from leachate is required after 15 yr natural attentuation. This is an important implication for the guidelines for the treatment of leachate. Based on these findings, the leachate should be collected separately from the closed landfill units with different refuse age and treated respectively, which would facilitate the treatment and reduce the costs.

Shanghai Refuse Landfill is the largest landfill in China. It occupies around 6 km^2. It was constructed along the shore of East China Sea, which was formed by the sedimentation of sand carried by Yangtze River. The refuse is filled by area method, with a filling height of 4 m. The lanfdill is separated into many landfill units by the clay dug in situ, with 10 hectares for each unit. Leachate is one of the wastewaters that are difficult to be treated biologically. Hence, the discharging standard in China has been regulated to COD < 300 mg L^{-1}, BOD < 150 mg L^{-1}, NH_3-N < 25 mg L^{-1}, SS < 200 Mg L^{-1} in 1998. In this case, according to the research results in this work, the leachate generated in the landfill units closed in 1989 and 1990 could be discharged into the water receivers (East China Sea) when the NH_3-N concentration is reduced to lower than 25 mg L^{-1} by chemical processes.

6. Conclusions

A large testing landfill was constructed in situ at Shanghai Refuse Landfill. The leachate generated in the testing landfill was collected and analysed. The corresponding mathematical simulation formula for Cl^-, turbidity, NH_3-N, COD_{Cr} and BOD_5 were established and their validity was justified by the monitoring data of the closed landfill units from 1989 to 1993. The formulas were used for the long-term prediction of these parameters in leachate and the implications and possible practices were discussed.

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