

Sorption Parameters of Pb and Cu on Natural Clay Soils from Selangor, Malaysia

(Parameter Penjerapan Logam Pb dan Cu oleh Tanah Lempung dari Selangor, Malaysia)

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ABSTRACT

The sorption parameters of two heavy metals (i.e. Pb and Cu) in clay soils from Selangor were studied using batch equilibrium test. The test was conducted in two separate systems, i.e. single and mix solutions. The sorption isotherms data from this test were then used to calculate the sorption parameters, i.e. distribution coefficient (K_d) and maximum adsorption capacity (A_m). Langmuir sorption equation was used to model the sorption data with the correlation coefficients (r^2) higher than 0.6. The study has revealed that different soils have different sorption capacity for different heavy metals. The K_d values are proportional to the sorption capacity of the soils. The sorption of Pb and Cu in single solution is higher than in mix solution, due to the competition for sorption sites among heavy metals in mix solution. The K_d values for Pb in single solution ranging from 36.18 – 334.48 L/g and for Cu is 9.29-66.19 L/g. In mix solution, the K_d values for Pb and Cu are much smaller, ranging from 23.13-31.79 L/g and 3.95-18.53 L/g respectively. The A_m values in single solution for both Pb and Cu are ranging from 0.48-1.09 mg/g dan 0.18-0.70 mg/g respectively. While in mix solution, the values of A_m for Pb ranging from 0.20-1.11 mg/g and Cu within 0.18-0.60 mg/g.

Keywords: Adsorption; Langmuir; distribution coefficient; maximum adsorption capacity

ABSTRAK

Parameter penjerapan dua logam berat (Pb dan Cu) oleh tanah lempung di kawasan Selangor dikaji dengan menggunakan kaedah ujian keseimbangan berkelompok. Ujian dilakukan dalam dua sistem, iaitu sistem larutan tunggal dan sistem larutan campuran. Data isoterma penjerapan dari ujian ini digunakan untuk mengira parameter penjerapan, iaitu nilai pekali taburan (K_d) dan nilai penjerapan maksima (A_m). Persamaan penjerapan Langmuir digunakan untuk memodelkan data penjerapan dengan nilai pekali korelasi (r^2) lebih dari 0.6. Kajian ini menunjukkan tanah yang berbeza mempunyai kapasiti penjerapan yang berbeza untuk logam berat yang berbeza. Nilai K_d berkadar langsung dengan nilai penjerapan maksima. Penjerapan logam Pb dan Cu dalam larutan tunggal adalah lebih tinggi berbanding larutan campuran, disebabkan oleh kesan perebutan tapak perlekatan oleh logam berat dalam larutan campuran. Nilai K_d untuk Pb dalam larutan tunggal berjulat 36.18 – 334.48 L/g dan untuk Cu ialah 9.29-66.19 L/g. Dalam larutan campuran, nilai K_d untuk Pb dan Cu adalah lebih kecil, masing-masing berjulat dari 23.13-31.79 L/g dan 3.95-18.53 L/g. Nilai A_m dalam sistem larutan tunggal berjulat untuk Pb dan Cu masing-masing berjulat dari 0.48-1.09 mg/g dan 0.18-0.70 mg/g. Dalam sistem larutan campuran pula, nilai A_m untuk Pb berjulat dari 0.20-1.11 mg/g manakala untuk Cu berjulat dari 0.18-0.60 mg/g.

Kata kunci: Jerapan; Langmuir; nilai pekali taburan; nilai penjerapan maksima

INTRODUCTION

Soils have been widely used as engineered clay liner (or compacted liner) in *state-of-the-art* sanitary landfills to prevent leachate migration into the environment (Taha & Kabir 2005; Wan Zuhairi 2000). Soils have an ability to retard the migration of contaminant species in leachate. Soils are capable to adsorb inorganic pollutants (Buchter et al. 1989; Bunde et al. 1998; Anderson & Christensen 1988; Alumaa et al. 2001) and organic pollutants (Kim et al. 2000; Hiriart et al. 2000; Taha et al. 2003). There are two sorption parameters that will be discussed in this study, i.e. distribution coefficient (K_d) and maximum adsorption capacity (A_m). These values can be obtained by applying

the Langmuir equation on the adsorption data from batch equilibrium test. Many researchers have utilized the Langmuir equation to calculate the sorption parameters (Singh et al. 2001; Harter & Baker 1977; Taha & Depankar 1998; Wan Zuhairi 2000). The mobility of heavy metals in soil is described by a distribution coefficient, K_d . It is defined as the ratio of metal concentration in the solid phase to that in the liquid phase at equilibrium (Alumaa et al. 2001).

The distribution coefficient (K_d) is one of the input parameters in contaminant transport modeling to model the contaminant flow in groundwater. The K_d parameter is due to the contaminant sorption or attenuation, i.e. the

contaminants are adsorbed on to the active soil components such as clay minerals, carbonates and organic matter. This sorption processes will delay (i.e. retard) the movement of pollutants in soils.

This paper highlights the determination of K_d and A_m values using clay soils from active landfill sites in Selangor, Malaysia. These soil samples were subjected to 24 hours of shaking with heavy metals in two separate systems (i.e. single solution and mix solution). In addition, this paper also focuses on the competitive sorption among heavy metals and its effect on the K_d and A_m values.

MATERIALS AND METHODS

Five samples were collected from active landfill sites in Selangor, Malaysia namely weathered metasediments from Air Hitam Landfill in Puchong (AHQ), river alluvium clay (SSC) and lateritic soil (SSL) from Sungai Sedu Landfill in Banting and lateritic soil from Taman Beringin Landfill (TBL) and finally, weathered graphitic schist from Ampang Pechah (APS) (Figure 1). Prior to testing with contaminants, all samples were physically characterized using physical tests according to the British soil testing standard BS1377 (1990). Meanwhile, chemical tests performed consists of cation exchange capacity (CEC) determination, soil pH and clay mineralogy. CEC was conducted using ammonium acetate solution using a method described by ASTM D4319 (ASTM 1984). Clay mineralogy in soil was determined using X-Ray Diffraction (XRD) technique. These soils are currently being used as natural soil liner underneath the

landfill sites to protect the leachate from polluting the groundwater. All soils were tested using batch equilibrium test method. This test provides a quick method of estimating the contaminant retention capacity of any liner material. It represents an extreme test method, since experimental technique completely destroy the soil structure (Jessberger et al. 1997). Batch experimental techniques are relatively simple and inexpensive, and many experiments can be done simultaneously (Bunde et al. 1998).

Soil samples were air-dried, ground and sieved to obtain aggregates that are less than 2 mm. Batch equilibrium test was undertaken by mixing 4 g of soil with 40 ml of known concentration of heavy metal solution in the centrifuge tubes (USEPA 1992; Wan Zuhairi, 2000, 2003; Wan Zuhairi et al. 2004). In this study, nitrate salts of Pb and Cu were used as a test solution. Nitrate salts were chosen as test solution, because nitrate has poor ability to complex the metallic cations (Msaky & Calvet 1990). The test was carried out in two separate systems, i.e. single-solution and mix-solution. In mix-solution, all four heavy metals were mixed together (i.e. Pb + Cu + Ni + Zn) to study the effect of competition between heavy metals in soil. The soil-solution system was then shaken for up to 24 hours to achieve equilibrium. Subsequently, the tube was centrifuged and the supernatant was analysed for equilibrium concentration (C_{aq}) using Atomic Absorption Spectrophotometer (AAS). The concentration of heavy metals adsorbed onto the soil solution (C_{ads}) was then calculated using the formula as follow:



FIGURE 1. The landfill and sampling locations in Selangor, Malaysia

$$C_{\text{ads}} \square \frac{(C_o - C_{\text{aq}}) \cdot V}{(M \square 1000)} \quad (1)$$

where C_{ads} is the amount of contaminant removed from solution (mg/g), C_o is the concentration of contaminant in the solution before mixing with soil (mg/L), C_{aq} is the equilibrium concentration of contaminant left in the solution after the experiment (mg/L), V = Volume of solution in the centrifuge tubes (40 mL) and M is the mass of soil in the tubes (4 g).

The amount of contaminant removed from solution (C_{ads}) was then plotted with the equilibrium concentration (C_{aq}) to determine the sorption response curves (i.e. sorption isotherms). There are three models that have been widely used to represent the response curves; namely linear, Freundlich and Langmuir models. The decision to select the best model is based on the pattern of the sorption curves (Deutsch 1997).

Sorption can be defined as the interaction of a contaminant with a solid (Piwoni & Keeley 1990). The sorption data were nicely fitted using the Langmuir equation which is given by equation 2 below. According to Harter and Baker (1977), Langmuir equation has become well established in summarizing a mass of adsorption data from the batch experiment.

$$C_{\text{ads}} \square \frac{K_d \cdot A_m \cdot C_{\text{aq}}}{1 \square K_d \cdot C_{\text{aq}}} \quad (2)$$

where C_{ads} and C_{aq} are previously defined. A_m is maximum sorption capacity of the soil and K_d is distribution (partition) coefficient. The Langmuir equation can be rearranged into a linear form as shown in equation 3, after substituting $1/B$ with K_d . Then, by plotting C_{ads} on the y-axis and ($C_{\text{ads}}/C_{\text{aq}}$) on the x-axis, one can determine the value of $-B$ as the gradient of the slope and the value of A_m at the intercept (USEPA 1999). A linear form of equation (2) is given by

$$C_{\text{ads}} = -B (C_{\text{ads}}/C_{\text{aq}}) + A_m \quad (3)$$

K_d is given by $K_d = -1/B$. The K_d parameter is very important in estimating the potential for the adsorption of dissolved contaminants in contact with soil. As typically used in fate and contaminant transport calculations, K_d is defined as the ratio of the contaminant concentration associated with the solid to the contaminant concentration in the surrounding aqueous solution when the system is at equilibrium.

RESULTS AND DISCUSSIONS

The physico-chemical properties for all soil samples are summarized in Table 1. Soils AHQ, SSC and SSL possess high percentage of clay fraction compared to other soils where kaolinite is a dominant clay mineral present. The pH values of all samples are within an acidic range between 4-5. The CEC value is higher for soil SSL (19-23meq/100g). The CEC values for APS dan TBL are estimated to be lower as shown by the clay contents in these two soils. The sorption curves in Figure 2 were plotted between the amounts of Pb adsorbed from the Pb single-solution against concentration of Pb left in the solution. All curves show a nonlinear shape. The shape of adsorption isotherms depended on several factors and is not always linear (Msaky & Culvet 1990). The sorption curves show that the amount of Pb adsorbed by all soil samples increased with the increasing amount of Pb used in the experiment, especially at lower concentration of initial Pb. This is because at low concentration, clay particles tend to disperse due to the full development of the diffuse double layer, therefore clay particle surfaces in contact with the solution are at the maximum (Mc Laren et al. 1981; Mohamed et al. 1992).

All sorption curves showed linear pattern at lower concentration and became constant, i.e. reached the maximum adsorption (A_m) with increasing concentration of Pb. The sorption capacity was at maximum at lower concentration (i.e. the sorption sites were still available and enough for sorption at lower concentration), resulting higher sorption of Pb. The sorption curves that are plotted

TABLE 1. Physico-chemical characteristics of soil samples used in this study

Property	pH	CEC (meq/100g)	LL %	PI %	w_{opt}	ρ_{dry} Mg/m ³	Gs	Clay %	Clay mineralogy
AHQ	4.74-5.35	0.5-2	2.28-2.59	10-19	6.10-9.78	2.17-2.35	2.41-2.59	19-22	K>I>>M
SSC	4.01-4.04	13-19	2.28-2.58	11-12	6.7-6.8	2.32	2.28-2.58	33-38	K>>M
SSL	4.7-4.8	18.5-22.5	2.78	19	6.68	2.32	2.78	20.00	K>>I
APS	4.0-5.5	na	2.62-2.69	3-5	0.69-4.40	2.45-2.70	2.66-2.69	0	K>>I
TBL	4.5 – 5.0	na	2.64-2.65	4-5	2.50-4.70	2.44-2.57	2.64-2.65	4-8	K>>I

LL % = Liquid limit; PI % = Plastic limit; w_{opt} = optimum water content; r_{dry} = maximum dry density; g_{dry} = maximum dry unit weight; Gs = Specific gravity; AHQ = Air Hitam Landfill (Weathered metasediment) SSC = Sungai Sedu Estuarine Clay (Clay soils) TBL = Taman Beringin Landfill (Lateritic soils) SSL = Sungai Sedu Landfill (Lateritic soils) APS = Ampang Pechah Landfill (Graphitic Schist); CEC = cation exchange capacity; K = kaolinite; I = illite; M = montmorillonite; na = not available

close to y-axis indicate higher sorption capacity compared to curves which are located near the x-axis (Deutsch 1997). Therefore, from the sorption curves in Figure 2, it can be observed that AHQ soil has higher sorption capacity compared to SSL, SSC, TBL and APS. Based on the positions of the curves, the sorption capacity for all soils can be ranked as follow: AHQ>SSL>SSC>TBL>APS.

Similar behavior was observed for Cu as depicted in Figure 3. It is interesting to note a contrast sorption curve for SSL, where the amount sorbed increased with the increasing concentration of Cu in the solution (i.e. linear sorption). Other soils showed similar sorption curves, as previously described for Pb, where the sorption capacity

increased and became constant when the concentration of Cu is increased. Except for TBL, the sorption of Cu by these soils can be ranked as AHQ>SSC>SSL>APS.

Figure 2 and 3 indicate that AHQ is the best candidate for liner material and has better sorption for Pb and Cu compared to APS soil. The variation in sorption by these soils is due to discrepancy in their physico-chemical properties. Other studies undertaken by Wan Zuhairi (2000; 2001; 2003; Wan Zuhairi et al. 2004) on various soil samples from United Kingdom and other Malaysian soils indicated that soil's physico-chemical properties highly influenced the sorption. There is a positive correlation between the sorption capacity and basic properties of the

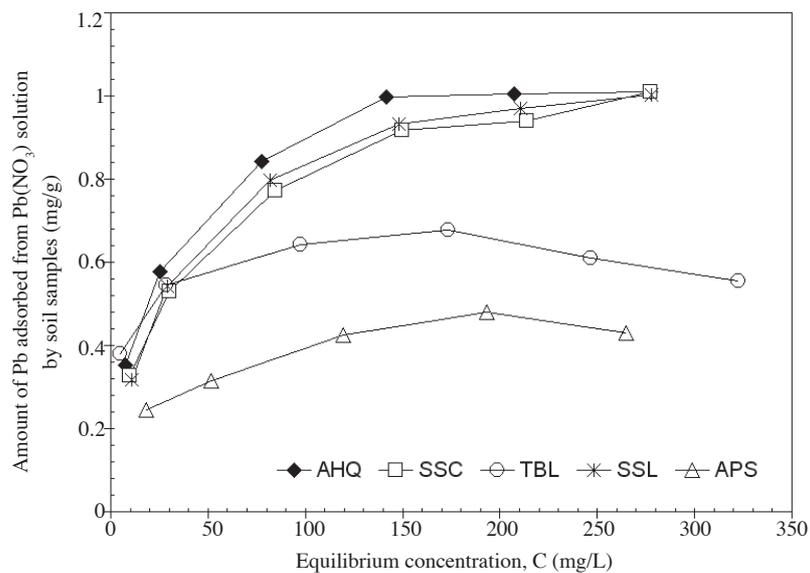


FIGURE 2. The amount of Pb removed by various soil samples per unit weight of soil from single nitrate solution (soil to solution ratio is 1:10)

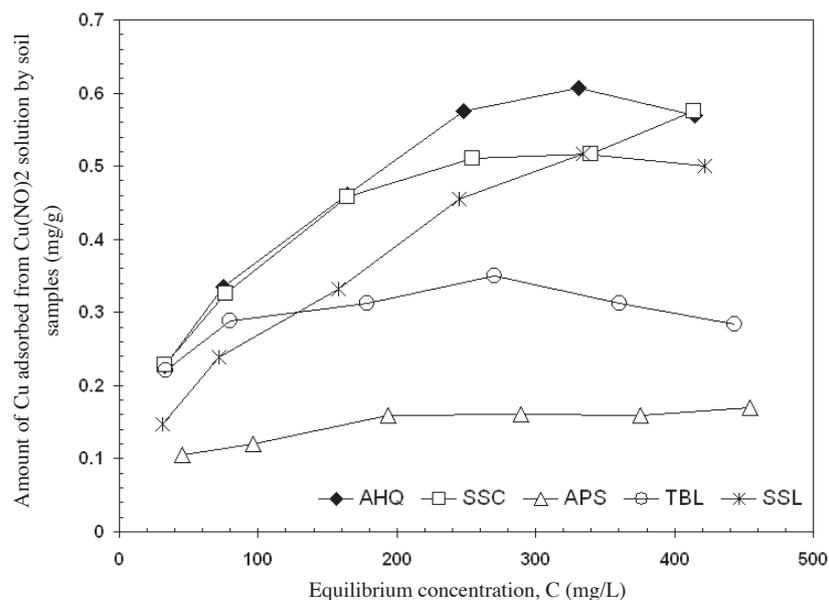


FIGURE 3. The amount of Cu removed by various soil samples per unit weight of soil from single nitrate solution (soil to solution ratio is 1:10)

soils from Wales, United Kingdom (Wan Zuhairi & Taha 2001). The adsorption maxima of soils are well correlated with the chemical, mineralogical and/or physical properties of the soils (Harter & Baker 1977). The clay content, pH, the amount of hydrous Fe and Mn oxides are significant factors in determining metal distribution (Anderson & Christensen 1988).

Figure 4 and 5 show the amount of Pb and Cu removed from mix-solution after 24 hours of shaking in batch equilibrium test. Both figures showed similar sorption behaviour as shown earlier by sorption curves in single-solution (Figure 2 and 3). The amount of Pb and Cu adsorbed increased with the increasing concentration of

Pb and Cu in mix-solution. The sorption curves were linear at lower concentration and approaching constant sorption at higher concentration, i.e. when the maximum adsorption was achieved. However, the maximum amount of Pb and Cu adsorbed by all soils in mix solution were relatively low compared to the amount sorbed in single-solution. Figure 6 highlights the difference on the amount sorbed between single and mix solutions for both Pb and Cu. The difference in sorption by these two systems is attributed mainly to the competition among heavy metals for sorption sites which are limited in soils. Heavy metals in mix-solution (i.e. four heavy metals Pb, Cu, Ni and Zn all mixed together) competed for a place onto the active soil

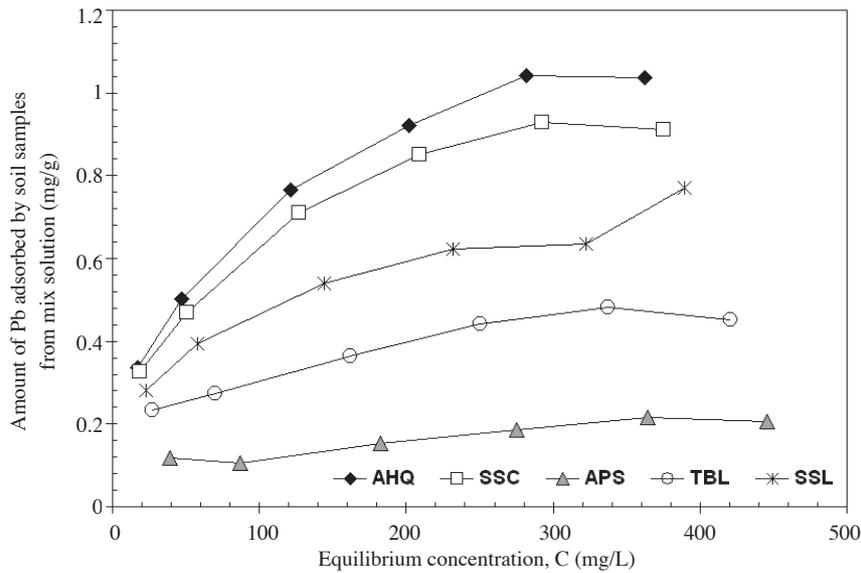


FIGURE 4. The amount of Pb removed by various soil samples per unit weight of soil from mix nitrate solution (soil to solution ratio is 1:10)

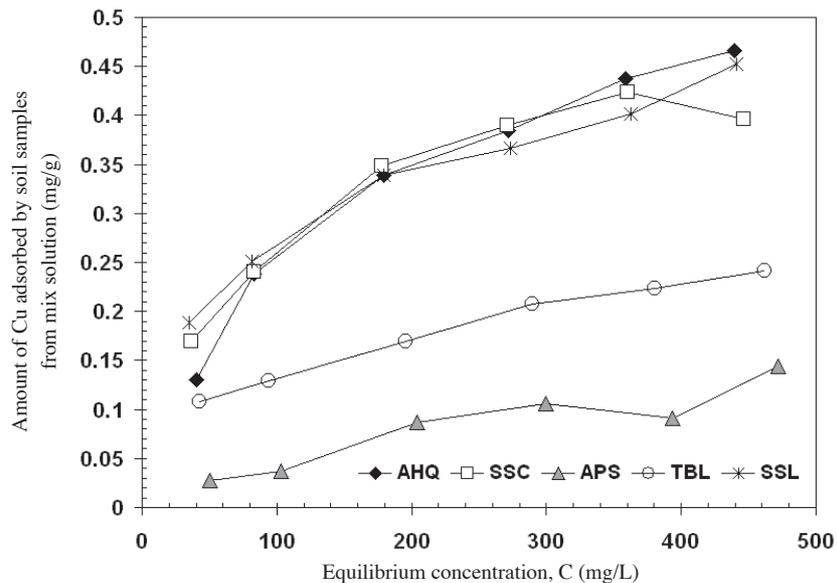


FIGURE 5. The amount of Cu removed by various soil samples per unit weight of soil from mix nitrate solution (soil to solution ratio is 1:10)

components (i.e. sorption sites). For example, the maximum Cu being adsorbed in single solution by soil AHQ (i.e. Figure 3) was 0.70 mg/g. Whereas in mix solution, the maximum amount sorption was 0.60 mg/g, a decrease of about 13.2%.

The sorption data for all samples can be best modeled using Langmuir equation (i.e. Equation 3). Figure 7 shows the Langmuir plot of Pb and Cu in single and mix solution

for soil AHQ. The graphs were plotted between C_{ads} (in y-axis) against C_{ads}/C_{aq} (in x-axis). The linear regression values (r^2) were higher than 0.89, indicating that the sorption data can be best fit using Langmuir's model. From the linear graphs in Figure 7, the parameters of K_d and A_m were calculated. Table 2 shows the K_d , A_m and r^2 values for both single and mix solutions for all soil samples. K_d values represent the retardation of pollutants by soils. High

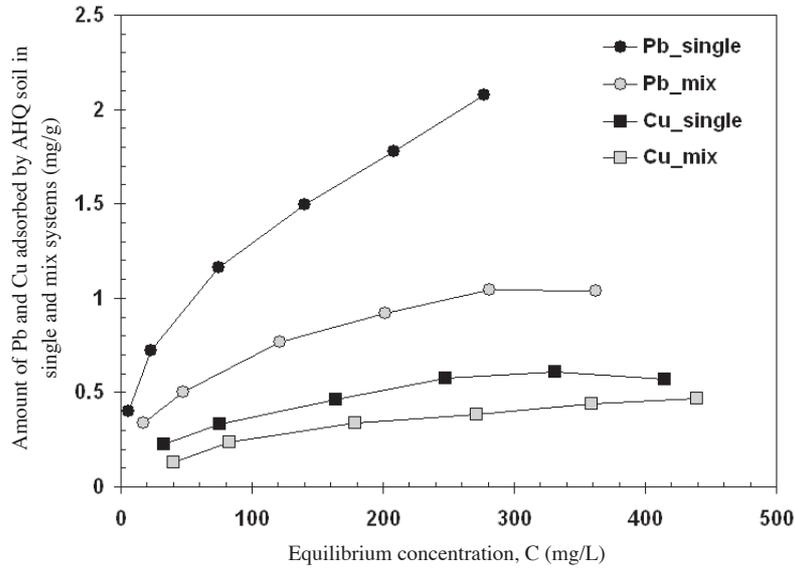


FIGURE 6. Pb and Cu removed from single and mix solutions (only soil AHQ is shown here)

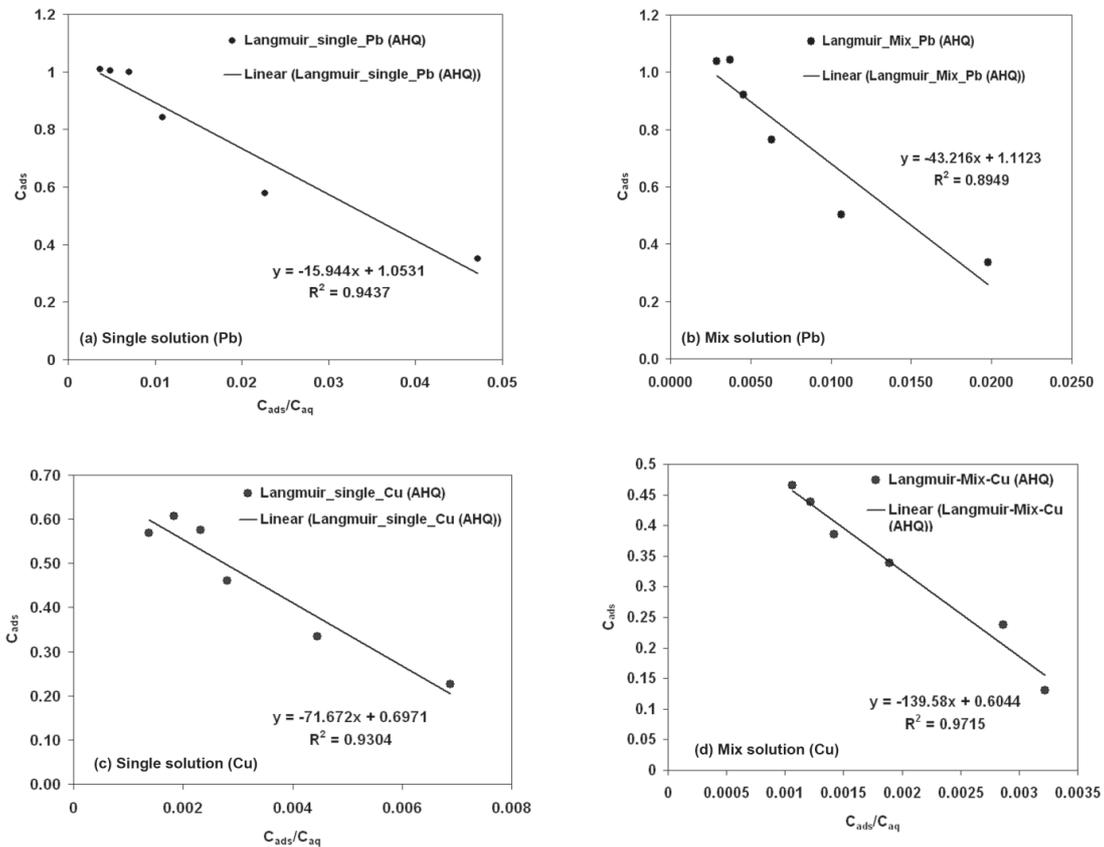


FIGURE 7. Langmuir plots between C_{ads} against C_{ads}/C_{aq} for Pb in single and mix solutions (only soil AHQ is shown here)

values of K_d indicate that heavy metals were retained by the soil solid through sorption reactions, while lower K_d indicate that most heavy metals remain in solution which is available for transport (Anderson & Christensen 1988).

Table 2 shows the correlation coefficients (r^2) higher than 0.6 except for APS soil in mix solution. K_d values that were calculated from single solution are higher compared to mix solution. The difference between these two K_d values was recorded for up to 90%. This indicates that in single solution, there is no competition for sorption, resulting higher K_d values (i.e. high amount of heavy metals being adsorbed by soil). Whereas, in the mix solution, there is intense competition for sorption sites, resulting less being adsorbed and reduce the values of K_d . The same results were also reported by Frost and Griffin (1977) after comparing the sorption results between pure nitrate salt and high levels of cations in leachate. They discovered

that K_d values were high in pure nitrate compared to multicationics in leachate.

One must be extra careful in selecting the K_d values in their transport model. The range of K_d values for Pb and Cu in single solution obtained from this study were from 36.18 – 334.49 L/g and 9.29 – 66.19 L/g, respectively. Lower range of K_d values were recorded from mix solution, i.e. 23.13 – 31.79 L/g for Pb and 3.95 – 18.53L/g for Cu. The highest K_d recorded in this study was for TBL soil, with K_d values for Pb and Cu are 334.5 L/g and 66.19 L/g, respectively. However, the K_d values for TBL soil do not correspond well with the sorption curves in Figure 2 and 3. One possible explanation is due to the fact that the particular sorption curve does not fit the Langmuir model.

Antoniadis and McKinley (2000) reported the K_d value for Zn in London Clay as 0.1 L/g. Taha and Depankar (1998) reported K_d value of Cd in granitic soil in Malaysia

TABLE 2. Langmuir isotherm parameters for all soil samples

Soils	Elements	Single solution			Mix solution		
		K_d (L/g)	A_m (mg/g)	r^2	K_d (L/g)	A_m (mg/g)	r^2
AHQ	Pb	62.72	1.0531	0.9437	23.13	1.1123	0.8948
SSC	Pb	44.03	1.0314	0.9419	24.48	0.9884	0.8950
APS	Pb	53.96	0.4786	0.8480	29.56	0.202	0.4873
TBL	Pb	334.49	0.626	0.8118	31.79	0.4743	0.7719
SSL	Pb	36.18	1.0918	0.9937	24.48	0.7412	0.8622
AHQ	Cu	13.95	0.6971	0.9304	7.16	0.6044	0.9715
SSC	Cu	16.46	0.6273	0.9583	14.25	0.4797	0.9441
APS	Cu	30.52	0.1773	0.8835	3.95	0.1755	0.3744
TBL	Cu	66.19	0.332	0.652	15.86	0.2489	0.7979
SSL	Cu	9.29	0.63	0.908	18.53	0.4577	0.8867

K_d = Distribution coefficient, A_m = Maximum sorption capacity; r^2 = correlation coefficient

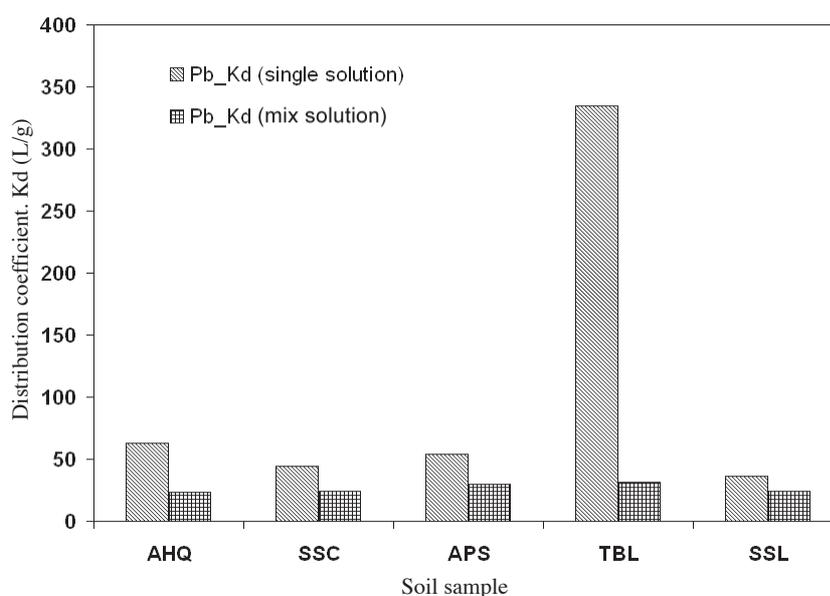


FIGURE 8. The comparison of K_d for Pb in single and mix solutions after 24 hours of shaking in batch experiment (soil to solution ratio is 1:10)

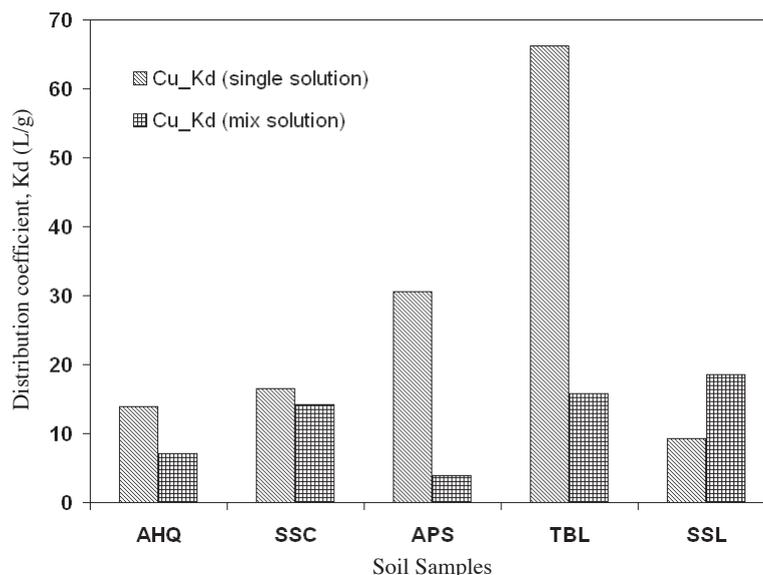


FIGURE 9. The comparison of K_d for Cu in single and mix solutions after 24 hours of shaking in batch experiment (soil to solution ratio is 1:10)

as 0.64×10^{-3} L/g. Small K_d reported in their study was due to the small initial concentration of Cd ranging only from 0.1-0.3 mg/L. Anderson and Christensen (1988) reported for Danish soils ranging from less than 0.01L/g to more than 1 L/g. The K_d values reported from the current study are somewhat higher compared to other researchers, but the values are still acceptable and within the range of K_d values reported by USEPA (1999). It is also interesting to note the values of maximum sorption capacity in soil (A_m). All A_m values reported in the Table 2 correspond well with the sorption curves in Figure 2, 3, 4, and 5.

Figure 8 and 9 shows the comparison between the K_d values for single and mix solutions. The K_d values are higher in single solution compared to mix solution due to the competition for sorption in mix solution, resulting decreasing amount of heavy metals adsorbed. The K_d values of TBL soil for Pb and Cu in single solution are far greater than in mix-solution, probably due to TBL sorption data that does follow Langmuir's equation. This argument can be supported using the sorption isotherm curves (in Figure 2 and 3), where TBL curves were plotted below AHQ, SSC and SSL. Further investigation is required to clarify this problem. Others however showed acceptable values of K_d .

CONCLUSIONS

The partition (distribution) coefficient, K_d is a parameter that is used to indicate the attenuation of contaminants when they migrate through the environment (i.e. groundwater). This contaminant transport parameter must be carefully estimated or tested using batch experiments or any other tests. This paper highlights the K_d values of five different soil samples in two different systems, i.e. single and mix solutions. The K_d values obtained in this study varies due to the variation in soil's physico-chemical

properties, different types of heavy metals and also due to the competition among those heavy metals in the solution. Higher K_d values are recorded in single solution compared to mix-solution. The K_d values for Pb and Cu in single solution ranged from 36.18 – 334.48 L/g and 9.29-66.19 L/g, respectively. In mix solution, the K_d values for Pb and Cu are much smaller, i.e. 23.13-31.79L/g and 3.95-18.53 L/g respectively.

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