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Isothermal Modelling of the Adsorption of Glyphosate onto Palm Oil Fronds Activated Carbon

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ABSTRACT

Glyphosate is a heavily usage herbicide in Malaysia. It inhibits amino acids synthesis in plants and microorganisms with the latter inhibition makes it difficult to find biodegradation microorganism as a bioremediation agent for this pesticide. One candidate for bioremediation is biosorption that has several positive aspects which include low operating expenses, very efficient detoxification of toxicants at low concentrations, low amount of disposal materials and does not need nutrient requirements as in bacterial-based remediation, the latter of which is limited by the presence of heavy metals and other toxicants. The biosorption of glyphosate on palm oil fronds activated carbon can be an efficient and low-cost tool for remediation of glyphosate. The absorption kinetics data of biosorption isotherm on the biosorption of glyphosate on oil palm fronds activated carbon were analyzed using modelled according to various models ranging from one to five parameters models such as Henry, Langmuir, Dubinin-Radushkevich, Freundlich, BET, Toth, Sips, Fritz-Schlunder IV, Baudu and Fritz-Schlunder V, and fitted using non-linear regression. Only the Henry, Langmuir, Freundlich, BET and Toth models can fit the data. Statistical analysis based on root-mean-square error (RMSE), adjusted coefficient of determination (adjR^2), bias factor (BF), accuracy factor (AF), corrected AICc (Akaike Information Criterion) showed that the Langmuir model is the best model. The calculated Langmuir parameters b_L value of 0.002 L/mg (95% confidence interval from 0.001 to 0.004) and q_{mL} value of 255.5 mg/g (95% confidence interval from 160.90 to 350.11).

INTRODUCTION

Glyphosate (N-(phosphonomethyl)glycine) is a common herbicide used primarily to kill weeds before the planting of fields, and for weed control in non-crop areas. Research showed an upward trend in glyphosate use worldwide within 1974-2014. By 2014, annual usage of glyphosate in the farm-sector increased to approximately 240 million pounds (~108.8 million kilograms) based on average annual crop use reported by the USDA's National Agricultural Statistics Service (NASS) [1]. The volume applied increased considerably due to higher rates of application in response to the development of glyphosate-resistant weeds and new, pre-harvest use patterns [2]. However, glyphosate transport to water bodies by spray-drift, run-off and leaching thereby causing harmful effects on non-target biota [3].

Glyphosate residue has been detected at elevated concentrations in both surface and groundwater and consequently in drinking water [4–7]. A recent meta-analysis study on data on glyphosate, AMPA and total phosphorus concentrations in surface waters in France showed that more than two-thirds of samples contained phosphorous with glyphosate, AMPA or both compounds [8]. Glyphosate prevalence in the environment has rising global concern of its ecotoxicological impacts on the environment, human and ecological systems [2,9–11]. Numerous studies linked the herbicide to a various health issue such as chronic kidney diseases [7] and cancer [9]. Hence, further research towards bioremediation of glyphosate in the environment is vital.

The existing method for removal of pesticides from water include biological and physicochemical treatment, advanced oxidation and combined treatment ([11,12]. However, the requirement of large infrastructure investments, expensive and specificity to particular contaminants are some of the major drawbacks of these methods. Biosorption is a metabolically passive process (made primarily by nonliving microorganisms or biological materials (e.g., agricultural waste) [13]. Biosorption is an efficient, selective, fast and economical process that makes the technique favorable for bioremediation of glyphosate in an aqueous environment. In addition, biosorption could be applied to many types of pollutant because the procedure could be performed in a wide range of temperature (4–90 °C) and pH (3–9) [14]

Several attempts have been made to remove glyphosate from aqueous solution by adsorption using different adsorbents which resulted to a various range of removal efficiencies i.e, three-dimensional graphene aerogels (578.0 mg g^{-1})[15],woody biochar (44mg/g) [16], biopolymer membrane (10.88 mg/g of membrane) [17] and water industrial residue (85.9 -113.6 mg/g) [18]. While some research has been carried out on glyphosate adsorption, only a few studies focused on adsorbent using agricultural biomass, [19–21] which also showed comparable results (~30 mg/g and 104.2 mg/g). Biosorption mechanisms are therefore various and, in some cases, still not very well understood [13]. Therefore, mechanisms involved in biosorption of glyphosate in different types of adsorbent remains a challenge.

Adsorption equilibrium information is the key point needed for a proper understanding of an adsorption process [22]. In order to understand the mechanism of glyphosate biosorption, the correct assignment of the kinetics and isotherms of biosorption is urgently needed. In many instances, a linearized from of an obviously nonlinear curve of these data is popularly reported in the literature. Linearization of nonlinear data disrupts the error structure of the data preventing and it is more difficult to estimate uncertainty, which is commonly shown in the form of a 95% confidence interval range [23]. In this study the published data from a glyphosate biosorption experiment on the oil palm frond [20] is remodeled with several more isotherms models (**Table 1**) and then regressed using nonlinear regression method and assessment of the best mode was carried out using various error function analysis.

Table 1. Isotherm models utilized in this study.

	Model	Formula	References
		Single-parameter model	
1	Henry's law	$q_e = HC_e$	[24]
		Two-parameter models	
2	Langmuir isotherm	$q_e = \frac{q_{mL} b_L C_e}{1 + b_L C_e}$	[25]
3	Freundlich isotherm	$q_e = K_F C_e^{\frac{1}{n_F}}$	[26]
4	Dubinin-radushkevich isotherm	$q_e = q_{mDR} exp\left\{-K_{DR}\left[RTln\left(1 + \frac{1}{C_e}\right)\right]^2\right\}$	[27,28]
		Three-parameter models	
5	Sips isotherm	$q_e = \frac{K_s q_{mS} C_e^{\frac{1}{n_S}}}{1 + K_s C_e^{\frac{1}{n_S}}}$	[29]
6	Toth isotherm	$q_e = \frac{q_{mT} C_e}{\left(K_T + C_e^{n_T}\right)^{n_T}}$	[30]
7	Bet isotherm	$q_e = \frac{q_{mBET} \alpha_{BET} C_e}{(1 - \beta_{BET} C_e)(1 - \beta_{BET} C_e + \alpha}$	[31]
		Four-parameter models	
8	Baudu isotherm	$q_e = \frac{q_{mB} b_B C_e^{(1+x+y)}}{1 + b_B C_e^{(1+x)}}$	[32]
9	Fritz-schlunder-iv isotherm	$q_e = \frac{A_{FS} C_e^{a_{FS}}}{1 + B_{FS} C_e^{b_{FS}}}$	[33]
		Five-parameter models	
10	Fritz-schlunder-v isotherm	$q_e = \frac{q_{mFS5} K_1 C_e^{\alpha_{FS}}}{1 + K_2 C_e^{\beta_{FS}}}$	[33]

MATERIALS AND METHODS

Data acquisition and fitting

Data from **Figure 1** from a published work [20] were digitized using the software Webplotdigitizer 2.5 [34]. Digitization using this software has been acknowledged for its reliability [35,36]. The data were then nonlinearly regressed using the curve-fitting software CurveExpert Professional software (Version 1.6).

Statistical analysis

Commonly used statistical discriminatory methods such as corrected AICc (Akaike Information Criterion), Bayesian Information Criterion (BIC), Hannan and Quinn's Criterion (HQ), Root-Mean-Square Error (RMSE), bias factor (BF), accuracy factor (AF) and adjusted coefficient of determination (R^2).

The RMSE was calculated according to **Eq. (1)**, [23], and smaller number of parameters is expected to give a smaller RMSE values. n is the number of experimental data, Ob_i and Pd_i are the experimental and predicted data while p is the number of parameters.

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(Pd_i - Ob_i)^2}{n-p}} \quad \text{(Eqn. 1)}$$

As R^2 or the coefficient of determination ignores the number of parameters in a model, the adjusted R^2 is utilized to overcome this issue. In the equation (**Eqns. 2** and **3**), the total variance of the y-variable is denoted by s_y^2 while RMS is the Residual Mean Square.

$$Adjusted\left(R^2\right) = 1 - \frac{RMS}{s_Y^2} \quad \text{(Eqn. 2)}$$

$$Adjusted\left(R^2\right) = 1 - \frac{\left(1-R^2\right)(n-1)}{(n-p-1)} \quad \text{(Eqn. 3)}$$

The Akaike Information Criterion (AIC) is based on the information theory. It balances between the goodness of fit of a particular model and the complexity of a model [37]. To handle data having a high number of parameters or a smaller number of values corrected Akaike information criterion (AICc) is utilized [38]. The AICc is calculated as follows (**Eqn. 4**), where p signifies the quantity of parameters and n signify the quantity of data points. A model with a smaller value of AICc is deemed likely more correct [38].

$$AICc = 2p + n\ln\left(\frac{RSS}{n}\right) + 2(p+1) + \frac{2(p+1)(p+2)}{n-p-2} \quad \text{(Eqn. 4)}$$

Aside from AICc, Bayesian Information Criterion (BIC) (**Eqn. 5**) is another statistical method that is based on information theory. This error function penalizes the number of parameters more strongly than AIC [39].

$$BIC = n.\ln\frac{RSS}{n} + k.\ln(n) \quad \text{(Eqn. 5)}$$

A further error function method based on the information theory is the Hannan–Quinn information criterion (HQC) (**Eqn. 6**). The HQC is strongly consistent unlike AIC due to the ln ln n term in the equation [38];

$$HQC = n \times ln\frac{RSS}{n} + 2 \times k \times ln(\ln n) \quad \text{(Eqn. 6)}$$

Further error function analysis that originates from the work of Ross [40] are the Accuracy Factor (AF) and Bias Factor (BF). These error functions test the statistical evaluation of models for the goodness-of-fit but do not penalize for number of parameter (**Eqns. 7 and 8**).

$$\text{Bias factor} = 10^{\left(\sum_{i=1}^{n}\frac{\log(Pd_i/Ob_i)}{n}\right)} \quad \text{(Eqn. 7)}$$

$$\text{Accuracy factor} = 10^{\left(\sum_{i=1}^{n}\frac{\log|(Pd_i/Ob_i)|}{n}\right)} \quad \text{(Eqn. 8)}$$

RESULTS AND DISCUSSION

The equilibrium data from [20] was analyzed using ten models—Henry, Langmuir, Dubinin-Radushkevich, Freundlich, BET, Toth, Sips, Fritz-Schlunder IV, Baudu and Fritz-Schlunder V, and fitted using non-linear regression. Only the Henry, Langmuir, Freundlich, BET and Toth models can fit the data whilst the rest did not converge and was abandoned (**Figs. 1-5**) Statistical analysis based on root-mean-square error (RMSE), adjusted coefficient of determination (adjR^2), bias factor (BF), accuracy factor (AF), corrected AICc (Akaike Information Criterion) showed that the Langmuir model is the best model (**Table 2**). This is the same to the published work [20] using linearized form. The calculated Langmuir parameters b_L value of 0.002 L/mg (95% confidence interval from 0.001 to 0.004) and q_{mL} value of 255.5 mg/g (95% confidence interval from 160.90 to 350.11) is different from the published work with a b_L and q_{mL} values of 104.2 mg/g and 0.054 L/mg, respectively [20]. The results from the published work has been improved in the form of the addition of a 95% confidence interval range which can statistically be used to discriminate models [23].

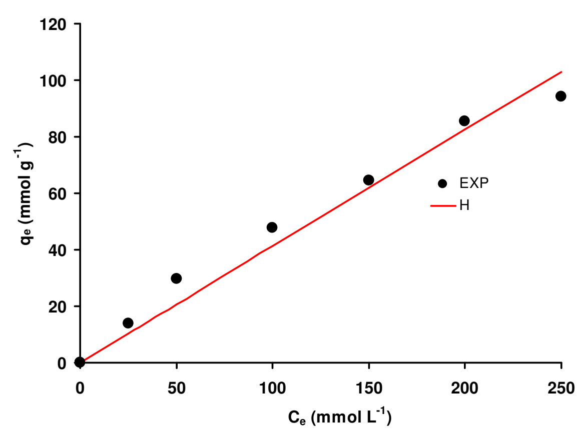

Fig. 1. Biosorption isotherm of glyphosate on palm oil fronds activated carbon as modelled using the Henry model.

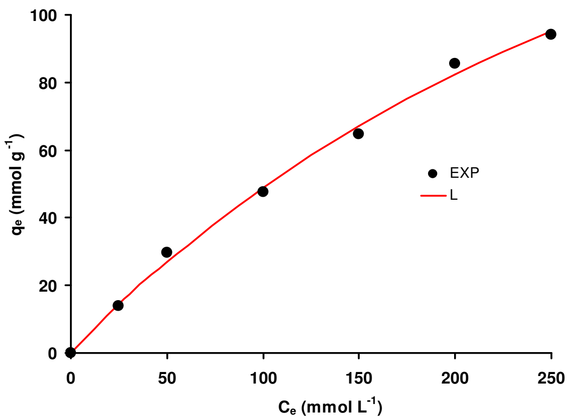

Fig. 2. Biosorption isotherm of glyphosate on palm oil fronds activated carbon as modelled using the Langmuir model

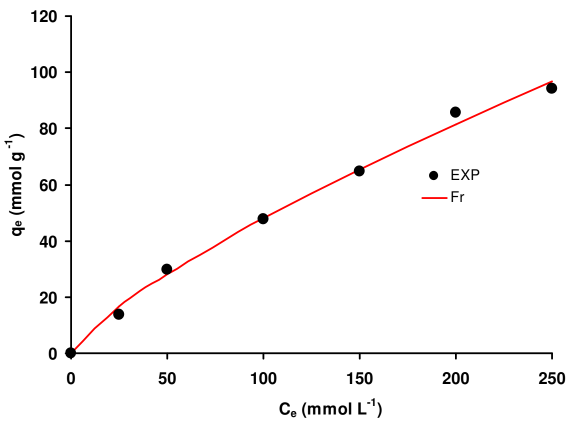

Fig. 3. Biosorption isotherm of glyphosate on palm oil fronds activated carbon as modelled using the Freundlich model.

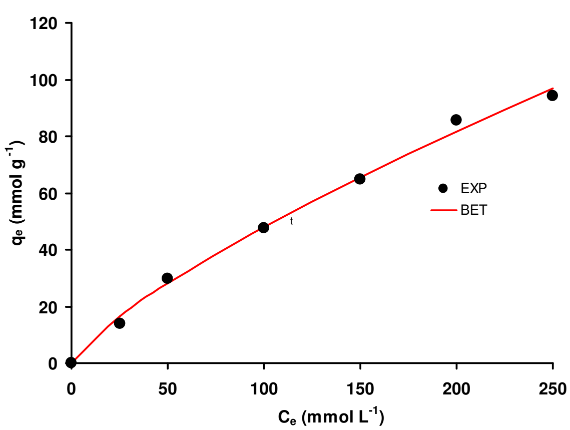

Fig. 4. Biosorption isotherm of glyphosate on palm oil fronds activated carbon as modelled using the BET model.

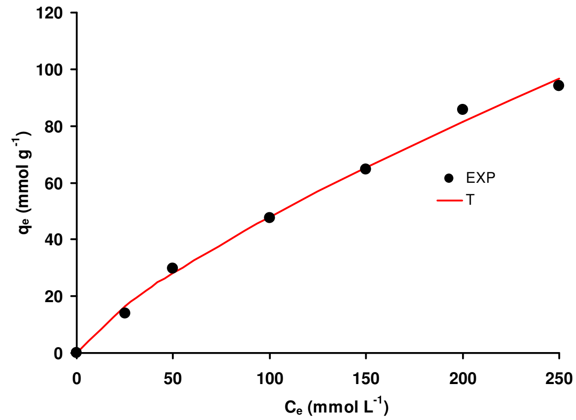

Fig. 5. Biosorption isotherm of glyphosate on palm oil fronds activated carbon as modelled using the Toth model.

Table 2. Error function analysis for the fitting of the isotherm of glyphosate on palm oil fronds activated carbon.

Model	p	RMSE	adR^2	AICc	BIC	HQC	BF	AF
Henry	1	6.25	0.968	33.57	26.52	25.9	0.8921	1.15
Langmuir	2	2.30	0.995	27.32	13.21	12.0	0.9969	1.03
Freundlich	2	2.57	0.993	28.88	14.77	13.5	1.0177	1.05
BET	3	2.57	0.993	43.32	15.16	13.3	0.9969	1.03
Toth	3	2.53	0.99	43.10	14.94	13.1	1.0034	1.04

Note:
RMSE Root mean Square Error
p no of parameters
adR^2 Adjusted Coefficient of determination
BF Bias factor
AF Accuracy factor
AICc Adjusted Akaike Information Criterion
BIC Bayesian Information Criterion
HQC Hannan–Quinn information criterion

The Langmuir isotherm is among the most cited best isothermal models to govern biosorption of xenobiotics such as the biosorption of Acid Orange 7 (AO7) and Remazol Black 5 (RB5) from aqueous solutions by canola stalks [41], malachite green adsorption onto copper nanowires loaded on activated carbon [42], the adsorption of betacyanin pigment onto the spun silk [43] and the adsorption of chlorothalonil by Nairobi River Sediment [44].

The Langmuir is also the best model in the biosorption of the pesticide 2,4-dichlorophenoxyacetic by modified jute [45] the biosorption of bromopropylate by activated carbons produced from agricultural residues (olive kernel, corn cobs, rapeseed stalks and soya stalks) [46], biosorption of paraquat by a hybdrid biopolymers consisting of κ-carrageenan and starch [47] and the biosorption of chlorpyrifos and monocrotophos by acid-treated palm shell powder [48]. On the other hand, in the biosorption of the butachlor pesticide by cantaloupe (*Cucumis melo*) seed shell powder Freundlich was found to be the best model [49].

CONCLUSION

In conclusion, the absorption kinetics data of biosorption isotherm on the biosorption of glyphosate on palm oil fronds activated carbon has been successfully analysed using modelled according to various models ranging from one to five parameters models such as Henry, Langmuir, Dubinin-Radushkevich, Freundlich, BET, Toth, Sips, Fritz-Schlunder IV, Baudu and Fritz-Schlunder V, and fitted using non-linear regression. Only the Henry, Langmuir, Freundlich, BET and Toth models can fit the data. Statistical analysis based on root-mean-square error (RMSE), adjusted coefficient of determination (adjR^2), bias factor (BF), accuracy factor (AF), corrected AICc (Akaike Information Criterion) showed that the Langmuir model is the best model. The calculated Langmuir parameters b_L value of 0.002 L/mg (95% confidence interval from 0.001 to 0.004) and q_{mL} value of 255.5 mg/g (95% confidence interval from 160.90 to 350.11).

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