Available online at www.sciencedirect.com
ScienceDirect
Journal of Hazardous Materials 147 (2007) 257–264
Journal of Hazardous Materials
www.elsevier.com/locate/jhazmat

Size-fractionation and characterization of refuse landfill leachate by sequential filtration using membranes with varied porosity

Lou Ziyang, Zhao Youcai *

The State Key Laboratory of Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Tongji University, Shanghai 200092, China

Received 29 August 2006; received in revised form 29 December 2006; accepted 29 December 2006
Available online 13 January 2007

Abstract

Fresh leachate and effluents samples were collected from the holding tank, anaerobic, anoxic, and aerobic lagoons at Shanghai Laogang Refuse Landfill, the largest landfill in China with a placement scale of 9000 t refuse per day. To characterize the difference in leachate along the treatment processes, especially the information about size distribution of colloids in those leachate, the organic matters were size-fractioned into suspended particles (SP, >1.2 μm), coarse colloids (CC, 1.2–0.45 μm), fine colloids (FC, 0.45 μm to 1 kDa MW, 1 Da = 1/16 O atomic mass unit), and dissolved organic matters (DM, <1 kDa MW) using micro-filtration and ultra-filtration membranes in order. The parameters, such as COD (chemical oxygen demand), TOC (total organic carbon), TS (total solid), pH, TP (total phosphate), TN (total nitrogen), FS (fixed solid), NH_4^+, IC (inorganic carbon), TC (total carbon), color, turbidity and conductivity in the filtrates resulting from sequential filtration of leachate, were then determined, and quantitative relationships between these parameters and the membrane molecular sizes used were established. Typically, the total removal of COD, NH_4^+, conductivity and P were found to be 75%, 75%, 42% and 85%, respectively, after the biological treatment processes used at Laogang Refuse Landfill. Dissolved fractions were predominant in fresh leachate and in effluents from treatment processes in terms of TOC with a content of over 47%. The molecular weight (MW) percentage distribution in leachate varied as the leachate was treated in the biological treatment stages. The percentages of TOC of fine colloid fractions increased from 6% to 38% while those of dissolved fractions decreased from 78% to 47%. TN in leachate also predominated in the dissolved fraction, occupying over 58%, while those TP in leachate were combined with the SS and CC fractions. The ratios of *ortho*-phosphate/TP and NH_4^+/TN in leachate and effluents were over 50% and 80%, respectively.

Keywords: Refuse landfill leachate; Membrane; Colloid; Size-fractionation; Biological treatment process

1. Introduction

Landfill is still one of the main predominant means for refuse disposal in the world, especially in the developing countries, and leachate, as one of the most important by-products in landfill, is a complex kind of waste water with considerable variations in both composition and volumetric flows [1–3]. The composition and concentration of contaminants in leachate are dependent on hydro-geological factors of the landfill, types of wastes deposited, climate of local place, and age of the waste placed in the landfill. Therefore, the leachate treatment processes used in a landfill should be able to cope with the changing quality and quantity of the leachate [4–6].

Integrated processes are usually used for leachate treatment [7,8], and most of them are derived from or referred to those used in municipal or industrial wastewater treatment processes, which are more or less modified while applied, while ignoring leachate's specific characteristics. Therefore, the actual widely used treatment units have been proved ineffective, as observed in practice, even if the biological and chemical processes are used together [9]. It has been well proved that COD and BOD in leachate may be biologically reduced to around 500–800 and 200–300 mg/L, respectively. Nevertheless further reduction would be very difficult, even with the aid of chemical or physical means, regardless of fresh leachate from operating landfill compartments or aged leachate from closed landfill compartments, as both of them are composed of some refractory materials, such as humus-like materials.

Abbreviations: SP, suspended particle; CC, coarse colloid; FC, fine colloid; DM, dissolved matter; TS, total solid; FS, fixed solid; TN, total nitrogen; TP, total phosphate

* Corresponding author. Tel.: +86 21 65982684; fax: +86 21 65980041.
E-mail address: zhaoyoucai@mail.tongji.edu.cn (Z. Youcai).

0304-3894/$ – see front matter
doi:10.1016/j.jhazmat.2006.12.084

In recent years, many new treatment methods for landfill leachate have been developed [7,10,11,25,26], among which reverse osmosis (RO) technology that showed its advantages in removing dissolved matters [27]. However, RO may be considered to be too expensive to be used in most developing countries due to the high energy consumption, the large operational costs and the severe membrane fouling, because there are higher strength concentration leachates produced due to the high content of food waste in refuse in China, comparing to leachate produced in west countries [9]. Therefore, the development of a cost-effective and highly efficient leachate treatment technology is still a challenging issue in the field of landfill management.

Though efforts have been made for the development of leachate treatment technologies, little knowledge on leachate is obtained due to its complex, including about its microstructures, colloidal particles, chemical properties, etc. Actually, the leachate treatment processes are being developed based on the blackbox theory. Indeed, leachate may be traditionally considered from a macro perspective and regarded as a whole [1,4,12,13], as expressed by COD, BOD, ammonia-nitrogen, TOC, etc. On the other hands, leachate was also considered from a micro-perspective. The chemical composition in leachate can then be characterized using GC–MS, LC–MS to obtain the concentrations of well-defined chemicals such as fatty acids, benzene, etc. with the aid of the recent literature about MS [23,24,28]. The disadvantage of these methods is that it is not representative of the real total content of COD in leachate, as it measures a COD equivalent to10–20 mg/L [2,5,14,15], in comparison with the real content equivalent to 10,000–15,000 mg/L COD in fresh leachate. Hence, COD is still the most important parameter characterizing the chemical property of leachate.

It has been preliminarily proved that leachate is not a homogeneous-phase liquid but a heterogeneous solution consisting of colloidal particulates with various diameters and a complex population of microorganisms. The molecular structure and elemental composition in landfill leachate is strongly influenced by microbial degradation processes. It has been proved that the coagulation with iron chloride can eliminate all high MW components (>5000 Da) in leachate, but does not trap compounds with a lower MW, which means that the coagulation process will not work to treat the fresh leachate after pre-treatment by biochemical treatments [16].

Nevertheless, although the molecular weight distribution in aged raw leachate in Canada had been reported [17], the details on the particle size distribution, percentages of colloidal and dissolved matters in fresh leachate in East Asia, where the refuse contains high contents of food wastes, are unknown yet. In addition, despite its importance [21,29], little knowledge is available about the composition and the fate of organic matter (OM) in leachate along the treatment processes [1,5]. It is important to understand the main components of leachate, including their molecular weight and particle sizes distributions, taking advantages of membrane technologies.

In this work, the colloids in leachate and effluents from biological treatment stages were separated in sequence using a series of micro- and ultra-membranes with different pore sizes, to have a better understanding of the mechanism of the biological processes at Laogang Landfill. The fresh leachate was collected from the operating landfill compartment directly, while the other samples were collected from the effluents discharged from the holding tank and the anaerobic, anoxic and aerobic lagoons established at Shanghai Laogang Refuse Landfill.

2. Materials and methods

2.1. Ultra-filtration apparatus

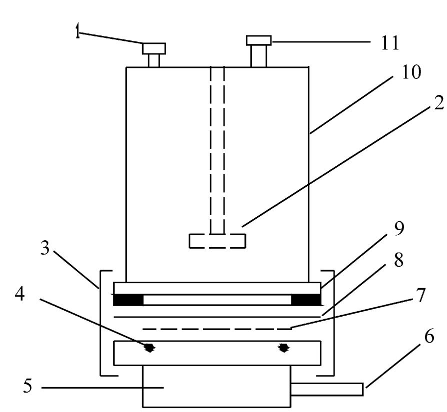
The ultra-filtration apparatus used in this work is shown in Fig. 1. It consists of a methyl-methacrylate glass holder with a volume of 300 mL. The effective membrane area is 0.02 m^2. The samples were pressed through the filters by pressure applied at the top of the barrel. N_2 was used to apply the pressure, the operation pressure of micro-filtration process and ultra-filtration process being maintained at 0.1 and 0.25 MPa, respectively. The filters were washed with 20 mL deionised water, and then conditioned with 20 mL of the sample before operation. Finally, 300 mL of sample were filtered until 20 mL leachate left in holder. The filtrate was then used for the analysis of COD, TOC and other parameters measurements.

2.2. Filtration and size fractionation of leachate

The leachate samples were separated into several size fractions, using a series of membranes with the following pore sizes (molecular weight cutoff): 1.2 μm, 0.45 μm, 1 kDa. The filtration scheme is shown in Fig. 2.

The leachate was first filtered by a 1.2 μm-membrane, then by a 0.45 μm-membrane, and finally by a 1 kDa MWCO membrane, all kindly supplied by Shanghai Nucleus Physics Institute of Chinese Academy of Sciences. The micro-membranes were made of mixed cellulose, while the ultra-membrane was made of polyether sulfone (PES).

In this work, the filtered residues retained by the 1.2 μm-membrane, the 0.45 μm-membrane and the 1 kDa MW-membrane were defined as suspended solid, coarse colloids and



1,admitting pipe; 2, stirrer;3,fixator; 4, O-ring seal packing; 5, seat 6, outlet; 7, flow deflector; 8, Ultra-membrane; 9, sealing plate; 10, ultrafilter cup; 11, feed inlet

Fig. 1. Schematic diagram of SCM type of ultra-filter.

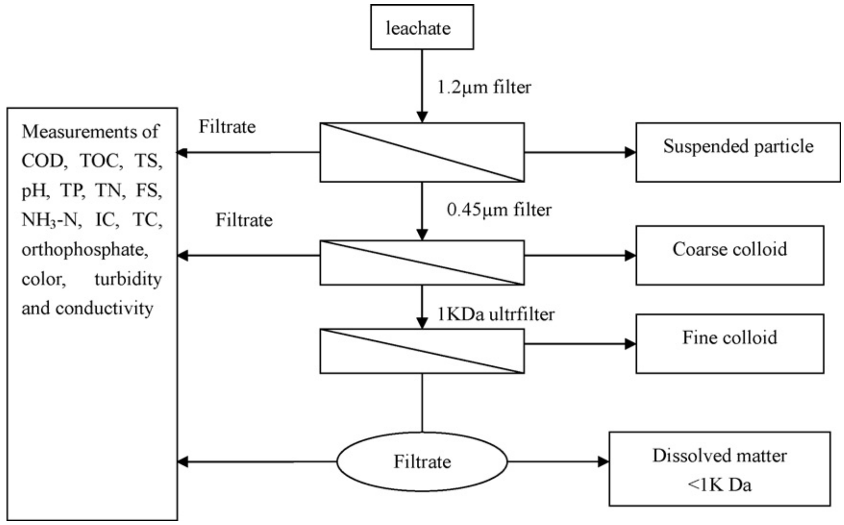


Fig. 2. Schematic diagram of organic matter size fractionation using filtration and ultra-filtration.

fine colloids, respectively whereas the filtrate from the 1 kDa MWCO-membrane filtration is considered as dissolved matter. Intermittent (batch) operation was used during the gradient size-fractionation.

2.3. Leachate and analytical methods

Laogang Refuse Landfill was constructed in 1985 along the shore of East China Sea, which was formed by the sedimentation of silt carried by Yangtze River and put into operation at the end of 1989. The landfill was separated into 56 landfill compartments through clay dikes, with 10 ha per compartment and a filling height of 4 m.

The oxidation ponds of 1000 t/day adopted at Laogang Refuse Landfill leachate treatment process consist of a holding tank and anaerobic, anoxic and aerobic lagoons in series, with a retention time of 90, 60, 30, and 20 days, respectively. The volume of the holding tank, anaerobic, anoxic and aerobic lagoons was 67,200 m^3 (140 m × 120 m × 4 m), 24,000 m^3 (100 m × 60 m × 4 m), 18,000 m^3 (100 m × 60 m × 3 m), 6000 m^3 (60 m × 40 m × 2.5 m), respectively. The anoxic lagoon is being stirred by 12 pumps with each power of 7.5 kW, and the oxygen supply devices in aerobic lagoon is ensured by four fans (three fans active and one fan standby) with each power of 75 kW. The effluent from the aerobic lagoon was then further treated by a wet land of reed field (Fig. 3). COD of raw leachate were 13,545–22,152 mg/L in the holding tank, 10,370–15,405 mg/L in the anaerobic lagoon, 6813–8421 mg/L in the anoxic lagoon, 2267–5447 mg/L in the aerobic lagoon, and NH_4^+ were 1891–3443 mg/L, 1265–3121 mg/L, 1030–2432 mg/L, 600–1137 mg/L, respectively in March 2003, according to the data from Laogang Landfill management department. The variation of COD and NH_4^+ values at Laogang Landfill is due to the meteorological changes. In general, COD ca n be reduced from 15,000 mg/L in leachate in the holding tank to around 3000 mg/L in the effluents from the aerobic lagoon, though higher influent COD will lead to a higher effluent COD. Currently, COD in effluents can not be reduced to lower than 300 mg/L, the discharge standard used in China, even if it is further treated in the wetland.

The samples tested in this work were taken from the biological treatment stages.

All the samples in this work were collected along the side of the tank or lagoons below the surface 0.1 m using a bucket (2 L volume) made of stainless steel for seven times at different points for both the tank and the lagoons and then immediately transferred into 10 L polyethylene containers, which were rinsed two times with deionised water and conditioned two times with sample waste water before sampling. Then, these polyethylene containers were kept in the dark and transported to the lab. In this work, the samples were stored at 4 °C until the end of the analysis in order to reduce the microbial activity in the samples.

The conductivity of leachate was also measured with conductivity instrument (DDS-NC); the pH by digital pH instrument

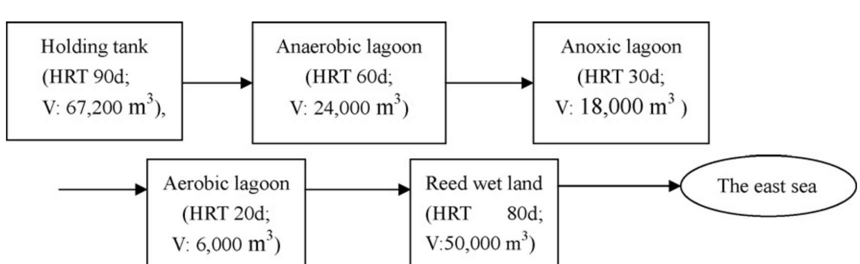


Fig. 3. Schematic diagram of biological treatment processes for leachate at Shanghai Laogang Refuse Landfill.

(PHB-4); the TOC by a specific instrument (TOC-V CPH/CPN, Shimadzu Corporation); while the total solid (TS) and fixed solid (FS) in each fraction of leachate were measured by weighting the residue of 20 mL corresponding fractions after drying in porcelain crucibles, using the classical desiccation (105 °C; 24 h) and calcination (600 °C, 1 h) methods. The COD, NH_4^+, phosphate, *ortho*-phosphate, turbidity and color were determined by the standard methods recommended by USEPA [22].

3. Results and discussion

3.1. Fresh leachate characterization

The fundamental parameters in fresh leachate and its effluents from each lagoon in the biological treatment processes are given in Table 1, where *fresh* represents fresh leachate, *holding* refers to leachate in holding tank; *anaerobic* to effluent in anaerobic lagoon; *anoxic* to effluent in anoxic lagoon; *aerobic* to effluent in aerobic lagoon, unless otherwise indicated in the text.

It can be found from Table 1 that COD, NH_4^+ and TS concentrations in fresh leachate were quite high, due to the high contents of food wastes (50–70%) in the refuse placement, and decreased significantly after being treated by the biological processes mentioned above, however COD in the effluent from the aerobic lagoon was still as high as approximately 3800 mg/L.

The COD concentration of leachate from the aerobic lagoon can be lower if the one in the holding tank is lower, and it may be reduced to less than 500 mg/L when the concentration in the influent in the holding tank is below 5000 mg/L (according to the statistics result of the leachate treatment process at Laogang Landfill resulting from the long term observations).

3.2. Effect of membrane porosity on COD

COD concentration of fresh leachate decreased from 54,651 to 44,573 mg/L after sequential separation by a series of membranes with differential pore sizes until 1 kDa, with a total removal of about 18.5%, indicating that the dissolved matters are predominant in fresh leachate (Fig. 4). COD removal in fresh leachate was 14.2% after filtration with a 1.2 μm membrane, meaning that the suspended particles had a great contribution to the COD removal by membranes in series. COD concentration in leachate from the holding tank decreased from 15,193 to 7289 mg/L after the sequential separation until 1 kDa, with a total removal of 52%, among which the suspended particle occupied 24%. The corresponding values of COD in effluents from anaerobic lagoon, anoxic lagoon, and aerobic lagoon decreased from 12,000 to 7744 mg/L, 7744 to 5968 mg/L, and 3773 to 2248 mg/L, with total removals of about 36%, 23% and 40%, respectively, while the suspended particles occupied about 21%, 21% and 32% in the COD removals.

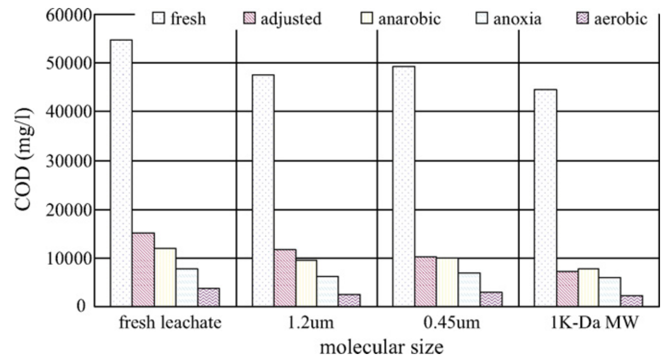


Fig. 4. Relationship between COD in filtrate and molecular sizes.

Obviously, the major part of matter in the fresh leachate or effluents in the treatment lagoons was the dissolved matter with a molecular weight cutoff below 1 kDa MW. Furthermore, the suspended particles actually had a great contribution to the COD removal due to the retention by the membranes used, ranging from 50% to 90%.

3.3. Effect of membrane porosity on TOC

TOC in fresh leachate and effluents from the treatment lagoons are given in Figs. 5–9, respectively. TOC concentration was 16,232 mg/L in the fresh leachate, and decreased to 745 mg/L in the aerobic lagoon effluent after the biological treatment processes. Meanwhile, it was found that the percentages of the dissolved matters in leachate from the different treatment

Table 1
Elemental analysis data of fresh leachate and effluents from different lagoons at Shanghai Laogang Refuse Landfill (mean ± S.E.)

Items	Fresh leachate	Effluent in holding tank	Effluent in anaerobic lagoon	Effluent in anoxic lagoon	Effluent in aerobic lagoon
COD (mg/L)	54,651 ± 72	15,193 ± 46	12,000 ± 37	7744 ± 64	3773 ± 57
TOC (mg/L)	16,232 ± 37	5438 ± 25	3839 ± 22	2312 ± 9	745 ± 5
NH_4^+ (mg/L)	3143 ± 18	2894 ± 6	2074 ± 4	2053 ± 3	808 ± 3
TN (mg/L)	3952 ± 3	3018 ± 2	2103 ± 3	2074 ± 3	1366 ± 2
ortho-Phosphate (mg/L)	16.7 ± 0.9	9.9 ± 0.2	9.9 ± 0.3	2.7 ± 0.1	2.7 ± 0.1
TP (mg/L)	25.4 ± 0.3	13.6 ± 0.1	11.8 ± 0.2	5.4 ± 0.1	3.8 ± 0.2
TS (mg/L)	54,800 ± 11	19,950 ± 23	13,450 ± 50	12,200 ± 43	11,650 ± 23
FS (mg/L)	20,450 ± 21	10,100 ± 15	9000 ± 29	7850 ± 31	6840 ± 18
Conductivity (ms/m)	2.81 ± 0.01	2.1 ± 0.02	2.06 ± 0	2.14 ± 0.01	1.65 ± 0
pH	7.13 ± 0.01	7.27 ± 0.01	7.75 ± 0	8.07 ± 0.01	8.66 ± 0
Color	1000 ± 58	600 ± 17	550 ± 29	500 ± 17	400 ± 17
Turbidity (NTU)	1749 ± 5	1135 ± 12	1007 ± 9	930 ± 13	674 ± 7

Sampling date March 2003 (T = 15 °C).

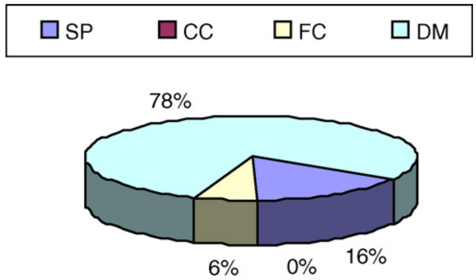


Fig. 5. Distribution of TOC in fresh leachate.

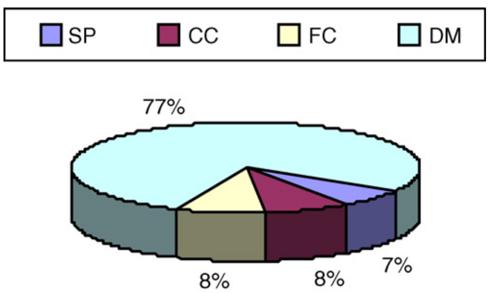


Fig. 6. Distribution of TOC in effluent from holding tank.

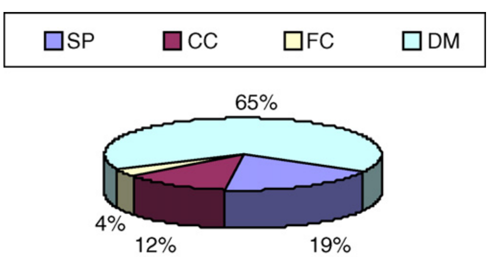


Fig. 7. Distribution of TOC in effluent from anaerobic lagoon.

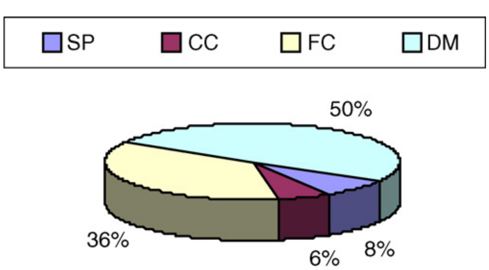


Fig. 8. Distribution of TOC in effluent from anoxic lagoon. *Note*: the percentage 0% means that the change in terms of TOC in this fraction is very low, and it beyond the detectability of the method.

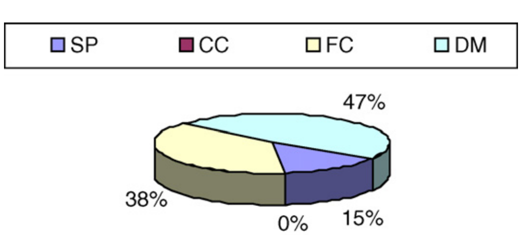


Fig. 9. Distribution of TOC in effluent from aerobic lagoon.

lagoons in terms of TOC decreased, varying from 78% in the fresh leachate, 77% in the holding tank, 65% in the anaerobic lagoon, 50% in the anoxic lagoon, and to 47% in the aerobic lagoon, respectively.

However, the percentages of fine colloidal particles in TOC increased from 6% to 38% as treated by the biological treatment lagoons, possibly due to the degradation of suspended particles into middle- and micro-particles during the treatment process, what changed the molecular weight distribution in leachate.

3.4. Effect of membrane porosity on pH

The pH of leachate or effluents in the treatment stages after gradient separation by the membranes is shown in Fig. 10. It can be seen that pH values increased when the fresh leachate was treated in series by the stages used, from pH 7.13 in fresh leachate to pH 8.66 in the effluent from aerobic lagoon, and it also slightly increased as the same effluents were sequentially filtered by the membranes. Hence, it can be assumed that the acidic matters (maybe due to the stripping of CO_2 from the leachate during the separation) can be removed when the leachate is treated in the treatment units or through the gradient membrane separation.

3.5. Effect of membrane porosity on fixed solid (FS) and total solid (TS)

The total solid (TS) of fresh leachate was 54,800 mg/L, while the TS in the leachate from the holding tank was 19,950 mg/L, indicating that the holding tank is an important unit in the leachate treatment process for the removal of solid particles (Fig. 11). The TS of leachate from holding tank, anaerobic, anoxic, and aerobic lagoons occupied 36%, 24%, 22%, and 21% of that in fresh leachate, respectively.

TS of fresh leachate decreased from 54,800 to 32,950 mg/L during the membrane separation process, while that of effluents from the various stages of the treatment process decreased slightly after being separated by the membranes in series, possibly due to the removal of the high molecular weight volatile matters during the separation and perhaps also due to the hydrolysis of macro-molecular matters into middle-, micro-molecular matters along the treatment process.

The FS of the effluents from the holding tank, anaerobic, anoxic, and aerobic lagoons (Fig. 12) occupied respectively 49%, 44%, 38% and 33% of that in fresh leachate. It was

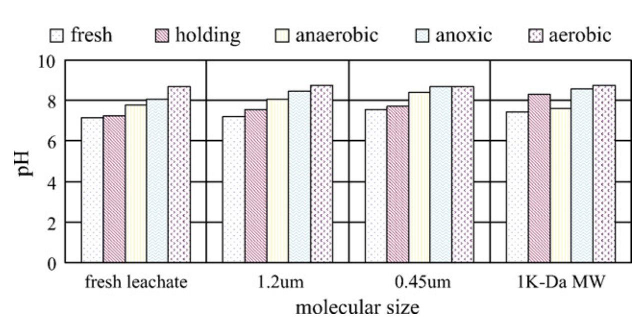


Fig. 10. Relationship between pH and molecular sizes.

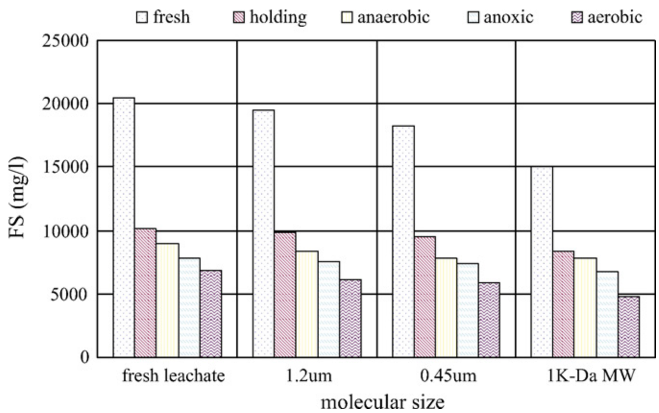


Fig. 11. Relationship between FS and molecular sizes.

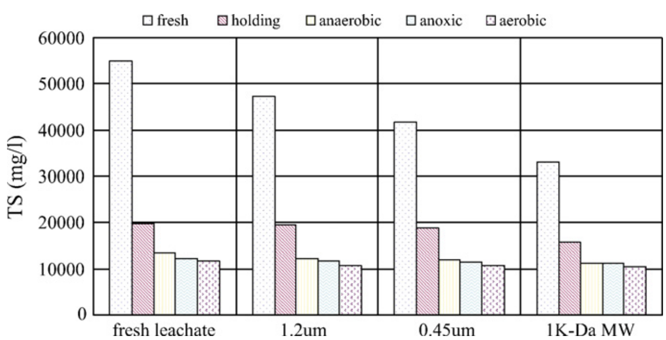


Fig. 12. Relationship between TS and molecular sizes.

also confirmed that the fraction <1 kDa MW predominated in leachate in terms of FS.

3.6. Effect of membrane porosity on conductivity

From Fig. 13, it can be shown that the conductivity of leachate was much higher than that of typical municipal wastewater, and it decreased from 2.81 mS/m for the fresh leachate to 1.65 mS/m for the effluent from the aerobic tank. Meanwhile, the conductivity varied from 1.55 to 2.90 mS/m after gradient separation by the membranes, possibly because of the change of ion activity in leachate during the filtration. The conductivity changed slightly when the leachate was treated in series by anaerobic, anoxic and aerobic lagoons, indicating that the salts cannot be effectively removed by these biological treatment processes, as the conductivity can be related to the salt concentration.

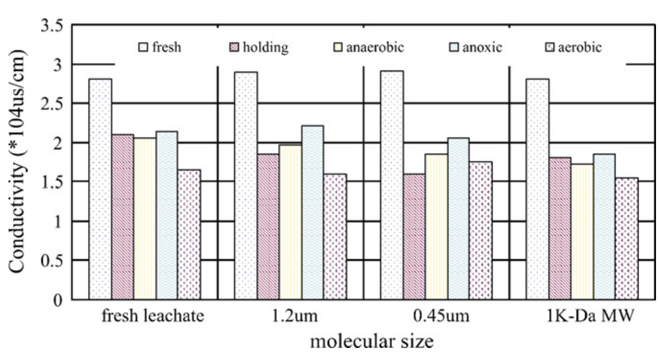


Fig. 13. Relationship between the conductivity and molecular sizes.

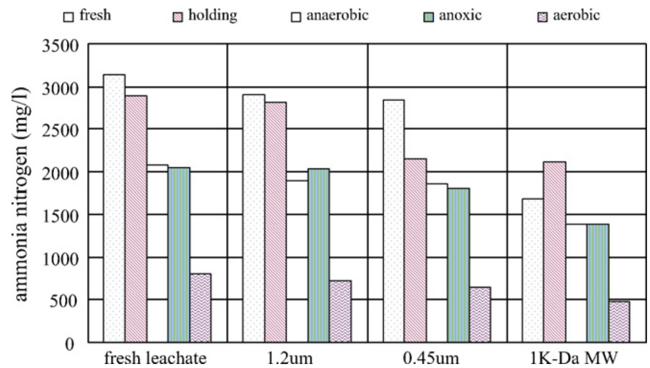


Fig. 14. Relationship between the ammonium nitrogen and molecular sizes.

3.7. Effect of membrane porosity on N

Figs. 14 and 15 showed the relationships between NH_4^+ and total N (TN) in leachate, effluents and the membrane porosities used. TN and NH_4^+ in fresh leachate were 3952 and 3143 mg/L, respectively, and decreased slowly after sequential filtration by the membranes.

Around 60–70% of TN and NH_3-N in fresh leachate can be removed after sequential filtration by the membranes and the remaining TN and NH_3-N should be considered as soluble forms. It was also found that NH_4^+ occupied about 80–90% of total N, and that the ratio of NH_4^+/TN in leachate decreased after it being treated by these biological processes (from 90% to 60% in aerobic tank).

3.8. Effect of membrane porosity on TP and ortho-phosphate

Total P (TP) and *ortho*-phosphate (OP) in fresh leachate were 25.4 and 16.7 mg/L respectively, and decreased considerably after the filtration using 1.2 and 0.45 μm membranes, showing that most of the Phosphorus in the fresh leachate can be presented as suspended particles and coarse colloids, as given in Figs. 16 and 17. Meanwhile, TP and phosphate in fresh leachate decreased to 14.7 and 2.9 mg/L after filtration using 1.2 μm membrane respectively.

TP in leachate decreased remarkably after the treatment processes, which was 25.4, 13.5, 11.8, 5.3, and 3.8 mg/L in series, with a total removal of 85%. The ratios of *ortho*-phosphate/TP in leachate were 66%, 73%, 83%, 50%, 50%, and 70%, indicating that *ortho*-phosphate occupied the major part of TP in leachate.

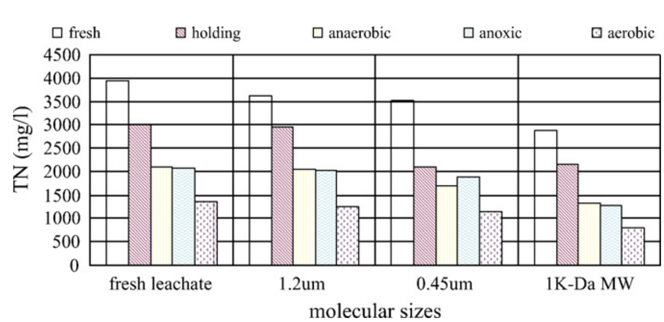


Fig. 15. Relationship between TN and molecular sizes.

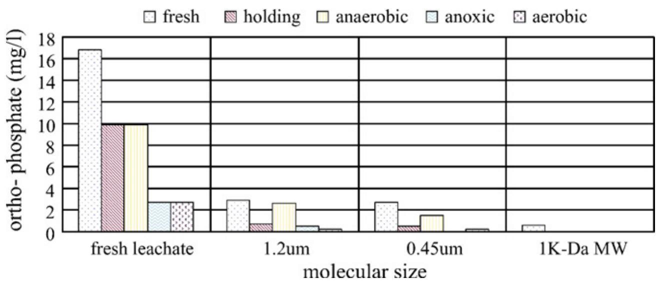


Fig. 16. Relationship between *ortho*-P and molecular sizes.

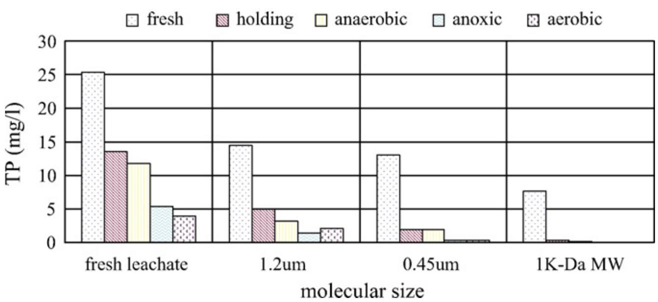


Fig. 17. Relationship between total P and molecular sizes.

3.9. Effect of membrane porosity on color and turbidity

Turbidity in fresh leachate was 1749 NTU, and color was 1000 units, which decreased to 674 NTU and 400 units after the treatment processes, with a removal of about 50–60% (Figs. 18 and 19). Turbidity in fresh leachate was reduced to 300 NTU after gradient separation using the membranes. Hence, suspended solids and coarse colloids in leachate can contribute greatly to the turbidity. The color of effluents from the treatment processes remained 200–300 units after sequential filtration by the membranes.

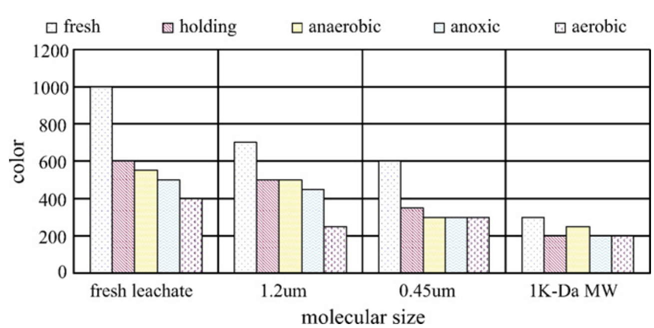


Fig. 18. Relationship between the color and molecular sizes.

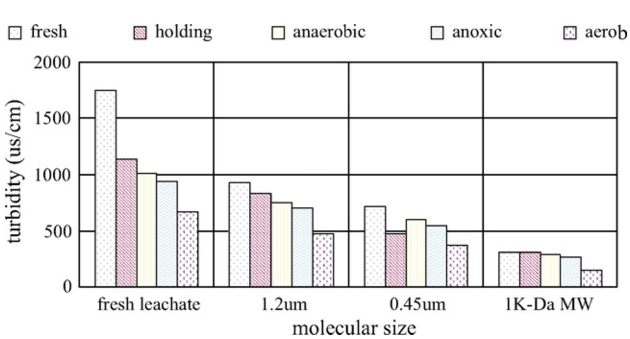


Fig. 19. Relationship between the turbidity and molecular sizes.

3.10. Discussion

Leachate contains a large amount of organic matter with wide range of molecular sizes, and its oxidation/reduction potential will decrease during storage. Concerning the dissolved matter (DM), the ratio DM/TS in fresh leachate, effluents from holding tank, anaerobic, anoxic and aerobic lagoons were about 0.60, 0.78, 0.83, 0.92 and 0.89, respectively. The percentages of DM in terms of TOC in leachate decreased in the biological treatment processes, indicating that the matters removed by the biological treatment should be predominantly in some soluble matters. Furthermore, it can also be assumed that the biochemical treatment techniques are effective for the removals of the matters with a molecular weight below 1000 Da. In comparison, coagulation, activated carbon absorption, biological treatment processes, and RO technology can remove macro-molecular weight with >3000 Da, 500–3000 Da MW, micro-molecular, and dissolved matter, respectively [18]. Therefore, it demonstrates that a RO unit should be used in leachate treatment process at Laogang Refuse Landfill after the biological treatment according to the result of effluents from aerobic tank.

Similarly, the ratios of FS/TS in the corresponding leachate or effluents were 0.37, 0.51, 0.67, 0.64, and 0.59, respectively, indicating that FS predominated in the leachate or effluents issued from the treatment stages (Fig. 12). Comparing to the effluents from the treatment stages, fresh leachate contained more organic matter and most of the removals were volatile organic matter, occurring during the biochemical processes.

It is observed that both carbon and nitrogen have similar size distribution patterns, with the DM having the largest share for both OC and N. The MW distribution in terms of TOC, TN, NH_4^+ in leachate from Laogang Landfill showed that the dissolved matters with a MW <1000 Da predominated in leachate, and that the suspended particles with a MW >1.2 μm also had a great contribution to the removal of COD, comparing to the other fractions over 1000 Da, while a wide range of the MW distribution of N in leachate was among the fractions >1000 Da. However, a considerable portion of the TP and *ortho*-phosphate was associated with the SP and CC, defined as the size fraction >0.45 μm, and the results is consistent with the fact that the fresh leachate contained mainly micro-molecular dissolved organic matter, i.e. fatty acid [19], and it is also consistent with previous observations of leachate's molecular weight fractions distribution (Christian et al., 2000; [20]).

4. Conclusion

The size fractionation procedure used to characterize the matter contained in leachate and to discuss the mechanism of the leachate's treatment techniques seems to work satisfactorily. Colloids were found in all the landfill leachate samples and the majority of the components in leachate were those with molecular weight below 1 kDa MWCO. P in leachate should be presented as suspended solid and coarse colloids, which can be readily separated by the membranes. NH_4^+ contributed to the major part of N in leachate, occupying about 80–90% of the TN. The decease of dissolved fraction percentages and increase of middle-molecular matters in leachates along the treatment process at Laogang Landfill shows that the biological treatment processes can remove micro-molecular weight matters more efficiently, comparing to the matter in coarse fraction and suspended matter.

Acknowledgements

This work was financially supported by the China National Natural Science Foundation with contract No. 20177014 and No. 29777019, Doctoral Program Foundation of the China Education Ministry.

References

[1] G. Christian, Organic halogens in landfill leachates, Water, Air, Soil Pollut. 120 (2000) 331–345.

[2] F. Caron, S. Elchuk, Z.H. Walker, High-performance liquid chromatographic characterization of dissolved organic matter from low-level radioactive waste leachates, J. Chromatogr. A 739 (1996) 281–294.

[3] K. Klaus, G. Georg, H. Ludwig, et al., The composition of dissolved organic matter in forest soil solutions: changes induced by seasons and passage through the mineral soil, Org. Geochem. 33 (2002) 307–318.

[4] E. Benfenati, G. Facchini, P. Pierucci, et al., Identification of organic contaminants in leachates from industrial waste landfills, Trends Anal. Chem. 15 (8) (1996) 305–310.

[5] D.L. Jensen, T.H. Christensen, Colloidal and dissolved metals in leachates from four Danish landfills, Water Res. 33 (9) (1999) 2139–2147.

[6] M.A. Nanny, N. Ratasuk, Characterization and comparison of hydrophobic neutral and hydrophobic acid dissolved organic carbon isolated from three municipal landfill leachates, Water Res. 36 (2002) 1572–1584.

[7] D. Trebouet, J.P. Schlumpf, P. Jaouen, et al., Stabilized landfill leachate treatment by combined physicochemical-nanofiltration processes, Water Res. 35 (12) (2001) 2935–2942.

[8] J. Bohdziewicz, M. Bodzek, J. Gorska, Application of pressure-driven membrane techniques to biological treatment of landfill leachate, Process Biochem. 36 (2001) 641–646.

[9] Z. Lou, Y. Zhao, Q. Zhang, Leachate Treatment and Practical Application, Chemical Industry Press, Beijing, 2006 (in Chinese).

[10] A. Chianese, R. Ranauro, N. Verdone, Treatment of landfill leachate by reverse osmosis, Water Res. 33 (1999) 647–652.

[11] R. Rautenbach, T.H. Linn, High-pressure reverse osmosis and nanofiltration, a zero discharge process combination for the treatment of waste water with severe fouling/scaling potential, Desalination 105 (1996) 63–70.

[12] H. Russell, Plumb Jr., Characterization of Mine Leachates and the Development of Ground-Water Monitoring Strategy for Mine Sites, EPA/600/R-99/007, Las Vegas, Nevada, 1999.

[13] T. Urase, M. Salequzzaman, S. Kobayashi, et al., Effect of high concentration of organic and inorganic matters in landfill leachate on the treatment of heavy metals in very low concentration level, Water Sci. Technol. 36 (12) (1997) 349–356.

[14] E. Benfenati, P. Pierucci, R. Fanelli, et al., Comparative studies of the leachate of an industrial landfill by gas chromatography-mass spectrometry, liquid chromatography-nuclear magnetic resonance and liquid chromatography-mass spectrometry, J. Chromatogr. A 831 (1999) 234–256.

[15] G. Franz-Josef, P. Wilhelm, Structural investigation of isolated aquatic fulvic and humic acids in seepage water of waste deposits by pyrolysis-gas chromatography/mass spectrometry, Water Res. 31 (7) (1997) 1609–1618.

[16] E. Chian, Stability of organic matter in landfill leachates, Water Res. 11 (1977) 225–232.

[17] F. Wang, D.W. Smith, M.-G. El-Din, Aged raw landfill leachate: membrane fractionation, O_3 only and O_3/H_2O_2 oxidation, and molecular size distribution analysis, Water Res. 40 (3) (2006) 463–474.

[18] X. Liu, L. Cao, Z. Wang, Study on the removal of the different molecular-weight organics by water purification process, China Environ. Sci. 18 (4) (1998) 341–344 (in Chinese).

[19] Y. Zhao, The Application Manual of Environmental Engineering, Chemical Engineering Publishing House, Beijing, 2002 (in Chinese).

[20] N. Calace, A. Liberatori, B.M. Petronio, et al., Characteristics of different molecular weight fractions of organic matter in landfill leachate and their role in soil sorption of heavy metals, Environ. Pollut. 113 (2001) 331–339.

[21] T. Assmuth, S. Penttilä, Characteristics, determinants and interpretations of acute lethality in daphnids exposed to complex waste leachates, Aquat. Toxicol. 31 (1995) 125–141.

[22] Editorial Board of Analytical technique on monitoring water and wastewater belonging to China agency of environmental protection. Analytical technique on monitoring water and wastewater, *Chinese Press of Environmental Science* Beijing (1989) 296–299 (in Chinese).

[23] L.A. Gary, et al., Comparing gel chromatography and ultrafiltration for the molecular weight characterization of aquatic organic matter, J. AWWA 79 (1) (1987) 43–49.

[24] G.L. Joseph, S.S. Gina., The effect of organic carbon on the sequential reductive dehalogenation of tetrachloroethylene in landfill leachates, Water Res. 34 (8) (2000) 2390–2396.

[25] L. Kristina, J. Ann-Sofi, Nanofiltration of salt solutions and landfill leachate, Desalination 103 (1995) 223–232.

[26] X.Z. Li, Q.L. Zhao, Efficiency of biological treatment affected by high strength of ammonium-nitrogen in leachate and chemical precipitation of ammonium-nitrogen as pretreatment, Chemosphere 44 (2001) 37–43.

[27] K. Linde, A-s. Jönsson, R. Wimmerstedt, Treatment of three types of landfill leachate with reverse osmosis, Desalination 101 (1995) 21–30.

[28] M.F. Dignac, P. Ginestet, A. Bruchet, et al., Changes in the organic composition of waster during biological treatment as studied by NMR and IR spectroscopes, Water Sci. Technol. 43 (2) (2001) 51–58.

[29] M.C. Scordal, Colloidal pumping, evidence for the coagulation process using colloids tagged with Hg, Environ. Sci. Technol. 30 (11) (1996) 3335–3340.