



Autocorrelation Test for the Residual Data from the Pseudo-1st Order Kinetic Model of the Brominated Flame Retardant 4-Bromodiphenyl Ether Adsorption onto Biochar-immobilized *Sphingomonas* sp.

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HISTORY

Received: 15th May 2021
Received in revised form: 24th June 2021
Accepted: 4th July 2021

KEYWORDS

polybrominated diphenyl ethers adsorption
biochar-immobilized bacteria
autocorrelation
Durbin-Watson statistic

ABSTRACT

Because of their fire-retardant properties, polybrominated diphenyl ethers (PBDEs) are frequently used in the manufacturing industry. PBDEs are mixed with polymers as additives and employed in a range of sectors, including plastics and textiles. They are, nevertheless, capable of leaking from the surfaces of these items and into the environment since they are not chemically connected to plastics or textile materials. The adsorption of PBDEs onto biochar-immobilized bacteria is a useful method to remediate PBDEs from the environment. Understanding the kinetics of adsorption can be done by using models such as pseudo-1st or pseudo-2nd. The pseudo-1st order kinetic model was previously found to best fit the data via a nonlinear regression exercise for brominated flame retardant 4-bromodiphenyl ether adsorption onto biochar-immobilized *Sphingomonas* sp. However, the use of nonlinear regression requires the residual of the fitted curve to be non-autocorrelated. The Durbin-Watson statistic, which is derived from the Durbin-Watson distribution, is one of the most commonly used ways for determining whether or not there is autocorrelation. In this study, the calculated value of the Durbin-Watson statistics was $d = 2.260$. The Durbin-Watson's lower critical value for dL was 0.700, while the upper critical value dU was 1.252. Since the d value was greater than the upper critical value or dU , this resulted in the null hypothesis not being rejected or indicating that there is no evidence of autocorrelation. This demonstrates that the pseudo-1st model used in the nonlinear regression model is adequate.

INTRODUCTION

PBDEs have been found in a wide range of environmental media, including soils, water, sediments, and even the air. Polybrominated diphenyl ethers (PBDEs) can biomagnify in food webs due to their strong lipotropy, posing a significant risk to human health. Furthermore, several PBDEs are dangerous and persistent in the environment due to their aromatic structures and bromide substituent groups [1–11]. To protect human health and the environment from polybrominated diphenyl ether (PBDE) pollution, these compounds in industrial effluent must be treated before being released into the environment. Adsorption is the most widely used treatment technology for removing contaminants from industrial wastewater because of its multiple advantages, which include simplicity, high efficiency, and ease of application. Another key adsorption challenge is the selection of efficient and cost-effective adsorbents, and many different

materials have been examined in prior research initiatives to solve this issue. The current study is focusing on biochar (a type of charcoal made from biomass pyrolysis) as a potential low-cost adsorbent for sequestering toxins and reducing pollution spread [11–21].

A biochar-based soil amendment, biochar can limit the biological uptake, storage, and absorption of organic contaminants, hence reducing the danger to the environment. 4-Bromophenyl phenyl ether (4-BE) and other brominated flame retardants (BFRs) have been utilised in a variety of consumer and commercial products for many years, including clothes and furniture. Since then, they have advanced to the status of a top-priority environmental pollutant on a global scale, and they have been discovered in the tissues of practically everyone who has been tested thus far [22–29]. The chemical 4-Bromophenyl phenyl ether has been detected in raw drinking water, mineral

water, and river water in other parts of the world. The United States Environmental Protection Agency (USEPA) recommends that the absolute maximum allowed level to protect freshwater aquatic life be 6.2 ug/L to protect aquatic life. When tested on the aquatic organism *Daphnia magna* (Water flea), the concentration that causes 50% fatality (LC50) was found to be 0.36 mg/L/48 hours [30]. (4)-BDE is another refractory top priority pollutant in which a study using activated sludge microbes indicated that it must not be appreciably reduced in the least; a subsequent study under aerobic circumstances demonstrated that it degrades at extremely low levels.

The degree of correlation (similarity) between two or more adjacent observations is measured by autocorrelation, which is defined as A variable's association with itself through time and space is measured by spatial autocorrelation, which can be either negative or positive depending on the variables involved. It is observed when undifferentiated values are found close to one another that there is negative spatial autocorrelation; on the other hand, positive spatial autocorrelation is observed when distinct values are discovered close to one another that there is positive spatial autocorrelation. Its properties and calculations are, however, frequently misinterpreted and distorted, even though it is a key theory in spatial statistics [31–35]. It has several pros as well as downsides. However, while it has advantages in that it allows for spatial interpolation, it also has downsides in that it makes statistical testing more complicated.

Temporal autocorrelation is an extension of this concept, however, it is a little more difficult to comprehend and implement than spatial autocorrelation. The time that simply moves in one direction is taken into consideration in temporal autocorrelation, whereas things with complicated shapes and more than two dimensions are taken into consideration in spatial autocorrelation, were recognising what is close by might be difficult to determine [36]. When a variable's structured spatial variation in a dataset is measured, it is referred to as organised spatial variation. Observed in areas that are close to one another and have values of variables that are indistinguishable from one another is positive spatial autocorrelation, which is a positive correlation between two or more variables. Because of this, the values that are adjacent to each other do not have the same value as one another whenever there is a negative spatial autocorrelation [37–43].

In normal nonlinear regression, the least-squares technique is used to ensure that data points do not rely on one another and that the value of a data point is not affected by the value of data points that came before or after it in the process. During the most extreme form of autocorrelation, temperature drift happens continuously throughout the duration of time measurements, and this drift has an impact on the findings of the measurements because they appear in a sequence of visually discernible patterns. Another example is a spectrophotometer that has been misused and has a tungsten light source attached to it.

In some situations, such as when the number of creatures that appear each year in a specific area is highly associated with and dependent on the number of creatures that appeared the year before, autocorrelation cannot be avoided. For example, when the number of creatures that appear each year in a specific area is highly associated with and dependent on the number of creatures that appeared the year before [44].

Among the most often employed approaches for determining whether or not there is autocorrelation is the Durbin–Watson statistic, which is derived from the Durbin–Watson distribution. When determining the level of significance in this method, the researchers employ Draper and Smith's strategy for calculating the level of significance [45–47]. In this study, the Durbin-Watson test was used to determine whether or not the pseudo-1st order kinetic model from a previously published work [48] for the adsorption of the brominated flame retardant 4-bromodiphenyl ether onto biochar-immobilized *Sphingomonas* sp. was adequate in terms of autocorrelation.

MATERIALS AND METHODS

Acquisition of Data

Residual data were acquired from a previously published work [48] from the pseudo-1st order Kinetic (2 regressors) modelling of adsorption of the brominated flame retardant 4-bromodiphenyl ether onto biochar-immobilized *Sphingomonas* sp.

Durbin-Watson test

In the Durbin–Watson test, a statistical calculation is carried out to test for the level of significance [46].

$$d = \frac{\sum_{t=2}^T (\hat{e}_t - \hat{e}_{t-1})^2}{\sum_{t=1}^T \hat{e}_t^2} \quad (\text{Eqn. 1})$$

In this test, the usual hypothesis where $H_0: \rho = 0$ versus the alternative $H_1: \rho > 0$ is performed. The statistic is approximately equal to $2(1 - p)$. When the value is zero, the Durbin-Watson test statistic is 2, and when the value is one, the Durbin-Watson test statistic is 0. Non-autocorrelation was indicated by a d value near 2, while positive autocorrelation was indicated by a d value around 0. Negative autocorrelation is shown by d values approaching 4 (Eqn. 1).

When the Durbin-Watson test statistics are low, the null hypothesis should be rejected because it indicates the presence of autocorrelation. Because there is no distribution of the -value in the Durbin-Watson test statistics associated with d, such as the t- or z-statistics, tables must be used in hypothesis testing.

The decision rule for the Durbin-Watson bounds test is

- if $d >$ upper bound, fail to reject the null hypothesis of no serial correlation, or there is no autocorrelation.
- if $d <$ lower bound, reject the null hypothesis and reach the conclusion that positive autocorrelation exists.,
- if lower bound $< d <$ upper bound, the test is inconclusive

RESULTS AND DISCUSSION

The calculated value of the Durbin-Watson statistics (Table 1) was $d = 2.260$. The statistic is approximately equal to $2(1 - p)$. We then test the hypothesis $H_0: \rho = 0$ versus the alternative hypothesis of $H_1: \rho > 0$. From the Durbin-Watson table [45,49] for 2 parameter models ($k'=2$) the lower critical value for dL was 0.700, while the upper critical value dU was 1.252. According to this, the d value was greater than the upper critical value or dU, resulting in the null hypothesis not being rejected or indicating that there is no evidence of autocorrelation. This demonstrates that the pseudo-1st model used in the nonlinear regression model can be adopted.

Table 1. The calculation for Durbin Watson statistics.

et	et ²	(et - et-1) ²
0.00E+00	0.00E+00	0.00E+00
1.17E-03	1.36E-06	1.36E-06
-2.67E-04	7.10E-08	2.06E-06
-2.60E-03	6.75E-06	5.44E-06
1.39E-03	1.92E-06	1.59E-05
-1.11E-03	1.22E-06	6.21E-06
3.65E-04	1.33E-07	2.16E-06
-5.33E-04	2.84E-07	8.06E-07
4.33E-04	1.87E-07	9.32E-07
-8.89E-04	7.90E-07	1.75E-06
-3.92E-04	1.54E-07	2.47E-07
-1.92E-03	3.68E-06	2.33E-06
-1.18E-03	1.39E-06	5.49E-07
-1.83E-04	3.34E-08	9.90E-07
-2.52E-04	6.37E-08	4.83E-09
		6.37E-08

Note

et= residual

Auto-related data causes the degree of freedom from statistics on inferential tests and leads to faux correlations between variables [50]. In a fundamental modelling exercise such as modified Gompertz and other models, the usage of the Durbin Watson test to test for autocorrelation data in time series are widespread [51–55]. The Breusch-Godfrey Lagrange multiplier test is another method of detection of autocorrelation. When autocorrelation is identified, the researcher can fix the condition using numerous methods of transformation, such as Cochrane-Orcutt [56], Hildreth-Lu, or Prais-Winsten that can alleviate the presence of autocorrelation [57].

The Durbin-Watson test statistic compares the null hypothesis that residuals in normal least-field regression are not auto-related to the alternative that residuals in an AR1 process, in which the current value is based on the immediately preceding value, are auto-related. Durbin-Watson has a statistical range of 0 to 4. Non-self-correlation has a value close to 2; positive autocorrelation indicates a value close to 0, and negative autocorrelation indicates a value close to 4. Because any computed Durbin Watson value is dependent on the related data matrix, the exact critical values of Durbin-Watson statistic are not given in all possible cases [51,52].

Durbin and Watson, on the other hand, established the critical values at the upper and lower boundaries. Because positive autocorrelation is far more common in practice than negative autocorrelation, the hypothesis of zero autocorrelation against the alternative positive self-relation of the first order is commonly used in tabular bounds. To utilise the table, cross-reference the sample size to the number of regressors, removing the constant from the regressors count. Traditional Durbin Watson tables are inapplicable when there is no permanent regression term. Instead, a suitable set of Durbin-Watson tables must be used. Traditional Durbin-Watson tables do not applicable when the lagged variable is shown on a regressor. Durbin proposed various testing procedures in this case.

Several factors might have contributed to the introduction of autocorrelation into the data [58], including the following:

1. Because of the regularity with which it occurs, carryover of effect is a primary cause of autocorrelation. Statistics on monthly household expenditures, for example, are influenced by the same category of spending from the previous month's data. Autocorrelation can be seen in cross-sectional and time-series data sets. When examining cross-sectional data, the feature under discussion is common in that it enables the discovery of units that are equivalent to one another. When working with time series data, the element of time is what creates self-correlation. When certain sample units are ordered in the data, autocorrelation might occur. The effect of omitting specific variables from an equation is another aspect that contributes to autocorrelation. When employing regression modelling approaches such as regression modelling, it is not possible to include all of the variables in a regression model. There are several reasons for this, not the least of which is that some variables are qualitative in character, direct observations on the variable are not always available, and so on. The autocorrelation in the resulting data is created

by the combined effect of the variables that were removed [59,60,33,61–63].

The introduction of autocorrelation into the data could be due to an erroneously defined kind of connection. Its goal is to establish a linear relationship between the research and the explanatory variables in the link between the research and the explanatory variables. The data exhibits autocorrelation as a result of a log or exponential factor in the model. This is due to the model's linearity being called into question. When the discrepancy between observed and actual values is greater than one standard deviation, it is referred to as a measurement error or error-in-variable for that variable. Furthermore, the presence of measurement errors in the dependent variable may result in undesired autocorrelation in the data set. It's also referred to as serial correlation or autocorrelation. It is defined as the time-delayed correlation of one signal with a delayed replica of itself. Informally, it is the degree to which two observations are similar as a function of the time interval between them.

CONCLUSION

When it comes to detecting recurring patterns, autocorrelation analysis is a mathematical method that can be used to determine the presence of a periodic signal that has been obscured by noise or to locate the missing fundamental frequency in a signal revealed by its harmonic frequencies. It is commonly used in signal processing to analyse functions or series of values, such as time-domain signals, and is very useful in signal processing. In this study, the calculated value of the Durbin-Watson statistics was $d = 2.260$. The Durbin-Watson's lower critical value for DL was 0.700, while the upper critical value dU was 1.252. Since the d value was greater than the upper critical value or dU, this resulted in the null hypothesis not being rejected or indicating that there is no evidence of autocorrelation. This demonstrates that the pseudo-1st model used in the nonlinear regression model is adequate.

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