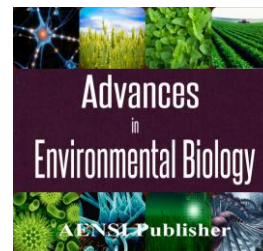




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Ammoniacal Nitrogen Contaminated Groundwater at Taman Beringin Ex-Landfill site: Implication to Health and the Environment

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ABSTRACT

Ammoniacal nitrogen make up the frequent contaminant of surface and groundwater, exceeding appropriate standards around landfills in Malaysia. Leachate and groundwater samples were collected from an ex-landfill site at Taman Beringin in Kuala Lumpur, Malaysia and were tested for Various physico-chemical parameters to investigate the characteristics of the groundwater and possible impacts on the environment. The results showed considerable impact of leachates on groundwater with high concentration of most chemical parameters in groundwater samples. COD (101.75±99.42), BOD (28±30.99), NH-N (31.10±19.12), F-(0.72±0.32) and heavy metal; Pb (0.042±0.045), Ni (0.016±0.006), Fe (0.41±0.68) were above the Malaysian National Drinking Water Quality Standard (NDWQS) and National Water Quality Standards (NWQS) CLASS II A. In particular, the levels of NH₃-N recorded in all the groundwater wells investigated at BH3 (12.89), BH5 (22.22), BH6 (32.01) and BH7 (57.30) were far above the NWQS CLASS IIA set at 0.3mg/L. Remedial measures are suggested to minimize the effects on the environment; improving the quality of the leachates and prevent further spreading of leachates into river, drinking water wells and other groundwater resources via groundwater flow are recommended.

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INTRODUCTION

Contaminations of groundwater by landfill leachates constitute a major problem arising from improper landfill practices. The problem of the Malaysia situation have been aggravated due to the fact that most of the existing solid waste disposal sites are not engineered landfill type; very few of the landfills could be regarded as basic sanitary landfill type. Worse is the case as ammoniacal nitrogen (NH₃-N) is identified as one of the main pollutants to render many of the rivers in Malaysia polluted and elevated level had resulted in the temporary closing of some of the landfills such as the Cheras WTP, thrice in 1997[6]. Furthermore, as pointed out in [6], the Malaysian National, water Resources Study 2000-2050, showed that the parameters which have frequently exceeded Class III limits (water which may be used for water supply with extensive/advanced treatment) along side with ammoniacal-nitrogen includes organic carbons, heavy metals, oil and grease. These chemical substances corroborate with the frequent and potential contaminant of soil, surface and groundwater noticeable around landfills worldwide and therefore elevated concentrations of these substances is a potential threat to public health and the environment.

Improper landfill practice such as landfilling without liner systems is one of the major way NH₃-N contaminant could occurs in groundwater aquifers. NH₃-N In groundwater aquifer reacts through complex physical- chemical and biological processes to form other associated nitrogen bearing compounds such as NO₂, NO₃ which makes the risk even more complicated. The term "ammoniacal – nitrogen" or total ammonia as referred to in this work is the sum of ammonia (NH₃) and ammonium (NH₄⁺) concentrations. Their toxicity is attributed to the influence of pH, temperature, and dissolved oxygen concentration. However, pH is the most important factor in considering ammonia toxicity. The investigation of the groundwater quality at the ex-landfill

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sites at Taman Beringin for proper updates of water quality standards and remediation are necessary for the management of the environment as well as for the sustainability of water resources in Malaysia. Presented herein is the discussion surrounding the potential impacts on groundwater quality associated with $\text{NH}_3\text{-N}$ contamination from leachates as evident of the post disposal of waste in an ex-MSW landfill at Taman Beringin Malaysia. Figure 1 shows Taman Beringin Landfill (TBL) site in Kuala Lumpur and the position of groundwater monitoring wells in the landfill site.



Fig. 1: Taman Beringin Landfill (TBL) site and the position of groundwater monitoring wells.

MATERIALS AND METHODS

Description of Study Area:

Taman Beringin ex- landfill (TBL) site is a 30-acre site located at Northern part of Jinjang; about 10km North West of Kuala Lumpur Federal Capital Territory of Malaysia at latitude $03^{\circ} 13.78'$ North and longitude $101^{\circ} 39.72'$ East. TBL was not designed as sanitary landfill type and was designated municipal solid waste (MSW) landfill by DBKL to accommodate only domestic and commercial waste origin collected in the city. The landfill area is mostly surrounded by residential apartment at the very edge and near surroundings. Three of the groundwater monitoring well stations is within the three residential areas (Figure 1). The Taman Beringin solid waste transfer station is located on the north-western flank of the landfill close to monitoring well BH3.

Currently climate are unpredictable in Malaysia, however, Taman Beringin landfill in Kuala Lumpur has a tropical climate. There are two monsoons between March-April and October-November coinciding with the southeast and southwest monsoon seasons respectively. October, November and April are the wettest months of the year and November receives more than 400 mm of precipitation while March-April gets around 700 mm of rainfall. May and June can be considered as the driest one and June gets less than 140 mm of rainfall. Total annual precipitation averages 2366.2 mm (93.2 in). Temperature varies a little from season to season with the average high of 28°C - 32°C while the low hardly falls below mid twenties. In the northern flank of the study area is Sungai Jinjang which is an important tributary of a major upstream, River Batu. River Jinjang flows into the landfill at BH3 at the north-eastern part. In the studied area, a groundwater modeling confirmed the groundwater flows towards the river / pond in a direction of east to south-east (See Figure 1). The aquifer in this site is a shallow sandy type of about 5-10 m thickness and a deeper fractured bedrock limestone.

Groundwater and leachates sampling:

The sampling of the groundwater was conducted between 19 December 2012 and 11th - 29th January 2013 in some selected groundwater points. Four existing observation wells, previously installed were selected to collect groundwater samples. The choosing monitoring wells have been designated BH3, BH5, BH6 and BH7 and well located exactly on the pathway of the groundwater flow (Figure 1). Monitoring wells were first being purged

with low flow pumping in order to remove standing water in the borehole and obtain fresh groundwater for sampling. Sample collection was carried out as soon as the field parameters such as pH, temperature had stabilized. Water collected in the sampling bailer was transferred to the sample bottles prepared for the various analyses. The respective preservatives were then added to the samples before being kept at temperature of 4°C for onward transportation to the laboratory. Raw surface leachates from six different points at the base of the landfill were also collected between 17-19 December 2012. The chain-of-custody form was then filled for onward transmission to the receiving laboratories for analysis.

Laboratory Analysis:

Leachates and groundwater samples were tested in the laboratories of Spectrum Sdn. Bhd and E. S. Tech venture Sdn. Bhd. Physico-chemical analyses followed Standard Methods for the Examination of Water and Wastewater by the American Public Health Association (APHA) 2005. Physico-chemical parameters examined in groundwater and leachates samples include: color (ADMI), total dissolved solids (TDS), total suspended solids (TSS), total hardness (TH), biological oxygen demand (BOD), chemical oxygen demand (COD), Oil/grease, phenol, sulphides (S^{2-}), fluoride (F^-), chloride (Cl^-), ammonical nitrogen ($NH_3^+ - N$), sulphate (SO_4^{2-}) and nitrate (NO_3^-). The heavy metals included: cadmium (Cd), copper (Cu), iron (Fe), nickel (Ni), lead (Pb) zinc (Zn), and manganese (Mn).

RESULT AND DISCUSSIONS

Results of surface raw-leachates analysis:

Table 1: Descriptive statistics for physico-chemical parameters of leachate samples from Taman Beringin landfill and the comparison with DOE regulation 2009 standard.

Parameters	Mean \pm Standard Deviation	Minimum	Maximum	DOE Regulation 2009
pH	7.8 \pm 0.084	7.6	7.8	6.0-9.0
Color(ADMI)	1469.8 \pm 619.3	995	2267	100
COD	456.16 \pm 102.42	309	586	400
BOD	90.5 \pm 20.8	60	116	20
BOD/COD	0.19	0.19	0.19	NS
TSS	81.5 \pm 75.8	21	182	50
Phenol	0.008 \pm 0.002	0.006	0.011	0.001
Oil/grease	10 \pm 5.06	4	16	5.0
$NH_3 - N$	175.5 \pm 73.3	96.2	273	5.0
S^{2-}	0.15 \pm 0.07	0.1	0.2	0.50
F	2.72 \pm 1.63	0.7	4.4	2.0

The pH is dimensionless; except otherwise stated, all units are in mg/L, maximum number of samples (n) = 6, Regulation 2009 [3]

The descriptive statistics for physico-chemical parameters of leachate samples from Tama Beringin Landfill (TBL) and comparison with DOE Regulation 2009 standard are presented in Table 1. The pH of the six leachate samples from TBL site is generally alkaline with pH values varying between 7.6 and 7.8 with mean value of 7.8 and standard deviation value of ± 0.084 indicating that the pH values were in close range. However, the mean values of most contaminants were relatively high in the leachates samples from the Taman Beringin landfill relative to the DOE regulation 2009, with the exception of S^{2-} at 0.15 \pm 0.07 (see Table 1) The ratio of BOD/ COD (0.19) of raw leachate remains similar throughout the range of concentrations. $NH_3 - N$ was present in excessively higher level at concentration of 175.5 mg/L relative to the compared standard set at 5.0mg/L.

The measured values of heavy metals characteristics of the leachate samples from the Taman Beringin ex- landfill site and the comparison with DOE Regulation 2009 standard are presented in Table 2. The mean concentration of heavy metals in the leachate rarely occur at concentration large enough when compared to the DOE regulation 2009 standard, with the exception of Mn. The concentrations of Mn (0.27 \pm 0.07) slightly exceeded the stipulated values of 0.20 set by the compared 2009 standard. Although the mean concentration for Fe (4.78mg/L) is slightly below the standard but some of the leachate samples show higher Fe concentration with maximum values at 5.783mg/L above tolerance levels of 5mg/L. Therefore Fe and Mn are considered as the heavy metal contaminants of the leachates constituents in the study area within the context of this study.

Table 2: Heavy metal characteristics of surface raw leachates samples from Taman Beringin ex- landfill and comparison with DOE regulation 2009 standard.

Parameters	Fe	Mn	Pb	Cu	Ni	Cd	Zn
Regulation 2009	5.0	0.20	0.20	0.20	0.20	0.01	2.0
Minimum	3.327	0.181	0.021	0.018	0.019	0.001	0.81
Maximum	5.783	0.355	0.123	0.069	0.043	0.002	0.248
Mean \pm SD	4.78 \pm 0.98	0.27 \pm 0.07	0.07 \pm 0.041	0.041 \pm 0.019	0.031 \pm 0.009	0.0018 \pm 0.0004	0.17 \pm 0.067

All units are in mg/L; SD-Standard Deviation, maximum number of samples (n) = 6, Regulation 2009[3]

Result of groundwater analysis:

Table 3 shows results of Physico-chemical characteristic of groundwater samples at the landfill site with increasing distance from the landfill and the comparisons with the National Drinking Water Quality Standard (NDWQS). In cases where there were no stipulated values by NDWQS, results were compared with the proposed National Water Quality Standards (NWQS) for Malaysia CLASS II A.

The results of temperature of the groundwater samples were almost identical with a mean temperature value of 29.1°C. The pH in the groundwater samples range from 7.8-7.6 and were found to be generally alkaline at mean values of 7.7. For the most part values of pH were in close range, and are indicative of the standard deviation value of ± 0.081 . However, the pH values recorded for the groundwater samples in all the wells in this study is within the stipulated pH tolerance range of 6.5 to 9.0 set by the Malaysian National Drinking Water quality standard (NDWQS). The results of BOD in each of the individual well sampled have exceeded the NWQS (Class II A) with the exception in BH6 which are at same concentration of 3mg/L with the compared standard. It was observed that the values recorded for COD in all the wells sampled were relatively higher than the corresponding BOD values of similar well. The mean and standard deviation of COD were found to be 101.75 ± 99.42 and these values are by far above the NWQS (CLASS IIA) stipulated value. Further analysis showed that the mean and standard deviation value of BOD/COD value in groundwater samples is 0.26 ± 0.10 which signify generally low organic content of the groundwater samples. However, the pattern observed in the BOD/COD ratios from upstream to downgradient wells as presented in Table 3 indicates a slow to unsustained biodegradability throughout the landfill. Exceptionally high Cl^- concentration (301 mg/L) was recorded for groundwater samples in aquifer around BH7 which is closet downgradient well. This value exceeded the stipulated chloride levels allowed in groundwater according NDWQS.

Table 3: Physico-chemical characteristic of groundwater samples from Taman Beringin landfill site with increasing distance from the landfill and the comparisons with the National Drinking Water Quality Standard (NDWQS).

Parameters	BH3	BH7	BH5	BH6	Mean \pm SD	NDWQS
Distance(m)	169.73	223.95	341.56	396.68	282.98	-
Temperature (° C)	29.5	29.0	29.1	29.1	29.1 ± 0.22	NS
pH	7.6	7.7	7.7	7.8	7.7 ± 0.081	6.5-9.0
Color(ADMI)	15	149	56	15	58.75 ± 63.19	NS
T.H (EDTA)	243	436	354	206	309.75 ± 105.06	500
COD	23	196	179	9	101.75 ± 99.42	25 ^(*)
BOD	4	37	68	3	28 ± 30.99	3 ^(*)
BOD/COD	0.17	0.18	0.37	0.33	0.26 ± 0.10	NS
TSS	-	82	2	-	42 ± 56.56	50 ^(*)
TDS	392	2200	510	400	875.5 ± 884.63	1000
$\text{NH}_3\text{-N}$	12.89	57.30	22.22	32.01	31.10 ± 19.12	0.3 ^(*)
SO_4^{2-}	8.4	3.7	195	ND	69.03 ± 109.11	250.0
NO_3^-	1.26	2.23	ND	1.92	1.80 ± 0.49	10.0
Cl^-	5.4	301	40.8	39.7	96.72 ± 137.17	250
S^{2-}	ND	ND	ND	ND	-	NS
F ⁻	0.4	1.0	0.5	1.0	0.72 ± 0.32	0.4-0.6
Oil/grease	ND	ND	ND	ND	-	NS
Phenol	ND	ND	ND	ND	-	0.002

The pH is dimensionless; except otherwise stated, all units are in mg/L, ND: Not detected, NS- not Specified, SD-Standard Deviation, NDWQS- National Drinking Water Quality Standard (NDWQS) [2] ^(*)-National Water Quality Standards (NWQS) for Malaysia CLASS IIA (represent water supply of good quality set for the protection of human health and sensitive aquatic species) [5].

Relative high concentration values of $\text{NH}_3\text{-N}$, and F were observed in the groundwater samples. The concentration of F (1mg/L) recorded in BH6 and BH7 were similar and the value obtained in the two wells and their overall mean value (0.72mg/L) obtained in the groundwater sample are above the Malaysian drinking water standard (NDWQS). The highest value recorded for $\text{NH}_3\text{-N}$ is 57.30mg/L in BH7 and the lowest is 12.89 mg/L in upstream BH3. It was observed that, levels of $\text{NH}_3\text{-N}$ concentration were above the compared standard in all the wells at upstream BH3, and downgradient wells at BH5, BH6, and BH7. In addition, the level of $\text{NH}_3\text{-N}$ in BH6 (32.01mg/L) about 396.68m away from landfill was observed to be relatively higher than the levels in BH5 (22.22 mg/L) which is closer to the landfill at a distance of 341.56m. These pattern shows that $\text{NH}_3\text{-N}$ is undecending with increasing distance from the landfill. However, there is evidence of biological oxidation process of $\text{NH}_3\text{-N}$ (nitrification) which could be related to the presence of NO_3^- in the groundwater aquifer with the exception of the aquifer in BH5 where NO_3^- were undetectable (no nitrification). The concentrations of NO_3^- in groundwater samples were relatively at low level with mean and standard deviation value at 1.80 ± 0.49 . The attenuation pattern observed for NO_3^- with respect to distance from the landfill is in an irregular pattern to undetectable below 0.02mg/L in the groundwater samples around the aquifer in BH5. Further analyses were carried out and test of significance of the observed correlation coefficient of data set of $\text{NH}_3\text{-N}$ and NO_3^- with some selected parameters as presented in Table 4

Table 4: Correlation coefficient between data set of NH₃-N and NO₃ with some selected Parameters from Taman Beringing Landfill.

Variables	NH ₃ -N	NO ₃ ⁻	Temp	pH	BOD	COD
NH ₃ -N	1	0.656	-0.7785	0.4081	0.1658	0.5491
NO ₃ ⁻		1	-0.1898	0.2726	-0.633	-0.2277

The result shows that out of the total of nine correlation found between the data sets of NH₃-N and NO₃ with some selected parameters (Temperature, BOD and COD), five were found to be positively correlated and four were negatively correlated (refer to Table 4). Highly negative correlation existed between NH₃-N and Temp ($r = -0.7785$).

In the case of Heavy metals parameters of the groundwater samples, Mn, Cu, Zn, occur in trace concentration, while Cd rarely occurred at concentrations large enough to be detected. The limit of detection (LOD) for Cd is < 0.001 . The acceptable level of 0.3, 0.01, 0.02mg/L for Fe, Pb and Ni respectively set by NDWQS in Malaysia was not met in some of the wells, therefore Fe, Ni, and Pb are considered to be the heavy metal contaminants of importance in the groundwater sample.

Table 5: Heavy metal characteristics of groundwater samples from Taman Beringin ex-Landfill and comparison with Malaysian National Drinking Water Quality Standard (NDWQS).

Samples	Distance (m)	Fe	Mn	Pb	Cu	Ni	Cd	Zn
NDWQS		0.3	0.1	0.01	1.0	0.02	0.003	3.0
BH3	169.73	0.047	0.053	0.013	0.013	0.012	ND	0.023
BH5	223.95	0.139	0.082	ND	0.044	0.018	ND	0.034
BH6	341.56	1.442	0.028	0.094	0.037	0.024	ND	0.487
BH7	396.68	0.028	0.077	0.019	0.010	0.010	ND	0.012
Mean \pm SD	282.98	0.41 ± 0.68	0.06 ± 0.024	0.042 ± 0.045	0.026 ± 0.017	0.016 ± 0.006	-	0.13 ± 0.23

Except otherwise stated, all units are in mg/L, SD-Standard Deviation, ND: Not detected

Discussion:

Elevated temperature of 29.1 ± 0.22 observed than the temperature of natural groundwater water is due to decomposition of landfill waste. In addition, a strong negative correlation between NH₃-N and Temp ($r = -0.7785$) shows that temperature influences the activity of NH₃-N. However, the general alkaline pH of 7.8 of raw surface leachate is characteristic of landfill in their methanogenic stage due to the age of the landfill which is greater than 10 year. This shows that solubilisation of the majority of organic components is near completion. Furthermore. The range of pH(8.07-8.50), BOD (560-1520mg/L) and COD (2050-5230mg/L) of raw leachates as recorded by Agamuthu [1] in Taman Beringin landfill were much higher compared to the range obtained for pH(7.6-7.8), BOD (60-116mg/L) and COD (309-586mg/L) of raw leachates in this present study. The declining in the concentrations of these parameters is purely due to the age of the landfill. However, the unsustainable biodegradability and the BOD/COD value of 0.37 and 0.33 in BH5 and BH6 respectively shows that there are still organic materials in the groundwater which are still biodegradable but some condition in the wastewater is probably inhibiting bacteria that uses organic. The exceptionally high concentration of Fe in BH5 (1.442mg/L) compared to the other wells is due to the anaerobic-reducing condition of the aquifer at BH5 (reduction of Fe³⁺ to Fe²⁺). High concentrations of redox sensitive species such as Fe and Mn can be attributed to reducing conditions [2]. This reducing condition in the landfill is supported by the undetectable NO₃ (< 0.02 mg/L) in groundwater samples around the aquifer in BH5. From the mentioned mean results of BOD/COD ratio of raw leachates (0.19) and groundwater samples (0.26), the leachate will be easy to treat biologically but is deemed to undergo a chemical treatment before the routine biological treatment due to heavy metal contaminant such as Fe, Pb and Ni.

Landfill greater than 10 years old are predominantly methanogenic and nitrogen based [4]. As pointed out in [8], during the active phase of the landfill at Taman Beringin, leachate was strong and methanogenic, and contained 4000-5000mg/l of COD, and in excess of 1000mg/l of NH₃-N. The level of NH₃-N in raw leachates recorded in this present study at 175.5 ± 73.3 shows that the parameters COD and NH₃-N are declining. However, the levels reaching the groundwater at 57.30-12.89 indicates the high degree of organic decomposition in the leachate deteriorating the ground water quality at TBL. Even though the levels of NH₃-N in the landfill are decreasing as presented in this study and the earlier studies as discussed in [1,4]. However the levels are still far above the acceptable limits for raw leachate and groundwater set at 0.3 and 0.5 mg/L. In contrast with other contaminants, NH₃-N occurs in all the wells. In particular, the levels of NH₃-N recorded in all the groundwater wells investigated at BH3 (12.89), BH5 (22.22), BH6 (32.01) and BH7 (57.30) were also far above the NWQS CLASS IIA set at 0.3mg/L and show undecending concentration decrease only over a few hundred meters away from the landfill although suggesting that nitrification of NH₃-N is occurring but only to some extent.

Implication of NH₃-N in receiving water to health and the environment:

There is no known health effect directly linked to ammonia toxicity therefore health-based guideline for human has not been established for NH₃ – N in drinking water [7]. However, it is vital to keep the levels of NH₃ – N/NH₄-N in drinking water at minimal level because ammonia compound in water can cause some dangerous diseases depending on the anion that it forms a compound with. Ammonium will form the ammonium chloride with chloride which increases the toxicity of NH₃ – N. It could also react with chlorine to create chloramines, which is toxic at concentration above the required level. As pointed out in [9] a concentration of 33.7 mg of ammonium ion per kg of body weight per day, ammonium chloride influences metabolism by shifting the acid–base equilibrium, disturbing the glucose tolerance, and reducing the tissue sensitivity to insulin. With the chloride level observed in some of the wells in Taman Beringin increases NH₃-N increases the risk of the toxicity when the groundwater flows into the river and pollute drinking water and other groundwater resources. Groundwater drawn from the wells at the landfill should be avoided. On the other hand acute exposure of NH₃ – N by animals may result in lung oedema, nervous system dysfunction, acidosis, and kidney damage while long term exposure may lead to decreases in bone mass, calcium content, and blood pH. [9]. The spread of the groundwater to river in and around the landfill area should be prevented in a responsible way because conditions such as Alkaline pH and warm water temperatures are condition that favors NH₃ – N toxicity are prevalent in the landfill. Aquatic (e.g fish) in receiving NH₃ – N contaminated water are at greater risk.

Furthermore, NH₃ – N in the environment can create a ripple effect in receiving water and further results to adverse effect on the environment through nitrification (under favorable pH and oxygen condition to form nitrate) to eutrophications. Thereby significantly, degrading the water quality and usability. Nitrification process is observed in all the wells at Taman Beringin with the exception of BH5.. The additional effects are decreases in the oxygen concentration and pH, which will raise the BOD and COD levels in the water which makes waste water treatment difficult. Decreases in oxygen level are as well detrimental to aquatic life. In addition high level of NH₃ – N in the environment is also a hindrance for removal of Manganese and reduction of chlorine disinfection efficiency in drinking water treatment and distribution systems.

Conclusion:

The groundwater wells beneath the Taman Beringin landfill have shown considerable impact of leachates with high concentration of NH₃-N and other major pollution indicator parameters: COD, BOD, TSS, TDS, and heavy metals(Pb, Ni, and Fe) which were above the stipulated NDWQS/ and CLASSII A range in Malaysia. Most important, there is a great tendency for an increase in groundwater contamination around the landfill location considering the present level of NH₃-N concentrations in the landfill which are underlining. As much as other contaminants is present in higher concentration than the stipulated standard in some of the wells in the landfill, the risk is there for NH₃-N degradation on the environment and toxicity to human and aquatic life around the landfill when the groundwater flows into the river and pollute drinking water and other groundwater resources. The dependency as pointed out in this study has been attributed to the anaerobic condition, extent of biodegradability of the organic waste and to some extent, the hydrogeology of the area. It is recommended that remediation should be done in a responsible way to prevent the existing leachates from further spreading and leakage into the river. A combined process of routine chemical treatment prior to biological treatment is also highly necessary to improve the quality of the leachate to minimize the effects on the surrounding environment.

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