

# Removal of lindane and *Escherichia coli* (*E.coli*) from rainwater using photocatalytic and adsorption treatment processes

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# Abstract

Low concentrations of contaminants in rainwater is the primary benefit of transforming it as an alternative source of drinking water. However, treatment is necessary as rainwater collects contaminants that are persistent in the environment such as lindane and E.coli. The combination of photocatalysis and adsorption processes was chosen as the treatment method in this study. From the results obtained, photocatalysis treatment process was able to degrade lindane in synthetic rainwater under different experimental conditions such as pH, titanium dioxide (TiO<sub>2</sub>) dosage, and initial concentration. The photodegradation process of lindane followed pseudo kinetic first order. In adsorption process, the adsorbents used in the process are limestone and laterite soil. The performance of these adsorbents are determined by carrying out an equilibrium batch study. The experimental works show that limestone and laterite soil able to remove E.coli at 99%. The optimum dosage of limestone and laterite soil to remove E.coli from synthetic rainwater is 6g and 2g, respectively. The results were then analysed by developing Langmuir and Freundlich isotherm model. Overall, the processes of photocatalysis and adsorption showed a good performance in removing lindane and E.coli from the synthetic rainwater respectively.

**Keywords:** synthetic rainwater, pH, titanium dioxide, photodegradation, adsorbents, limestone, laterite soil

# 1. Introduction

In order to provide an alternative solution to the water scarcity problem, rainwater can be collected to serve our water supply system. Generally, harvested rainwater provides a good quality in terms of physicochemical properties (Gikas and Tsihrintzis, 2012). The low concentrations of contaminants in the harvested rainwater is the primary advantage to convert it as an alternative source of drinking water. However, the presence of micropollutants such as organochlorine pesticides (OCPs) and *Escherichia coli (E. coli)* in the harvested rainwater may affect the human health (Ismail *et al.* 2013; Wilbers, Sebesvari *et al.*, 2013). OCPs are the persistent organic pollutants (POPs) urgently needed to be eliminated because of their high solubility in greases, which involves an accumulation in fat tissues of the living organisms (Pang, *et al.* 2009; He and Balasubramanian, 2010). OCPs mostly occur in waters that have been affected by agricultural discharges. Once released into the environment, these OCPs could remain for quite a long time and as the result, these compounds are being collected in the rain droplets as the wet precipitation occurs.

One of the most harmful pesticides that is persistent in the environment is lindane (Devipriya and Yesodharan, 2005). There are several studies that have proven the concentrations of lindane in rainwater (Basheer *et al.*, 2003; He and Balasubramanian, 2009; He and Balasubramanian, 2009; He and Balasubramanian, 2010; Mahugija *et al.*, 2015). Naturally, the presence of *E.coli* in a water body comes from the human and animal fecal waste. The effects of *E.coli* in water may cause illness and infection to any humans who drink it. Therefore, United States Environment Protection Agency (U.S EPA) has developed a standard limit for *E.coli* is zero. Hence, further treatment is necessary in order to make the quality of harvested rainwater parallel to the requirements according to the drinking water standard.

Treatment is required to improve the quality of the harvested rainwater. Several potential methods have been studied to treat the harvested rainwater. In 2011, Amin and Han used solar disinfection method with the use of lemon and vinegar as the catalyst. The study showed that solar disinfection was able to remove and inactivate microbiological contaminants from harvested rainwater (Amin and Han, 2011). Other than that, there were also studies of rainwater harvesting treatments by using combination process (Jordan *et al.*, 2008; Naddeo *et al.*, 2013). Jordan and his co-researchers has investigated the use of the combination treatment by using filtration and UV disinfection to remove microbiological contaminants from the harvested rainwater (Jordan *et al.*, 2008). Meanwhile, Naddeo and his co-workers applied the

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combination of filtration, adsorption (using granulated activated carbon), and disinfection (UV light) to enhance the quality of harvested rainwater as drinking water supply. The results showed that the combination process was able to produce clean water in terms of microbiological quality as disinfection process provides a good barrier for pathogens and several major contaminants (Naddeo *et al.*, 2013).

Advanced oxidation processes (AOPs) constitute the best option for the destruction of POPs in the near future (Esplugas et al., 2002). The versality of AOP is enhanced by the fact that they offer different possible ways for hydroxyl radical production thus allowing a better compliance with the specific treatment requirements. One of the most widely used AOPs is photocatalysis. Photocatalysis with TiO2 catalyst has been a promising method to be used in removing the organic and inorganic pollutants in the water treatment as reported in the previous studies (Zaleska et al., 2000; Muneer et al., 2005; Lair et al., 2008; Mahmoodi and Sargolzaei, 2014). The basic principle of semiconductor photocatalysis relies on the formation of an electron-hole pair upon the absorption of a photon with energy equal or bigger than the semiconductor band gap (Robertson et al., 2012). The unlimited supply of sunlight and oxygen, if natural sunlight is to be used, makes photocatalysis better to be used compared to other processes (Malato et al., 2009).

Adsorption process is one of the most commonly applied in rainwater harvesting treatments (Fuping et al., 2012; Gan, et al., 2012; Wang et al., 2013; Yang et al., 2015). Generally, adsorption process can be defined as a mass transfer operation, which a compound can be transferred from liquid phase to solid phase. The substance that is being removed from liquid phase at the interface is described as adsorbate, while adsorbent is the medium which the adsorbate accumulated with in the form of solid, liquid, or gas phase (Tchobanoglous, 2003). Numerous studies have revealed the usage of limestone in various kinds of water treatment (Aziz et al., 2004; Aziz, Adlan et al. 2008; Silva, Cunha et al. 2012; Wang et al., 2013). The presence of calcium carbonate (CaCO<sub>3</sub>) and magnesium carbonate (MgCO<sub>3</sub>) in limestone enable the adsorbent to remove pollutants such as heavy metals from a water body (Aziz et al., 2004; Aziz et al., 2008; Wang et al., 2013). On the other hand, the removal of pollutants by laterite soils was caused by the high content of silica compound. However, these adsorbents are rarely used to study the removal of E.coli.

Therefore, this study was conducted in order to determine the removal efficiency of lindane after being treated with photocatalytic treatment process. Under different experimental conditions: the effects of pH, catalyst loading, and the initial concentration on the degradation percentage of lindane were discussed. The kinetic study on the photodegradation of lindane was also investigated. Besides that, the optimum dosage of limestone and suitable adsorption isotherm model in removing *E.coli* from synthetic rainwater were examined in this study.

#### 2. Methodology

#### 2.1. Chemical Reagents and Materials

The chemicals and materials involved in this study were described. The analytical standards of lindane ( $\gamma$ -HCH) were purchased from Sigma-Aldrich (Steinhelm, Germany) with a purity of 97.5%. Chloroform used as the organic solvent, also dichloromethane and methanol used as GC eluent were of the analytical residue grade and obtained from Systerm. Titanium dioxide (Degussa P25) used in this work, is a known mixture of 70% anatase and 30% rutile, has a 99.5% purity with mean particle size of 30 nm and surface area of 50 m<sup>2</sup>/g, was supplied from R&M Marketing, Essex, U.K. Other chemicals were used without further purification. Deionized and doubly distilled water was used throughout this study.

Meanwhile, the source of E. coli in this study was collected froom oxidation pond in Universiti Sains Malaysia in Nibong Tebal, Penang, Malaysia. The materials used in this study are limestone and laterite soil. High quality limestone was sieved to obtain the size of 2.00 mm mesh. The adsorbent was then washed with distilled water for several times in order to remove dust and other impurities. After that, it was dried in the oven at 105 °C for 24 hours. The laterite soil were washed by adding 100g of laterite soil into 1000 ml of deionized water and were stirred with 250 rpm for 30 minutes. The mixture was then filtered to collect the treated laterite soil and was oven dried at 105 °C for 24 hours (Lau et al., 2014; Rout et al., 2014). Then, the washed laterite soil was sieved and the sample that retained at 630µm were used for the adsorption process (Maiti et al., 2013).

#### 2.2. Experimental works

#### 2.2.1. Preparation of Synthetic Rainwater

The synthetic solutions for lindane and E. coli were prepared at low concentration to imitate the condition of real rainwater. Stock solution of lindane was prepared by dissolving the powdered standard of lindane with chloroform and stored at 4 °C (Huang *et al.*, 2009; Rianawati and Balasubramanian, 2009). Meanwhile, the synthetic rainwater of *E.coli* was prepared by spiking 1mL of wastewater into 1L of distilled water. The initial content of *E.coli* in the synthetic rainwater is 1986 MPN/100mL after the dilution.

# 2.2.2. Photocatalysis and Adsorption Batch Equilibrium Study

Photocatalytic treatment was carried out by exposure of the synthetic rainwater with titanium dioxide to natural sunlight in different experimental conditions, such as pH, initial concentration of lindane, and dosage of titanium dioxide.

Adsorption batch equilibrium study was formed in determining the optimum adsorbent dosage and adsorption isotherm models. The experiments were carried out with a series of twenty 250 mL of conical flasks contained of synthetic rainwater for *E.coli* at pH 7 with adsorbent. The adsorbent dosage for limestone and laterite soil is in a range of 2 - 20g. The conical flasks were placed on the orbital shaker at 200 rpm for 60 minutes.

#### 2.3. Analytical Method

The concentration of lindane was obtained by using the liquid-liquid extraction method: 100 ml of the synthetic sample was transferred into a conical flask and mixed with 100 ml of dichloromethane. The mixtures were shaken in a shaking incubator at room temperature for two hours with 120 rpm of shaking speed. After 30 minutes, the two layers were formed. The mixtures were transferred into a separating funnel. From the separating funnel, the lower layer that formed the organic layer was collected and evaporated to dry. After being dried, the extract was mixed with 1 mL of dichloromethane and ready to be analyzed by GC-MS. The standard solution of lindane was prepared in dichloromethane at concentration of 1 mg/mL and diluted at concentration of 200000, 2000, 2000, 200, and 20 µg/L.

The quantification of lindane was conducted using an Agilent 6890N Series GC System coupled with Agilent 5973 Inert Mass Selective Detector. The column used was Agilent HP-5MS fused capillary column (USA) with description as follows: 30.0 m x 250  $\mu m$  x 0.25  $\mu m.$  The carrier gas was helium. The inlet temperature was 300 °C. 1  $\mu$ l of the sample was injected into the GC. The temperature program was as follows: initial oven temperature was held at 70 °C for 2 min, and ramped to 290 °C at a rate of 15 °C/min. The mass spectrometer was operated under electron impact (EI), choosing 70eV as the electron energies, while the ion source temperature was set to 230 °C. Selective ion monitoring (SIM) mode was used for monitoring the analyte. Quantitation ion for lindane is 181. Lindane was identified by comparing the retention time (chromatographic column) and mass spectra (mass detector) with the standard (full scan mode).

On the other hands, *E.Coli* content was determined by using IDEXX Colilert Method, which has been approved by U.S EPA and included in Standard Methods of Quality of Water and Wastewater (APHA Standard Method 9221 A-F). This experiment was carried out within 24 hours after preparing the synthetic rainwater in order to maintain the *E.coli* content in the samples.

#### 3. Results and Discussion

#### 3.1. Photocatalytic Treatment Process

# 3.1.1. рН

In heterogeneous photocatalytic water system, pH is one of the most important operational factors that affect the charge on the catalyst particles and the position of the conduction and the valence bands. Due to the nature of TiO<sub>2</sub> catalyst used, any variation in the operating pH is known to affect the isoelectric point or the surface charge of the photocatalyst used (Chong *et al.*, 2010). The point of zero charge (PZC) of the TiO<sub>2</sub> (Degussa P25) is widely reported at pH 6.25 (Li *et al.*, 2008). While under acidic conditions, the positive charge on the TiO<sub>2</sub> surface increases as the pH decreases, and above pH 6.25 the negative charge on the surface of the TiO<sub>2</sub> increases with increasing pH. Fig. 1 shows the percentage removal of lindane from synthetic rainwater in accordance to different pH condition, that is from 3 to 11. Based on the figure, lindane was highly degraded in this process.



# Figure 1. Percentage removal of lindane from synthetic rainwater with pH 3 to 11

#### 3.1.2. TiO<sub>2</sub> Dosage

Fig. 2 shows the percentage removal of lindane from synthetic rainwater in accordance to different  $TiO_2$  dosage, that is from 0.1 to 0.5 g/L. Based on the figure, lindane obtained maximum degradation when the  $TiO_2$  dosage used is 0.2 g/L, followed by 0.5 g/L. From this result, it can be concluded that the higher the amount of  $TiO_2$  used, the higher the degradation rate. The reason generally for this is that the increase in amount of catalyst increases the number of active sites on the photocatalyst surface, which in turn increase the number of hydroxyl and superoxide radicals (Gaya and Abdullah, 2008).



Figure 2. Percentage removal of lindane from synthetic rainwater with  $TiO_2$  dosage 0.1 to 0.5 g/L

However, in the review of TiO<sub>2</sub>-assisted photocatalytic degradation of azo dyes in aqueous solution reported that in any solar reactor system, the initial rates were found to be directly proportional to catalyst concentration, indicating the heterogeneous regime, until it reached a limit of catalyst concentration at which the rate of photocatalysis will even decrease (Konstantinou and Albanis 2004).

#### 3.1.3. Initial Concentration

Fig. 3 shows the percentage removal of lindane from synthetic rainwater in accordance to different initial concentration of pollutant that is from 10 to 100  $\mu$ g/L. Based on the figure, the lowest degradation rate of lindane occurs at 70 and 80  $\mu$ g/L. This shows that the higher the initial concentration of each pollutant, the lower the oxidation rate. Owing to the photonic nature of the

photocatalysis reaction, excessively high concentration of organic substrate is known to simultaneously sature the  $TiO_2$  surface and reduces the efficiency of the light penetration into the solution, leading to photocatalyst deactivation (Muneer *et al.*, 2005). This may be due to the fact that at higher concentrations more substrate molecules could occupy more active sites, thus inhibiting the generation of the oxidants (Yang *et al.*, 2012).



Figure 3. Percentage removal of lindane from synthetic rainwater with initial concentration of pollutant from 10 to  $100 \mu g/L$ 

#### 3.1.4. Kinetics of the Photodegradation Process

For the kinetic photodegradation processes, pseudo first order equation is normally assumed (Bosco *et al.,* 2006). By using the following equation, the rate constant of the photodegradation process can be calculated:

$$\ln \frac{C_o}{C} = kt$$
 (1)

Where  $C_o$  and C are the pollutant concentrations at times 0 and t respectively, and k being the rate constant. Fig. 4 shows the photocatalytic degradation of lindane in the synthetic rainwater. Based on the plotted graph, the correlation coefficient of lindane is 0.585.



Figure 4. Photocatalytic degradation of lindane

The pseudo rate constants, half life times and the removal percentages of lindane are depicted as in Table 1. From the results below, it is suggested that further studies with wider range of experimental conditions need to be done in order to investigate the behavior and performance of lindane throughout the photocatalytic treatment process.

Table 2. Linearity of lindane

**Table 1.** Values of kinetic parameters for photodegradation of lindane.

Time (hour)	k (hour⁻¹)	t½ (hour)	% Removal Efficiency
1	0.6	1.2	45
2	0.0	17.6	8
3	0.9	0.8	94
4	0.3	2.7	64
5	0.4	1.9	84
6	0.2	3.1	74

3.1.5. Gas chromatography mass spectrometer

The retention time of lindane was approximately 10.00. Fig. 5a shows a chromatogram of lindane at concentration of  $100\mu$ g/L. Fig. 5b shows a chromatogram of lindane after treatment.



Figure 5a. Chromatogram of lindane at 100µg/L



Figure 5b. Chromatogram of lindane after treatment

The calibration curves were established by plotting peak area of analyte. The linear regression equation was calculated by using Microsoft Excel program and is summarized in Table 2.

The LOD is the lowest amount of analyte that can be detected, but not necessarily quantitated as an exact value. LOQ is the lowest amount of analyte that can be quantitatively determined with suitable precision.

Analyst	Regression equation	Correlation coefficient	Linearity (µg/L)	LOD* (µg l <sup>-1</sup> )	LOQ** (µg l <sup>-1</sup> )
Lindane	y=38.40x-138360	0.9980	20 - 200000	2	20

\*LOD = Limit of detection, \*\*LOQ = Limit of quantification

## 3.2. Adsorption Process

#### 3.2.1. Removal of E. coli

The percentage removals of *E.coli* were plotted by in Fig. 6. The figure depicted the removals of *E.coli* by limestone and laterite soil at the range of 2 - 20g. Based on the figure, results indicate the removal of *E.coli* reached more than 90% at 2g and 6g for laterite soil and limestone, respectively.



Figure 6. Percentage removal of *E.coli* from synthetic rainwater

The percentage removal of E.coli for laterite soil was more efficient than limestone due to the smaller particle size of the medium. Smaller particles of adsorbent expose a larger surface area compared to coarse particles. Therefore, it provides more adhesion site of adsorbent (Fontes et al., 1991; Tan et al., 1992). Besides that, another factor that contributed to the removal of E.coli is the presence of divalent cation in the adsorbents. Generally, limestone is highly content of calcium carbonate, while laterite soil contained of iron oxide (Aziz et al., 2008; Maiti et al., 2013). Huysman and Verstraete stated that the presence of divalent cation increased the attachment of bacteria to the surface of adsorbent (Huysman and Verstraete 1993). Therefore, it can be concluded that the presence of Ca<sup>2+</sup> and Fe<sup>2+</sup> ions in the chemical compound enhanced the attachment of E.coli to the adsorbents.

#### 3.2.2. Adsorption Isotherm Model

Adsorption isotherm is a function of the concentration of adsorbate at constant temperature in order to determine the amount of material adsorbed. The adsorbent phase concentration can be computed by Equation (2) below (Tchobanoglous, 2003; Halim *et al.,* 2010; Rout *et al.,* 2014):

$$q_e = \frac{(C_o - C_e) V}{m}$$
(2)

Where;

qe = adsorbent phase concentration after equilibrium, MPN E.coli adsorbate/ g adsorbent

C<sub>o</sub>= initial concentration of adsorbate, MPN/100mL

 $C_e$ = final equilibrium concentration of adsorbate after adsorption has occurred, MPN/100mL

V= volume of liquid in the reactor, mL

m = mass of adsorbent, g

In this study, the chosen adsorption isotherms were Langmuir isotherm model and Freundlich isotherm model. These isotherms were chosen in order to determine the efficiency of adsorption process.

# 3.2.3. Langmuir Isotherm Model

Langmuir isotherm model assumes a fixed number of sites and only one solute molecule per site of adsorbent and suggest the monolayer adsorption on a homogeneous surface (Tchobanoglous, 2003; Halim *et al.*; 2010, Rout *et al.*, 2014). The Langmuir isotherm relates with  $q_e$  (mg of adsorbate adsorbed per gram of adsorbent media) and  $C_e$ (the equilibrium adsorbate concentration in solution) as shown in Equation 3 (Aziz *et al.*, 2008; Halim *et al.*, 2010; Ismail *et al.*, 2013):

$$q_e = \frac{QbC_e}{(1+bC_e)}$$
(3)

Where;

Q= maximum adsorption capacity, MPN/g

#### b= Langmuir constant, L/mg

The constant in Langmuir isotherm can be determined by plotting graph of  $Ce/q_e$  against Ce which can be rewrite as shown in Equation (4):

$$\frac{C_{e}}{q_{e}} = \frac{1}{Qb} + \frac{C_{e}}{Q}$$
(4)

Based on the Langmuir isotherm model equation, a dimensionless constant that indicates the nature of the monolayer adsorption can be determined. It is expressed in terms of equilibrium parameter,  $R_L$  as shown in Equation (5):

$$R_{L} = \frac{1}{1 + (1 + bC_{o})}$$
(5)

Where,

R<sub>L</sub>= equilibrium parameter

b = Langmuir constant (L/g)

Co= initial concentration of E.coli (MPN/100mL)

The  $R_L$  value determine the monolayer adsorption nature to be either unfavorable if  $R_L > 1$ , linear if  $R_L=1$ , favorable if  $O<R_L<1$  and irreversible if  $R_L=0$  (Halim *et al.*, 2010; Rout *et al.*, 2014).



Figure 7. Langmuir Isotherm Model for Removal of *E.coli* by Limestone

In this research, the Langmuir isotherm model equation and correlation coefficient, R<sup>2</sup> for *E.coli* uptake from synthetic rainwater to limestone and laterite soil are depicted in Fig. 7 and Fig. 8. Based on the figures, the values of R<sup>2</sup> for Langmuir isotherm model for limestone and laterite soil were 0.993 and 1, respectively. These values demonstrated a very good mathematical fit of the experimental data since it were higher than 0.96 (Rout et al., 2014). The maximum adsorption capacities of E.coli, Q can be determined from the obtained equation in the figures. Therefore, the obtained maximum adsorption capacities for limestone and laterite soil are 2500 MPN/g and 3333.3 MPN/g, respectively. The result demonstrates that laterite soil has higher adsorption capacities as compared with limestone. Since the media size used for laterite soil is smaller than limestone, therefore, it enhanced the higher capacities of E.coli to adsorb to the adsorbent due to the larger surface area as compared with larger particles of limestone (Fontes et al., 1991; Tan et al., 1992). Based on the obtained Langmuir isotherm equation, the calculated values of RL for limestone and laterite soil are -0.0002 and -0.000001, respectively. The values of  $R_L$  for both adsorbents were too small and nearly zero. The irreversible reaction might occur in the process. Bacterial adsorption and attachment to the surface of adsorbent is a two-step process. The first step is reversible adsorption. It takes place when the bacteria located in the secondary minimum overcome the secondary repulsive force between the bacteria and the porous material (Van Loosdrecht et al., 1990). In this phase, the primary forces involved were electrostatic force and van der waals force (Mozes et al., 1987). It is a reaction when the attached bacteria detached from the surface of the adsorbent particle and return the water phase. The second step is irreversible adsorption, which also referred to as adhesion. The reaction occur when bacterial polymers from bridges that connect the bacteria to the adsorbents (Stevik et al., 2004).



Figure 8. Langmuir Isotherm Model for removal of *E.coli* by laterite soil

# 3.2.4. Freundlich Isotherm Model

On the other hands, Freundlich isotherm model assumes the uptake of adsorbate occurs on a heterogeneous surface by multilayer adsorption and the amount of adsorbate adsorbed increases infinitely with an increase in concentration (Tchobanoglous, 2003; Halim *et al.*, 2010). The Freundlich isotherm can be expressed as shown in Equation (6) (Halim *et al.,* 2010; Ismail *et al.,* 2013; Rout *et al.,* 2014):

$$q_e = K_F C_e^{1/n} \tag{6}$$

Where;

K<sub>F</sub>= indicator of the adsorption capacity

## 1/n= adsorption intensity

The value of  $K_F$  and 1/n can be determined by plotting the graph in linear form of the Freundlich expression as shown in Equation (7), which  $K_f$  can be determined from the intercept of the graph while 1/n can be determined by the slope of the graph.

$$\log q_e = \log K_F + \frac{1}{n} \log C_e$$
(7)

In this study, the obtained correlation coefficient, R<sup>2</sup> and linearized Freundlich isotherm equation for limestone and laterite soil were showed in Fig. 9 and Fig. 10.



Figure 9. Freundlich Isotherm for Removals of *E.coli* by Limestone



Figure 10. Freundlich Isotherm Model for removal of *E.coli* by laterite soil

Based on the figures, the values of  $R^2$  for limestone and laterite soil were 0.62 and 0.18, respectively. The  $R^2$  values of this model for both adsorbents were lower than  $R^2$  of Langmuir isotherm model. Besides that, the poor correlation coefficient for both materials has confirmed that the removal of *E.coli* does not fit to Freundlich isotherm model (Rout *et al.*, 2014). However, the obtained values of n for limestone and laterite soil were 5.1894 and 1.673, respectively. The n values indicate whether the heterogeneous adsorption process were favorable if n >1, and unfavorable if n<1 (Halim *et al.*, 2010; Rout *et al.*, 2014).

#### 4. Conclusion

The photocatalytic treatment of synthetic rainwater using  $TiO_2$  was studied to eliminate linden in this study. The results indicate that this advanced oxidation process was able to degrade lindane successfully with different experimental conditions such as pH,  $TiO_2$  dosage, and initial concentration. The photodegradation process of lindane followed pseudo kinetic first order. The correlation coefficient of lindane is 0.585. Further studies need to be done in order to monitor the behavior and performance of lindane under a wider range of different experimental conditions.

The optimum adsorbent dosage of limestone and laterite soil in order to remove E.coli from synthetic rainwater is determined. The removal of E.coli was influenced by the size and chemical composition of media. Isotherm model were developed in order to determine the mechanism of adsorption in this study. Based on the obtained result, Langmuir isotherm model was the best fit model as compared with Freundlich isotherm model. It is because the correlation coefficients of Langmuir isotherm model for both materials were higher than Freundlich isotherm model. The equilibrium parameter of Langmuir isotherm, RL indicated small negative values for both materials. Since it is nearly zero, it can be concluded that the nature of the adsorption process was irreversible reaction. Therefore, it can be concluded that limestone and laterite soil are a potential media to remove *E.coli* from synthetic rainwater.

Overall, the processes of photocatalysis and adsorption showed a good performance in removing lindane and *E.coli* from the synthetic rainwater respectively. Based on the results, there is a potential to develop an integrated process by combining these two methods as a new treatment in the future. To date, there were also studies that have been done previously, that have utilized photocatalysis to remove *E.coli* while adsorption to remove lindane. Therefore, the idea of developing this integration method can be a good opportunity towards creating an efficient and advantageous method for removing biological and chemical contaminants present in the environment.

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