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Adsorption of lindane as an organo-chlorine pesticide from aqueous solutions by mineral sand

Kambiz Moradnejadi[1], Ali Torabian[2], Heshmatollah Nourmoradi[3,4*], Sayed Ahmad Mirbagheri[1]

1. Department of Environmental Engineering, School of Energy and Environment, Islamic Azad University, Science and Technology Branch, Tehran, Iran
2. Department of Environmental Engineering, University of Tehran, Tehran, Iran
3. Biotechnology and Medical Plants Research Center, Ilam University of Medical Sciences, Ilam, Iran
4. Department of Environmental Health Engineering, School of Health, Ilam University of Medical Sciences, Ilam, Iran

***Corresponding author:** Tel: +98 8432235733 Fax: +98 8432235733
Address: Department of Environmental Health Engineering, School of Health, Ilam University of Medical Sciences, Ilam, Iran
E-mail: Ilam_nourmoradi@yahoo.com
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Abstract

Introduction: Pesticide including lindane has been extensively applied in agriculture as insecticide. It is considered as one of the most significant organic pollutants that have reasoned environmental pollution especially surface and ground waters. In the present study, mineral sand, as a media of slow sand filtration in water treatment, was used to remove lindane from aqueous solutions.

Materials and methods: This study was carried out in batch mode. The influences of various parameters such as contact time, pH, lindane content and solution ion strength were investigated on the sorption process. The isotherm and kinetics of the sorption was also determined by the research.

Results: The maximum removal efficiency of lindane by sand was achieved at the contact time and pH of 150 min and 4, respectively. The removal efficiency of lindane at abovementioned condition was (28.86% equal to 2.88 µg/g) in the concentration of 100 µg/L. The results of various isotherms and kinetics models analysis also showed that the adsorption process described well by the Freundlich isotherm and pseudo-first-order kinetics models, respectively. Addition of the salt to lindane solution increased the removal efficiency to about 3.5 times.

Conclusion: This study showed that the sorption is considered as one of the major parameters of mineral sand as main media in sand filtration of water treatment plant.

Keywords: Adsorption, Sand, Lindane, Aqueous solution

Introduction

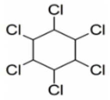
Environmental pollution, as a result of agricultural activities development and irregular usage of agricultural pesticides and fertilizers, has been taken into consideration as an important alarm in recent decades, particularly in developing countries (1). Pesticides are one of the most significant organic pollutants that have reasoned soil and surface and ground waters pollution. Among pesticides, organo-chlorine type is categorized as persistent organic pollutants (POPs), because of their high toxicity, chemical resistance and biological accumulative property (2). Hexachlorocyclohexane (HCH) is a synthetic chemical product which finds in eight isomers. Gamma-HCH (namely Lindane) is one of these isomers which has the most toxicity (3). Lindane is one of the organo-chlorine pesticides (OCPs) which has been extensively applied in agriculture as insecticide since the beginning of 1950s (4). In the past years, this insecticide was also used to wood preservatives and human health to fight against insect-borne diseases including lice and tick, commonly in developing countries (5-7). Lindane has detrimental effects on the aquatic organisms as well as human health. Lindane is toxic to the kidney, liver and reproductive tract by various contact routes such as orally, dermally or by respiration (8). Long-term exposure to this synthetic compound has been also connected to other human health effects like immunosuppression and neurological problems and liver cancer in rats and mice (9). The International Agency for Research on Cancer (IARC) has listed lindane as carcinogenic to humans (Group 1) (10). On the Basis of the world health organization (WHO) guideline, the maximum allowable concentration of lindane in drinking water should not be more than of 2µg/L (11). Therefore, nowadays, many countries have limited and banned the application of this pesticide; however, it is used in several parts of the world (12). In spite of restrictions, lindane inserts in the environment (water, sediments, soil, plants and animals) and because of its high solubility in lipids, lindane can bioaccumulate straightforwardly in the food chain (5, 12). Because of the abovementioned health effects, lindane has been considered as one of the priority organic pollutants and therefore, removal of it from the environment especially water and wastewater solution is necessary (13). Several treatment procedures like chemical oxidation, biodegradation and adsorption have been applied for the removal of organo-chlorine pesticides (OCPs) from aqueous solutions (3, 4, 15). Adsorption process, because of its easiness and cost-effectiveness particularly for low to medium adsorbate content, is one of the most commonly treatment methods for the removal of organic pollutants such as OCPs from the aqueous solutions (1, 16, 17). Adsorption of lindane through fly ash (18), activated carbon (19), fungal biomass (20), clinoptilolite (21) and zeolites (22) has been extensively studied for the sorption of lindane from aqueous solutions. In the present study, mineral sand, as a media of sand filtration in water treatment plant, was used to remove lindane from aqueous phase. The influences of various parameters such as contact time, pH, lindane content and solution ion strength were assessed on the sorption process.

Materials and methods

Materials: The mineral sand (quartz), as the main media of sand filtration in rural and municipal water treatment plants, was provided from Pars-Shimi Co (Iran). Lindane powder (97%) was obtained from Sigma Aldrich CO (USA). Table 1 shows the physical and chemical characteristics of lindane (23). Other chemicals including H2SO4 (96%), HCl, CaCl2 and NaOH were purchased from Merck Co (Germany). The lindane stock solution (1000 µg/L) was weekly prepared by distilled water and maintained in refrigerator at 4 °C. The desired working solutions were made by diluting the stock solution by distilled and applied for the experiments.

Purification of sand: Firstly, 100 g of raw sand was severally washed with distilled water, then, in order to remove more impurities, it was washed by HCl (0.1 N) (24). The sample was finally rinsed with distilled water to neutralized pH and dried in an oven at 150 oC for 2 h. In order to use the adsorbent in the experiments, the sand was graded through screens to obtain mesh size of 45 to 100 (0.15 to 0.35 mm).

Table 1. Physical and chemical characteristics of lindane.

General Name	Chemical Formula	Chemical Structure	Molecular Weight (g/mol)	Appearance	Solubility in water at 25 °C (mg/L)
Lindane	$C_6H_6Cl_6$		290.83	White powder	7.9

Analysis: The concentration of lindane in the solutions was measured by a gas chromatography equipped with electron capture detector (GC-ECD). A gas chromatograph Model YL (Yang Lin, South Korean) equipped with ^{63}Ni electron capture detection (ECD) system and split–splitless injector was applied to measure lindane concentration in the liquid solution. Nitrogen (purity 99.99%) with flow of 1.5 mL/min was used as carrier gas. A capillary column (60 m×0.53 mm×5µm was employed for the separation. A splitless injection of 1 µL was carried out at 240 °C. The temperature program of GC column; the initial temperature was 50 °C and then increased to 250 °C (15 °C/min) and maintained in 250 °C for 5 min. Detection temperature was performed at 240 °C.

Adsorption experiments: The influence of several factors like contact time (0-240 min), pH (4-10), pollutant content (0-250 µg/L), solution ion intensity (20-100 mg/L of Ca^{+2} ions) was assessed on the removal of lindane in the solutions by the sand. All the batch adsorption tests were conducted at room condition (25 ^{o}C). The experiments were performed by 2.5 g of the sand into 250 mL of lindane solution (100 µg/L) through 250 mL Erlenmeyer flask (sealed with stoppers) and blended by a rotary shaker (200 rpm). After mixing time, the suspensions were centrifuged (4000 rpm at 10 min), then filtered (with pore diameter of 0.2 µm) and the clear supernatant was finally analyzed for lindane concentration by GC-ECD. All the tests were carried out in duplicates and the average values were used in results. The removal efficiency of the sorption was determined by Eq. (1):

$$Removal\ (\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \quad Eq.(1)$$

Where, C_o and C_e (µg/L) are the initial and the equilibrium concentrations of lindane in the solution, respectively (25).

Results and Discussion

Effects of contact time: The influence of contact time for the uptake of lindane by mineral sand is shown in Figure 1(a). As seen, the removal efficiency of lindane was raised over time. The adsorption of lindane was quickly increased during the first 100 min (17.25%), then slowly increased during the remaining contact time up to 150 min (19.60%) and finally fixed after 150 min. The higher uptake rate of lindane at the starting adsorption time can be due to the richly available adsorption sites in this stage (1). In a study conducted by Oliver et al. (2013) on the adsorption of four pesticides (Atrazine, Duron, 2,4-D and Chlorpyrifos) in liquid phase by mineral sand mining, the results showed that the equilibrium time has been obtained at 6 h (26). In the present study, the contact time of 150 min was selected for the rest of the experiments.

Adsorption kinetics: Adsorption kinetic models have been widely applied to determine the mechanism of sorption. In the present study, the adsorption data of lindane by sand were analyzed via various kinetic models including pseudo-first-order, pseudo-second-order and intraparticle diffusion models. The pseudo-first order kinetic is expressed by the following equation (1):

$$ln(q_e - q_t) = ln\, q_e - k_1 t \quad Eq.(2)$$

Where, qe and qt are the adsorbed lindane amounts (µg/g) on the sand at equilibrium and at time (min), respectively. k1 (1/min) is the rate constant of pseudo-first order model. k1 and qe are calculated from the slope and intercept of the plotting ln(qe-qt) vs. t, respectively (1). The pseudo-second-order model is shown by Eq. (3):

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad Eq. (3)$$

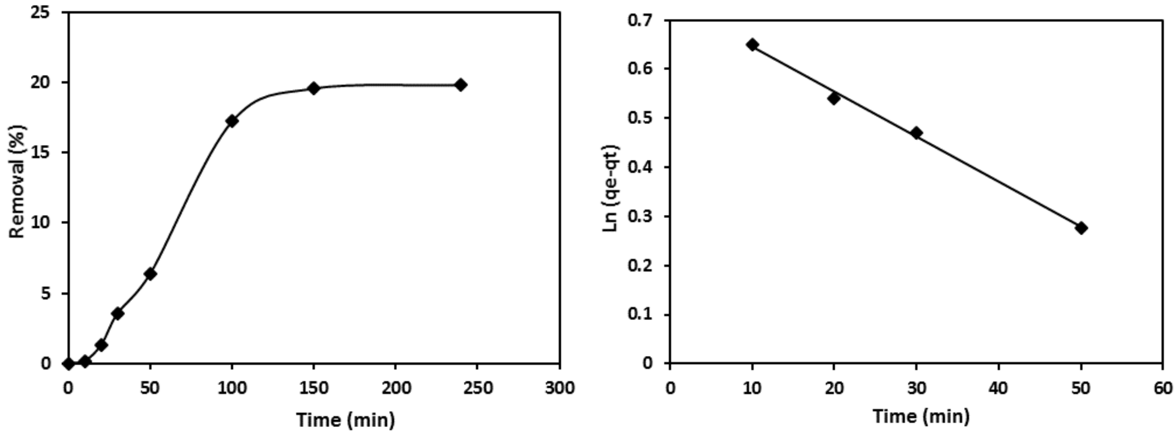


Figure 1. (a) The effects of contact time on lindane adsorption by mineral sand (lindane solution = 100 µg/L, initial pH =7.0, solution volume =250 mL and adsorbent conc. = 2.5 g) and **(b)** pseudo-first order kinetic model.

Where, q_e and q_t are the same as the pseudo-first order model parameters. k_2 (g/µg. min) is the rate constant of pseudo-second order model. The intercept and slope of plotting *t/qt* against *t* is used to determine k_2 and q_e, respectively (1).

The intraparticle diffusion kinetic model is presented by the following equation (25).

$$q_t = k_{id} t^{0.5} + C \quad Eq. (4)$$

Where, K_{id} (g/µg. min) is the intraparticle diffusion kinetic model rate constant. *C* is a constant parameter that is related to the boundary layer thickness. *C* and K_{id} are achieved by the intercept and gradient of the drawing q_t versus $t^{0.5}$, respectively (25). The results of various kinetic models analysis for the adsorption of lindane by mineral sand are presented in Table 2. As can be seen, the upper linear correlation coefficient (R^2) of the pseudo-first order model (R^2 =0.996) indicates that this kinetic model was better to fitting the experimental data in comparison with other kinetic models. Furthermore, the calculated sorption capacity ($q_{e,}$ calculated) value obtained by the pseudo-first order model was rationally alike to experimental sorption capacity (qe, experimental). Figure 1(b) also shows the pseudo-first order kinetic model for the adsorption of lindane from aqueous solutions by the mineral sand. Oliver et al. (2013) study on the adsorption of 2, 4-D pesticide by mineral sand mining also showed that the results had the best agreement with pseudo-first order kinetic model (26). Table 2 also shows the intraparticle diffusion model parameters. As presented, the C value obtained from the linear curve of this kinetic model did not pass from the origin of plotting qt versus t0.5. This illustrates that the intraparticle diffusion is not the only rate controlling factor and other mechanisms such as film (boundary layer) diffusion can control the sorption process to some extent (27).

Table 2. Parameters of various kinetic models for the adsorption of lindane by mineral sand.

	Pseudo-first order			Pseudo-second order			Intra-particle		
$q_{e,\ Experimental}$	$q_{e,\ Calculated}$ (µg/g)	k_1	R^2	$q_{e,\ Calculated}$ (µg/g)	k_2	R^2	*k*	*C*	R^2
1.96	2.08	0.01	0.996	1.17	0.006	0.506	0.237	0.869	0.968

Effects of solution pH: The solution pH has been identified as one of the most significant factors that impact on any sorption process. The surface characteristic of adsorbent and also the ionization degree of adsorbate are influenced by solution pH (28). The influence of initial solution pH on the sorption of lindane by the mineral sand was evaluated at a pH range 4 to 10 and the result is displayed in Figure 2. As can be seen, the solution pH changes had a substantial effect on the sorption. The uptake of lindane by the adsorbent was increased from 1.59% to 28.86% by decreasing solution pH from 10 to 4. This finding can be due to the protonation of the adsorbent surface that happened at the lower pH and as a result, the negatively charged lindane molecules were more readily adsorbed by the mineral sand (28). Ju et al. (1997), Young and Banks (1998), El-Kady et al. (2013) studies confirmed that the maximum sorption of lindane by bacterial biomass, fungal biomass and activated carbon in liquid phase has been occurred at acidic pH, respectively (4, 29, 30). The solution pH of 4 was chosen as the optimum condition for the subsequent of the experiments in this study.

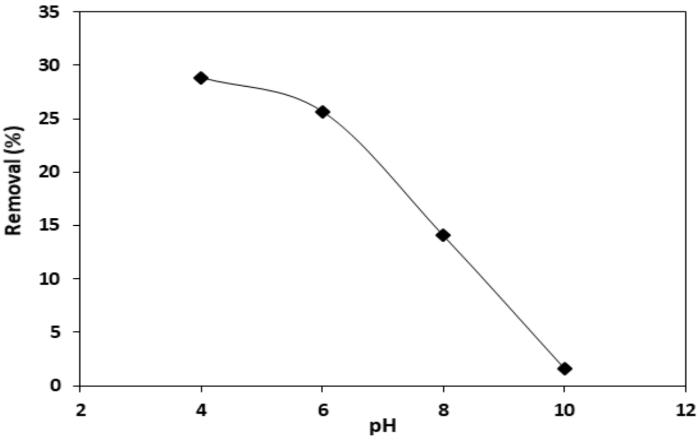


Figure 2. The effect of solution pH on the adsorption of lindane by the sand (lindane solution = 100 µg/L, contact time =150 min, solution volume =250 mL and adsorbent conc. = 2.5 g).

Effects of lindane concentration: The effects of initial lindane content on the sorption by the mineral sand were investigated and the results are presented in Figure 3(a). As shown, the adsorption capability of the mineral sand was raised by the increasing lindane concentration up to 100 µg/L. This may be as a result of an increase in driving force of lindane molecules like van der Waal’s force to the active sorption sites of the sand and finally reached saturated state at lindane concentration more than 100 µg/L (1).

Adsorption isotherms: Adsorption isotherms provide useful data in order to understand the sorption mechanisms, surface properties of the sorbent and design of any sorption system (1). The findings of the influences of lindane concentration on the adsorption were applied to determine the sorption isotherms. Three most commonly isotherms including Langmuir, Freundlich and Dubinin- Radushkevich (D-R) isotherm models were used for this purpose.

The Langmuir isotherm model describes the monolayer adsorption on a homogenous and uniform surface of the adsorbent (31). The linear form of Langmuir isotherm can be presented by Eq. (5):

$$\frac{C_e}{q_e} = \frac{C_e}{Q_m} + \frac{1}{bQ_m} \quad Eq. (5)$$

Where, Ce (µg/L) is lindane concentration in the solution at equilibrium time, qe (µg/g) is the sorption ability of the sand at equilibrium, b (L/µg) is the isotherm constant and Qm (µg/g) is the maximum

monolayer sorption capacity of the sand. Qm and b are achieved through the gradient and intercept of plotting Ce/qe against Ce, respectively (31).

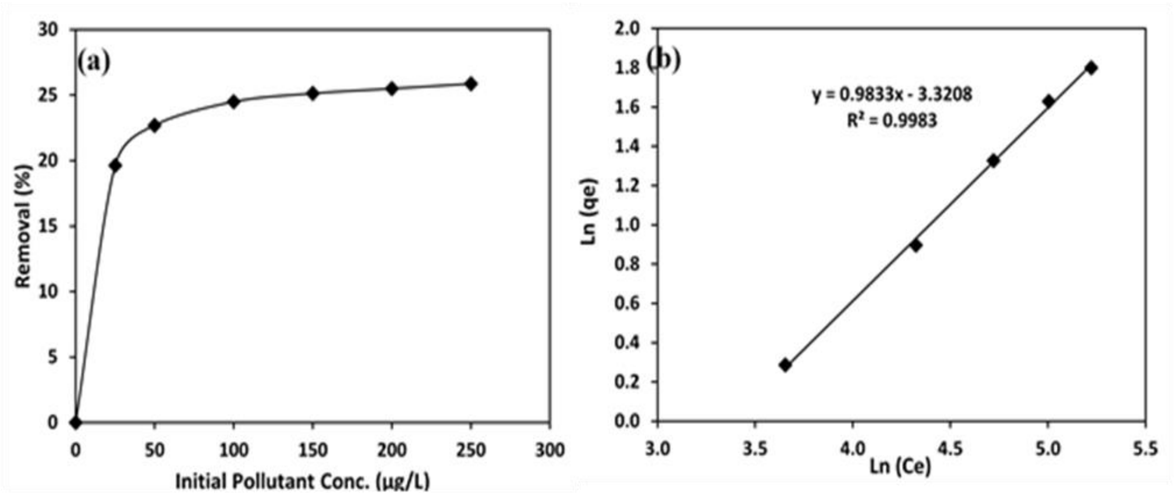


Figure 3. (a) The effects of lindane concentration on the adsorption by mineral sand (pH =4.0, contact time =150 min, solution volume =250 mL and adsorbent conc. = 2.5 g) and **(b)** Freundlich isotherm model.

Adsorption isotherms: Adsorption isotherms provide useful data in order to understand the sorption mechanisms, surface properties of the sorbent and design of any sorption system (1). The findings of the influences of lindane concentration on the adsorption were applied to determine the sorption isotherms. Three most commonly isotherms including Langmuir, Freundlich and Dubinin- Radushkevich (D-R) isotherm models were used for this purpose.

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Where, C_e (µg/L) is lindane concentration in the solution at equilibrium time, q_e (µg/g) is the sorption ability of the sand at equilibrium, b (L/µg) is the isotherm constant and Q_m (µg/g) is the maximum monolayer sorption capacity of the sand. Q_m and b are achieved through the gradient and intercept of plotting C_e/q_e against C_e, respectively (31).

The Freundlich isotherm model presents multilayer sorption on a heterogeneous and non-uniform surface of the adsorbent (25). This isotherm model can be written as equation (6):

$$lnq_e = ln\, k_f + \frac{1}{n}\, ln\, C_e \quad Eq.(6)$$

Where, K_f (L/g) and n are the Freundlich isotherm constants and they are related to the capacity and strength of the sorption, respectively. The intercept and slope of plotting $ln\, q_e$ vs. $ln\, C_e$ can determine K_f and n, respectively (25).

The Dubinin–Radushkevich (D–R) isotherm model explains the chemical or physical characteristic of the adsorption process (1). This isotherm model can be presented via Eq. (7):

$$ln\, q_e = ln\, q_m - \beta\, \varepsilon^2 \quad Eq.(7)$$

Where, q_m (µg/g) is the theoretical sorption capacity at saturation condition, ß (kJ/mol) is a constant parameter associated to sorption energy and $\mathcal{E}$ (Polanyi Potential) is obtained from the following equation:

$$\mathcal{E} = RT\, ln\left(1 + \frac{1}{C_e}\right) Eq.(8)$$

Where, *R* (kJ/mol. K) is gas constant (8.314 J/mol. K) and *T* (K) is absolute temperature of the solution. q_m and *ß* are calculated through the intercept and the gradient of plotting ln q_e against $\mathcal{E}^2$ in Eq. (7), respectively (1). *E* (kJ/mol), mean adsorption energy, is obtained from the constant *ß* as presented by Eq. (9).

$$E = \frac{1}{\sqrt{2\beta}} \quad Eq. (9)$$

The *E* value is used to find out the adsorption process type as physical or chemical. *E*< 8 kJ/mol and *E*> 16 kJ/mol indicate to physical and chemical adsorptions, respectively. The *E* value range of 8-16 kJ/mol determines the chemical ion exchange of the process (31).

Table 3 expresses the Langmuir, Freundlich and D-R isotherm parameters for the adsorption of lindane by mineral sand. As shown, because of the higher correlation coefficient (R^2) value, the Freundlich isotherm model was best described the sorption data. Therefore, the Freundlich model is the most appropriate equation for the sorption of lindane by mineral sand. If the *n* value in Freundlich isotherm is greater than 1, the sorption bond between adsorbent and adsorbate molecules is relatively potent (1). Hence, as listed in Table 3, the n value (1.02) in this isotherm model specified that lindane was favorably adsorbed by the sand. Figure 3(b) shows the data analysis by the Freundlich isotherm model for the adsorption of lindane from aqueous solutions by the sand. Table 3 also exhibits the D-R isotherm parameters for this study. From *E* value (0.57 kJ/mol) of D-R isotherm can be concluded that the physical lindane sorption was occurred by the mineral sand.

Effects of solution ion strength: To evaluate the effect of solution ion strength on the sorption of lindane by the mineral sand, $CaCl_2$ with concentration ranging from 20 to 100 mg/L Ca^{+2} was introduced in the lindane solutions (100 µg/L) at pH 4. The results indicate in Figure 4.

Table 3. Langmuir, Freundlich and D-R isotherm parameters for the adsorption of lindane by mineral sand.

Langmuir isotherm			Freundlich isotherm			D-R isotherm		
Q_m (µg/g)	*b* (L/µg)	R^2	K_f	*n*	R^2	q_m (µg/g)	*E* (kJ/mol)	R^2
16.39	0.002	0.705	0.04	1.02	0.998	4.55	0.57	0.918

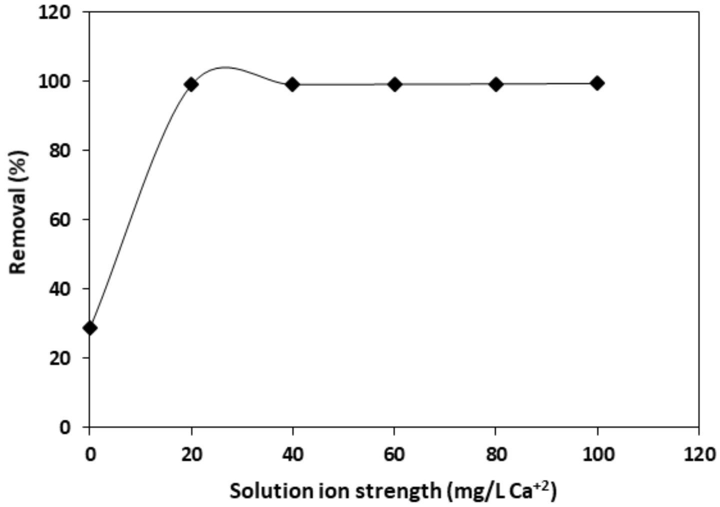

Figure 4. The effect of solution ion strength on the adsorption of lindane by the sand (lindane solution = 100 µg/L, contact time =150 min, pH =4, solution volume =250 mL and adsorbent conc. = 2.5 g).

As depicted, by increasing the solution ion strength from 0 to 100 mg/L Ca^{+2}, the removal efficiency of lindane was enhanced from 28.80% to 99.48%, respectively.

Addition of the salt to lindane solution increased about 3.5 folds the removal efficiency. The reason may be due to this fact that the solubility of lindane is decreased by increasing the salt and subsequently lindane is readily transported toward the sorbent (3, 32). An increasing the uptake capacity of lindane by addition the solution ion strength has been also reported through Tor et al. (2013) study (3).

Conclusion

In this study, mineral sand, as a media of sand filtration in water treatment plant, was applied to eliminate lindane from aqueous phase. The effects of several parameters including contact time, pH, pollutant content and solution ion strength were assessed on the sorption process. The maximum adsorption removal of lindane (28.86%) by mineral sand was achieved at contact time of 150 min and at pH 4. The sorption was physical in nature. The data analysis by various isotherms and kinetics models indicated that the adsorption process described well by the Freundlich isotherm and pseudo-first-order kinetics models, respectively. The solution ion strength had the substantially positive effects on the uptake of lindane by the mineral sand. This study showed that the sorption has an important role for mineral sand as main media in sand filtration of water treatment plant.

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