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Modelling the Adsorption of Arsenic from Water and Wastewater using Microsoft Excel Solver and SPSS

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Abstract
Arsenic (As) poisoning is toxic to human organs and tissues (the heart, bones, intestines, kidneys, and reproductive and nervous systems). This paper focused on the modelling fittings of removal of As from water and typical industrial wastewaters through adsorption. Equilibrium isotherm parameters were determined using Microsoft Excel Solver (MES) and SPSS methods. The study revealed that equilibrium isotherms for As adsorption from aqueous solutions can be grouped into two based on the values of correlation coefficient (R^2) as follows: adsorption equilibrium isotherms with R^2 greater than 0.95 and R^2 less than 0.95. Microsoft Excel Solver method was found to be as accurate as SPSS method based MSC, CD, R and AIC. The study concluded that MES can be used for model fitting based on accuracy and cost (at no additional cost of purchase and installation unlike SPSS).

Keywords: Microsoft Excel Solver, model fittings, arsenic, equilibrium isotherm, adsorption.

Introduction
Statistical and mathematical modeling methods are generally used in clinical science, physical sciences, engineering, epidemiology, environmental design and health services. Researchers do analyze data collected in clinical trials, physical and biological sciences and engineering experiments as well as observational and protptype studies of current data sources are using statistical and mathematical models (Henley *et al.*,2019). Design, diagnostic, analysis and prognostic inferences from statistical and mathematical models are acute to researchers advancing science, clinical practitioners, engineering and in making critical decisions, and policy makers impacting the environmental, health care and engineering systems to improve quality of life and reduce both initial and operational costs. The reliability of these inferences relies not only on the quality, quantity, and completeness of these collected data, but on statistical and mathematical models validity. A major key component of instituting model validity is fitting the model parameters. Models, which can satisfactorily represent the true original process by which the data were generated, are known as the Data Generating Process (DGP, Henley *et al.*, 2019). These DGP models serve a significant communicative function by simplifying 'model transparency' (Henley *et al.*, 2019), which maintenances future scientific inquiry and more actual distribution of research findings. Statistical and mathematical models that more accurate, approximate the underlying DGP. DGP include methods for regression analysis that pertain to generalized linear models, generalized additive models, and the exponential family non-linear models. The exponential family non-linear models includes methods using supervised learning that are routinely used in machine learning (Henley *et al.*, 2019). There are vast literature on these subjects, which provides an overview of methods with guidance and references underscoring the critical nature of considering model specification as part of the development process as it describes model fit, model misspecification tests, and model selection tests, addresses data representation strategies, discusses automated model-building approaches and validation methods, and reviews predictive and classification measures (Henley *et al.*, 2019).

Analysis should be performed on specific variables model which predicts the response variable for each variable to determine fit, specification, and predictive performance. Essential to data interpretation, model fit is a measure of the discrepancy between the observed empirical distribution of the observations in the data set and the 'best-fitting' probability distribution computed from the

estimated probability model the specification of a parameterized model and the data, model parameters may be estimated to fit the model. Graphical and numerical methods are important tools that are recommended as part of the model development process.

Microsoft Excel Solver is an example of numerical method commonly use. Microsoft Excel Solver is an add-in, which involves numerical method for solving mathematical and statistical problems. Tay (2006) anticipated step-by-step instructions on how to utilize a Casio (Malaysia) fx-570 MS calculator in numerical methods, while Guerreri-Garcia and Santos-Polomo (2008) presented a collection of keystroke sequences helping in solving numerical methods. Tay (2006) had suggested solving numerical methods using the Casio fx-570MS calculator to overcome the tedium of doing recursive computations. Tay and Kek (2008) established a solver to solve systems of linear models and equations using Excel tool. Tay *et al.* (2014) offered solutions to nonlinear equations by Newton Raphson, but none of the literature and researches dealt with a system of nonlinear and linear equations in adsorption isotherm equilibria and statistical evaluations were not conducted by these studies. Suwannahong *et al.* (2021) studied the evaluation of the Microsoft Excel Solver Spreadsheet-Based program for nonlinear expressions of adsorption isotherm models, which was limited to non – linear models without linear expressions. More information on adsorption can be found in literature such as Wongcharee *et al.* (2017, 2018), Ng *et al.* (2014), Bastami and Entezari (2012), Cheng *et al.* (2012), Chincholi *et al.* (2014), Kumar and Sivanesan (2006), and Jasper *et al.* (2020). More explanations on Microsoft Exel Solver and its application can be established in literature such as Guerrero- Garcia *et al.(*2008); Barati (2013), Bhattacharjya (2010), Gay and Middleton (1971), Jewell (2001), Briti *et al*. (2013), Hossain *et al*. (2013), Tay *et al*. (2014), Oke *et al*. (2014, 2015a, 2015b, 2015c, 2016), Adekunbi *et al. (*2019), Huang *et al*. (2019), Lukman *et al.* (2020) and Umaru *et al.* (2021). This study focuses on curve fittings of linear and non-linear adsorption isotherm models that are applied in adsorption practice on complete data and presented statistical evaluations.

Materials and Methods

Powdered eggshells were used as adsorbent for arsenic removal from both raw and synthetic wastewaters (detailed in Oke *et al.*,2008). Adsorption equilibrium isotherms were determined were determined using equation (1).

$$q_e = \frac{(C_0 - C_e)}{M} V \quad (1)$$

Where: q_e is the adsorption capacity of the Powdered eggshell (PESL) at equilibrium (mg/g), C_0 is initial the concentration of arsenic in the solution (mg/l), C_e is the final concentration of arsenic in the solution at equilibrium (mg/l).

Parameters of standard adsorption isotherms were determined using both Statistical Package for Social Sciences (SPSS, standard method) and Microsoft Excel Solver. The standard adsorption isotherms (single, two, three, four and five parameters) were determined and evaluated using standard statistical methods (Model Selection Criterion, (MSC), Coefficient of Determination (CD), Akaike Information Criterion, (AIC) and Correlation Coefficient, (R)). Table 1 presents the standard adsorption standard equilibrium isotherms. Microsoft Excel Solver was used for the determination of the standard adsorption isotherm's parameters based on availability of the add-in software on Microsoft Excel at no additional cost.

Table 1: Adsorption equilibrium isotherm for single component aqueous solutions

Type	Relationship	Parameters	Sources
Langmuir	$q_e = \frac{a_L b_L C_e}{1 + a_L C_e}$	a_L and b_L	Oke *et al.* (2008); Olarinoye *et al.* (2011); Nameni et *al.*, (2008)
Freundlich	$q_e = K_f C_e^{1/N_f}$	K_f and N_f	Oke *et al.* (2008); Olarinoye *et al.* (2011); Nameni et *al.*, (2008)
Redlich–Peterson	$q_e = \frac{\alpha_t C_e}{1 + \beta_t C_e^{\gamma}}$	α_t, γ and β_t	Yasmin *et al.* (2009)
Temkin	$q_e = a_t + 2.3 b_t \log C_e$ $q_e = \frac{RT}{b_t} \ln(a_t C_e)$	a_t and b_t	Otun *et al.*, (2006 a and b); Yasmin *et al*. (2009)
Crombie-Quilty and McLoughin	$q_e = K_m \left(\frac{C_e}{M}\right)^{1/N_m}$	K_m and N_m	Oke *et al.*, (2008)
Sips	$q_e = \frac{\alpha_{ts} C_e^{\gamma_{ts}}}{1 + \beta_{ts} C_e^{\gamma_{ts}}}$	α_{ts}, γ_{ts} and β_{ts}	Oke *et al.* (2008); Olarinoye *et al.*, (2011); Nameni et *al.*, (2008)
Langmuir-Freundlich	$q_e = \frac{q_m K_t C_e^{K_{LF}}}{1 + K_t C_e^{K_{LF}}}$	q_m, K_{LF} and K_t	Oke *et al.*, (2008); Olarinoye *et al.* (2011); Nameni et *al.* (2008)
Toth	$q_e = \frac{q_m \alpha_{tt}^{\gamma_{tt}} C_e}{\left(1 + \alpha_{tt} C_e^{\gamma_{tt}}\right)^{\left(\frac{1}{\gamma_{tt}}\right)}}$	q_m, $\alpha_{tt,}$ and γ_{tt}	Oke *et al.*, (2008); Olarinoye *et al.* (2011); Nameni et *al.* (2008)
	$q_e = \frac{\alpha_{tt} C_e}{\left(\beta_{tt} + C_e^{\gamma_{tt}}\right)^{\left(\frac{1}{\gamma_{tt}}\right)}}$	$\alpha_{tt,}$ β_{tt} and γ_{tt}	Khashimova *et al.*, 2008
Radke-Prausnitz	$\frac{1}{q_e} = \left(\frac{\alpha_{rp}}{(C_e)}\right) + \frac{\beta_{rp}}{\left(C_e^{\gamma_{rp}}\right)}$	α_{rp}, γ_{rp} and β_{rp}	Oke *et al.*, (2008); Olarinoye *et al.*, (2011); Nameni et *al.*, (2008)
	$q_e = \frac{q_m \alpha_{rp} C_e}{\left(1 + \alpha_{rp} C_e^{\gamma_{tt}}\right)}$	α_{rp}, γ_{rp} and q_{rp}	Kim *et al.*, (2002)
Khan	$q_e = \frac{q_{mk} \alpha_k C_e}{\left(1 + \alpha_k C_e\right)^{\gamma_k}}$	α_k, γ_k and q_{mk}	Kim *et al.*, (2002)

Type	Relationship	Parameters	Sources
Fritz and Schhunder	$q_e = \frac{q_{mfs}\alpha_{fs}C_e^{\gamma fst}}{\left(1+\alpha_{fs}C_e^{\gamma fst}\right)}$	α_{fs}, γ_{fst} and q_{mfs}	Olarinoye *et al.*, (2011);
Linear	$q_e = K_p C_e$	k_p	Olarinoye *et al.*, (2011);
Loading ratio	$q_e = \frac{q_{mL}\left(b_{Ll}C_e\right)^{(1/k_l)}}{1+\left(b_{Ll}C_e\right)^{(1/k_l)}}$	$q_{mL,}$ b_{Ll} and k_l	Olarinoye *et al.*, (2011);
Four parameters	$C_e = \alpha_1 Q_e^{(\alpha_2 q_m+\alpha_3)}$	$\alpha_{1,}$ $\alpha_{2,}$ α_3 and q_m	Olarinoye *et al.*, (2011);
Dubinin–Radushkevich	$q_e = X_m Exp\left(-k\varepsilon^2\right)$	X_m, k and ε	Nameni *et al.*, 2008; Yasmin et al., (2009)
Statistical	$q_e = \left(\frac{kC_e+(kC_e)^2\Gamma_{ii}}{1+kC_e+(kC_e)^2\frac{\Gamma_{ii}}{2}}\right)$	k	Olarinoye *et al.*, (2011);
Halsey	$q_e = \left(\frac{k_H}{C_e}\right)^{\frac{1}{n_H}}$	k_H and n_H	Yasmin *et al*. (2009)
Koble - Corrigan	$q_e = \frac{\alpha_{kc}C_e^{\gamma_{kc}}}{\left(1+\beta_{kc}C_e^{\gamma_{kc}}\right)}$	$\alpha_{kc,}$ β_{kc} and γ_{kc}	Han *et al.* (2009)
Frenkel- Halsey-Hill	$q_e = Exp\left(\frac{\ln\left(K_{fhh}-\ln\left(C_e\right)\right)}{n_{fhh}}\right)$	$K_{fhh;}$ and n_{fhh}	Olaosebikan *et al.* (2022)
Harkin Jura	$q_e = \left(\frac{\left(A_{hj}\right)}{B_{hj}-\log\left(C_e\right)}\right)$	A_{hj} and B_{hj}	Olaosebikan *et al.* (2022)
Bauder	$q_e = \left(\frac{\left(q_{mb}\right)b_o C_e^{1+x+y}}{1+\left(C_e^{1+x}\right)}\right)$	b_o, q_{mb}, x and y $1+x+y<1$ And $1+x<1$	Olaosebikan *et al.* (2022)
Marczewk	$q_e = q_{ms}\left(\frac{\left(C_eK_{ms}\right)^{\alpha}}{1+\left(C_eK_{ms}\right)^{\alpha}}\right)^{\left(\frac{\beta}{\alpha}\right)}$	K_{ms}, q_{ms}, α and β	Olaosebikan *et al.* (2022)
Liu	$q_e = \left(\frac{B_{li}\left(C_eA_{li}\right)^{\frac{1}{n_{li}}}}{1+\left(C_eA_{li}\right)^{\frac{1}{n_{li}}}}\right)$	B_{li}, A_{li} and n_{li}	Suwannahong *et al*. (2021)
Fritz- Schhunder (5 parameters)	$q_e = \left(\frac{q_{5m}K\left(C_e\right)^{\alpha_{fs5}}}{1+\beta_{fs5}\left(C_e\right)^{\alpha_{fs5}}}\right)$	Q_{5m}, K, α and β	Olaosebikan *et al.* (2022)

The procedure used for Microsoft Excel solver can be summarized as follows (Umaru *et al.* 2021):

a. Excel solver was added in the Microsoft Excel,
b. Target $\left(\left(q_p - q_t\right)^2 = 0\right)$, operation and changing cells were set, where; q_p is the experimental adsorption capacity and q_t is the calculated adsorption capacity using the adsorption equilibrium isotherms; and
c. MES was allowed to iterate at 200 iterations with 0.005 tolerance (Figure 1).

The AIC was derived from the Information Criterion of Akaike (Idi *et al*. 2020). AIC allows a direct comparison among models with a different number of parameters. The AIC presents the information on a given set of parameter estimates by relating the coefficient of determination to the number of parameters.

The higher MSC value indicates higher the accuracy, validity and the good fit of the methods. MSC was computed using equation (2) as follows:

$$MSC = \ln\left(\frac{\sum_{i=1}^{n}\left(Y_{obsi} - \overline{Y}_{obs}\right)^2}{\sum_{i=1}^{n}\left(Y_{obsi} - Y_{cali}\right)^2}\right) - \frac{2p}{n} \quad (2)$$

where, Y_{obsi} is the As concentrations from the experimental study; $\overline{Y}_{obs}$ is the average As concentrations from the experimental study; p is the total number of fixed parameters to be estimated in the methods; n is the total number of As concentrations calculated, and Y_{cali} is the As concentration calculated using the methods.

The AIC was determined using Equation (3) as follows:

$$AIC = n\left(\ln\sum_{i=1}^{n}\left(Y_{obsi} - Y_{cali}\right)^2\right) + 2p \quad (3)$$

The coefficient of determination (CD) can be interpreted as the proportion of expected data variation that can be explained by the obtained data. Higher values of CD indicate higher accuracy, validity and good fitness of the device. CD and Correlation Coefficient (R) can be expressed using equations (4) and (5).

$$CD = \frac{\sum_{i=1}^{n}\left(Y_{obsi} - \overline{Y_{cali}}\right)^2 - \sum_{i=1}^{n}\left(Y_{obsi} - Y_{cali}\right)^2}{\sum_{i=1}^{n}\left(Y_{obsi} - \overline{Y_{cali}}\right)^2} \quad (4)$$

$$R = \sqrt{\frac{\sum_{i=1}^{n}\left(Y_{obsi} - \overline{Y_{cali}}\right)^2 - \sum_{i=1}^{n}\left(Y_{obsi} - Y_{cali}\right)^2}{\sum_{i=1}^{n}\left(Y_{obsi} - \overline{Y_{cali}}\right)^2}} \quad (5)$$

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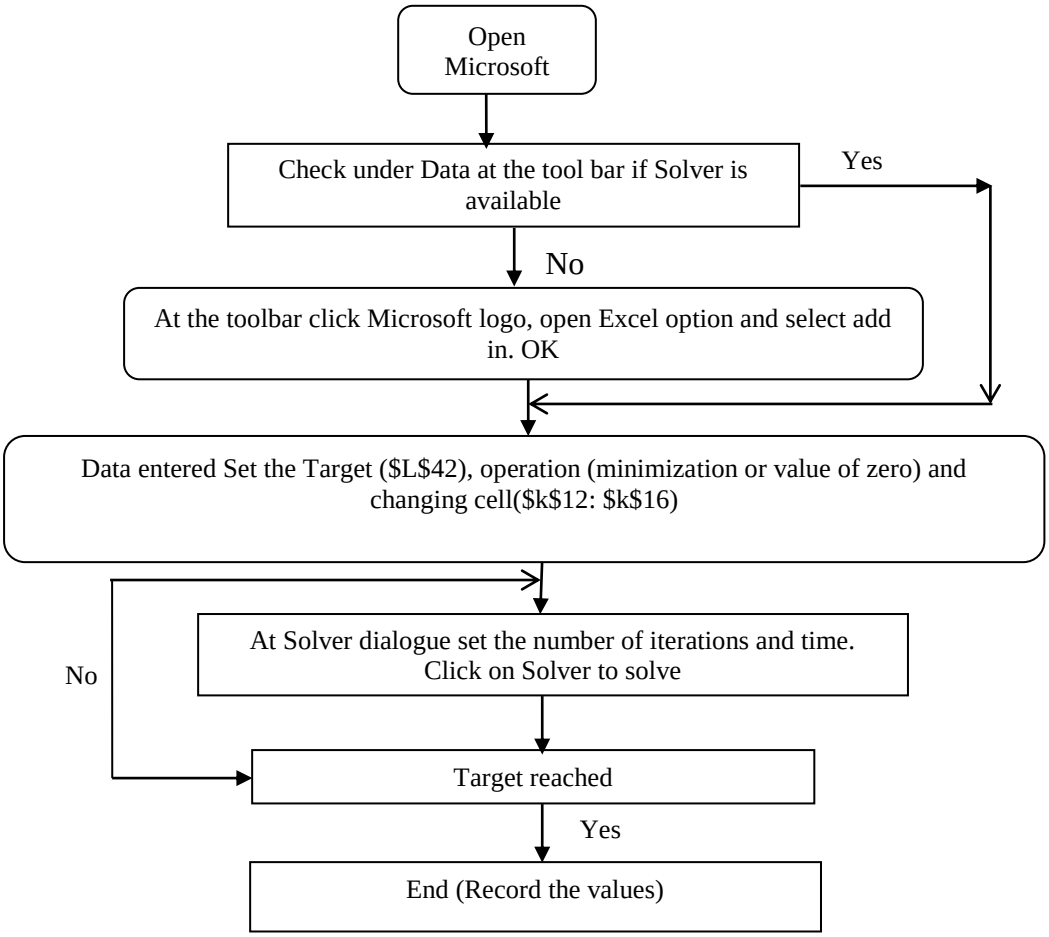

Figure 1: Flow chart of procedure for using Microsoft Excel Solver in the computation of the Isotherm's parameters

Results and Discussion

Table 2 presents the results of both synthetic wastewater and raw water. It presents statistical values (minimum, maximum, coefficient of variation, standard deviation and skewness) of C_e and q_e for both synthetic wastewater and typical raw water. Table 3 presents statistical evaluation of the two methods used (MES and SPSS). From Table 3, adsorption isotherm models of As onto PESL can be classified into four models as follows:

a) High accurate models: these are isotherm models with R equal or greater than 0.97, MSC > 3.03, CD > 0.97 and AIC < -18.46; these are Freudlich, Halsey, Temkin, Sips, Langmuir-Freudlich, Radke- Prausnitz1, Khan, Koble-Corigan Loading ratio, Liu, Fritz- Schlender (four and five parameters), Bauder, Marczewk, and Four parameters,
b) Medium accurate models: these are isotherm models with R less than 0.97, but greater than 0.87, 3.03 < MSC > 0.35,0.81 < CD > 0.90 and -9.47 < AIC < 7.10 These are adsorption equilibria isotherms such as Langmuir, Harins - Jura; and Radke - Prausnitz.
c) Average accurate models: these are isotherm models with R less than 0.87, but greater than 0.70, 0.35 < MSC > 0.14,0.7 < CD > 0.45 and -1.92 < AIC < -1.91; Redlich – Peterson, Toth 1 and 2
d) Low or poor accurate models: these are isotherm models with R less than 0.75, MSC < 0.45, CD < 0.51 and AIC > -1.92; This includes Linear.

From Table 3, averages of MSC, CD, R and AIC for these two methods were 2.618, 0.872, 0.927 and -16.420 and 2.153, 0.819, 0.882 and -13.630 for MES and SPSS methods, respectively. These results indicated that MES slightly performed better that SPSS, based on the values of average MSC (2.618 > 2.153), CD (0.872 > 0.819), R (0.972 > 0.880) and AIC (-16.420 < -13.630).

Table 2: Results of Synthetic Wastewater and Raw Water

Experiments	Synthetic Wastewater		Raw Water	
	q_e	C_e	q_e	Ce
1	0.364	0.288	2.508	1.649
2	0.424	0.299	3.313	1.727
3	0.707	0.321	4.933	1.789
4	0.977	0.360	6.971	1.877
5	1.115	0.385	2.937	1.688
6	1.321	0.399	9.756	1.880
Average	0.818	0.342	5.070	1.768
Standard Deviation	0.385	0.046	2.821	0.097
Coefficient of Variation (%)	47.042	13.532	55.643	5.500
Skewness	0.006	0.074	1.041	0.129
Minimum	0.364	0.288	2.508	1.649
Maximum	1.321	0.399	9.756	1.880

Table 3: Statistical Summary of the two methods

Adsorption Equilibrium Isotherms	MES				SPSS			
	MSC	CD	R	AIC	MSC	CD	R	AIC
Linear	0.35	0.50	0.71	-1.92	0.35	0.50	0.71	-1.92
Langmuir	0.35	0.92	0.96	-1.90	0.35	0.92	0.96	-1.91
Freudlich	3.24	0.97	0.99	-19.24	3.24	0.97	0.99	-19.24
Halsey	3.24	0.97	0.99	-19.24	3.24	0.97	0.99	-19.23
Temkin	4.28	0.99	1.00	-27.39	3.95	0.99	0.99	-25.38
Harkins-Jura	1.25	0.79	0.89	-9.16	1.30	0.81	0.90	-9.47
Redlich- Peterson	0.35	0.50	0.71	-1.92	0.35	0.50	0.71	-1.92
Sips	3.24	0.97	0.99	-19.24	3.19	0.97	0.99	-18.94
Langmuir- Freudlich	3.77	0.98	0.99	-22.42	3.39	0.98	0.99	-20.12
Radke- Prausnitz 1	3.24	0.97	0.99	-19.24	2.99	0.96	0.98	-17.74
Radke- Prausnitz 2	1.05	0.75	0.87	3.09	0.39	0.61	0.78	7.10
Khan	3.18	0.97	0.99	-18.89	3.11	0.97	0.98	-18.46
Koble- Corrigan	3.77	0.98	0.99	-22.42	3.20	0.97	0.99	-19.01
Loading Ratio	3.77	0.98	0.99	-22.42	3.77	0.98	0.99	-22.41
Toth 1	0.35	0.50	0.71	-1.92	0.35	0.50	0.71	-1.91
Toth 2	0.35	0.50	0.71	-1.92	0.35	0.50	0.71	-1.92
Liu	3.77	0.98	0.99	-22.42	3.77	0.98	0.99	-22.42
Fritz- Schulender	3.43	0.98	0.99	-20.36	3.55	0.98	0.99	-21.08
Bauder	4.09	0.99	0.99	-24.36	4.09	0.99	0.99	-24.36
Marczewk	4.25	0.99	0.99	-25.28	-2.14	-0.01	0.10	13.06
Four Parameters	3.03	1.00	1.00	-43.42	1.35	1.00	1.00	-33.35
Fritz- Schulender	3.24	0.97	0.99	-19.24	3.24	0.97	0.99	-19.24
Average	2.618	0.872	0.927	-16.420	2.153	0.819	0.882	-13.630

Tables 4, 5, 6 and 7 show the results of ANOVA for the evaluations. Table 4, which is ANOVA results for MSC revealed that there was no significant difference between the methods (MES and SPSS) at the 95 % confidence level ($F_{1,21}$ = 2.50; p =0.129), but between the adsorption isotherm models there were significant difference ($F_{21, 21}$ = 4.41; p = 0.001). Table 5, which is ANOVA results for CD revealed that there was no significant difference between the methods (MES and SPSS) at the

95 % confidence level ($F_{1,21}$ =1.35; p = 0.258), but between the adsorption isotherm models there were significant difference ($F_{12,21}$ = 3.71; p = 0.002). Table 6, which is ANOVA results for R revealed that there was no significant difference between the methods (MES and SPSS) at the 95 % confidence level ($F_{1,21}$ = 1.23; p = 0.280), but between the adsorption isotherm models there were significant difference ($F_{21,21}$ = 2.06; p = 0.053). Table 7, which is ANOVA results for AIC revealed that there was no significant difference between the methods (MES and SPSS) at the 95 % confidence level ($F_{1,21}$ = 2.50; p =0.129), but between the adsorption isotherm models there were significant difference ($F_{21,21}$ = 6.80; p = 0.000). The results from these tables agree with the literature (Nameni *et al.,* 2008; Wongcharee *et al., 2017;* 2018; Henley *et al.,* 2019; Jasper *et al.,* 2020; Suwannahong *et al.,* 2021; Olaosebikan *et al.,* 2022), which highlighted that adsorption equilibrium isotherm models are significantly different in expressions and in magnitude.

Table 4: Statistical evaluation of MSC

Source of Variation	*Sum of Squares*	*Degree of Freedom*	*Mean sum of square*	*F-value*	*P-value*	*F critical*
Within adsorption equilibrium isotherms models	88.16	21	4.20	4.41	0.001	2.08
Between MES and SPSS	2.38	1	2.38	2.50	0.129	4.32
Error	20.00	21	0.95			
Total	110.54	43				

Table 5: Statistical evaluation of CD

Source of Variation	*Sum of squares*	*Degree of freedom*	*Mean sum of square*	*F-value*	*P-value*	*F critical*
Within adsorption equilibrium isotherms models	1.78	21	0.08	3.71	0.002	2.08
Between MES and SPSS	0.03	1	0.03	1.35	0.258	4.32
Error	0.48	21	0.02			
Total	2.29	43				

Table 6: Statistical evaluation of R

Source of Variation	*Sum of squares*	*Degree of freedom*	*Mean sum of square*	*F-value*	*P-value*	*F critical*
Within adsorption equilibrium isotherms models	0.79	21	0.04	2.06	0.053	2.08
Between MES and SPSS	0.02	1	0.02	1.23	0.280	4.32
Error	0.38	21	0.02			
Total	1.20	43				

Table 7: Statistical evaluation of AIC

Source of Variation	*Sum of squares*	*Degree of freedom*	*Mean sum of square*	*F-value*	*P-value*	*F critical*
Within adsorption equilibrium isotherms models	4893.38	21	233.02	6.80	0.000	2.08
Between MES and SPSS	85.64	1	85.64	2.50	0.129	4.32
Error	720.09	21	34.29			
Total	5699.11	43				

Conclusion

The study evaluated two methods (Microsoft Excel Solver and SSPS) through the use of adsorption equilibrium isotherm and adsorption of Arsenic on powered eggshell. The study concluded based on the findings that:

a) MES performance was better than SPSS based on the MSC, CD, R and AIC
b) MES is better than SPSS based on no additional costs (purchase and installation costs) and
c) Procedures needed for the use of MES are simple and can be utilized on any computer with Microsoft Excel.

It is therefore recommended that other adsorption equilibrium isotherm models should be evaluated using MES, SPSS and other related software.

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