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Removal of lindane as a health-toxic pesticide in drinking water by slow sand filtration

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ABSTRACT

Lindane is applied in agriculture as insecticide and in veterinary medicine to control the insect-borne diseases. Contact to this synthetic chemical has various adverse effects on human health. This study was conducted to remove lindane in water by slow sand filtration (SSF). The effects of different parameters including lindane concentration, turbidity, filter bed depth and Schmutzdecke type were assessed on the removal of lindane by SSF. The results indicated that the filter media caused the removal of 85.4%, 95.4% and 100% inlet lindane concentration of 200, 100 and 20 μg/L, respectively, over 1 d after operation. These removal efficiencies were approximately constant by increasing operation period up to 5 d. Water turbidity up to 5 NTU had an insignificant effect on the removal of lindane. The filter depth more than 20 cm had negligible effect on the removal of lindane. The findings also showed that the filter media decreased 98.0% and 98.3% of water turbidity and heterotrophic plate count, respectively. The removal efficiency of lindane by the SSF-modified Schmutzdecke layer was nearly 15% higher than that of SSF with the natural Schmutzdecke layer. On the basis of the results, SSF can meet the WHO guideline for lindane in drinking water only in low concentration (up to 20 μg/L).

Keywords: Water treatment; Slow sand filtration; Pesticide; Lindane

1. Introduction

Organo-chlorine compounds are one of the most important constituents which due to various agricultural and industrial activities including accidental leakage, pesticide spray, inappropriate disposal, etc., have led to the environment pollution especially surface and ground waters [1]. Organo-chlorine pesticides (OCPs) such as lindane because of their high persistence and toxicity, bioaccumulation and non-biodegradability are recognized to contaminate water, soil, plants, animals as well as human health [2,3]. Lindane is the gamma isomer of hexachlorocyclohexane (HCH) which has been widely used as a scabicide and a delousing agent in the form of lotions and shampoos [4]. It has been applied in agriculture as insecticide and in veterinary to control insect-borne diseases, particularly in developing countries [5–7]. Lindane, as a lipophilic compound, can bio-accumulate in the food chain and consequently in fat tissues of humans. Contact with this synthetic chemical has been associated to adverse effects on human health such as immunosuppressive and neurological problems [4]. United States Environmental Protection Agency and World Health Organization (WHO) have classified lindane as a powerful carcinogenic and teratogenic chemical [8]. Although

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lindane has been currently forbidden in many countries, due to the economic causes it is still consumed in some part of the world and subsequently water, sediments, soil, plants and animals are being polluted [9–11]. Many researchers have reported the existence of various OCPs such as lindane in surface, ground and drinking waters [12,13]. It has been recommended that quantity of lindane in surface and ground waters should not exceed 0.98 μg/L [14]. The WHO has also set the value of 2 μg/L as the maximum permissible concentration for lindane in drinking water [15]. Because of the adverse health impacts of lindane, the removal of this compound in the environment, particularly in drinking water, is vital. Therefore, many treatment methods such as advanced oxidation processes, adsorption, biological treatment, membrane reactors, etc., have been effectively applied for the removal of OCPs in water [5,13,16–20]. Slow sand filtration (SSF) is mostly used in water treatment in rural communities. Application of this treatment procedure is well documented for the removal of various organic matters in water. Since, the product water of slow sand filter is nearly harmless for consumption [21]. Slow sand filters can nurture various microorganisms into the media and subsequently it can degrade pesticides and their degradation products [22]. As water penetrates through sand media of the filter by gravity force, organic material is removed by various treatment methods including mechanical (adsorption, diffusion, screening and sedimentation) and biological processes [23]. Several studies revealed that the sand filtration in water treatment can omit various organic chemicals, such as pesticides in drinking water [21,24–26]. In a study by Feld et al. [25] on the removal of four common pesticides by the sand filter in water, they indicated that 2,4-dichlorophenoxyacetic acid (2,4-D), aldicarb, chlorpyrifos and diazinon have been removed by this treatment method. Therefore, this study was applied to remove a common OCP, lindane, in water by SSF. The effects of various parameters including lindane concentration, turbidity, filter bed depth and Schmutzdecke type were assessed on the removal of lindane by SSF.

2. Materials and methods

2.1. Materials

The mineral sand (quartz), as the main media of SSF in rural water treatment plants, was purchased from Shimi Gostaran Pars Co. (Iran). Kaolinite (for the production of water turbidity) was provided from Tehran Ramfar Co. (Iran). Lindane powder (97%) was obtained from Sigma-Aldrich Co. (USA). Table 1 shows the physical and chemical characteristics of lindane [27]. The lindane stock solution (1,000 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 with drinking water and applied for the experiments.

2.2. Purification of sand

First, 50 kg of raw sand was washed several times with distilled water. Then, in order to remove more impurities, it was washed by HCl solution (0.1 N) [28]. The sample was finally rinsed with distilled water to neutralized pH and dried in an oven at 150°C for 2 h. In order to use the media in the experiments, the sand was graded through screens to obtain mesh size of 45–100 (0.15–0.35 mm). The characterization of filter media (sand) is presented in Table 2.

2.3. Experimental setup

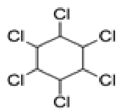
2.3.1. Pilot facility

Two sand filter pilots with a similar media and design were used to remove lindane from water. The schematic of slow sand filter (SSF) in this study is shown in Fig. 1. The pilots were made from plexiglass with height and diameter of 160 and 10 cm, respectively. The wall thickness of the filter was 5 mm. As seen from the figure, the sand layer of the filter was 100 cm deep. The height of water on the filter media was also 50 cm with 10 cm of free board. The five water sampling points (with diameter of 1 cm) into the sand bed circumference were located in the 20 cm intervals beneath the filter sand media. A metal plate with diameter and pore size of 10 cm and 0.1 mm was placed in the bottom of the filter media to avoid the sand leakage. A 50 L plastic tank was used as a water reservoir and the water from the tank was pumped through a plastic tube (with diameter of 2 cm) into the filter. A bypass line was also applied in the upper surface of the filter water in order to prevent water overflow from the filter.

Table 2
Characteristics of mineral sand in the filter media

Sand type	Quartz
Particle shape	Irregular
Effective size (mm)	0.25
Uniformity size	2.5
Density (g/cm^3)	2.65
Porosity (%)	43

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

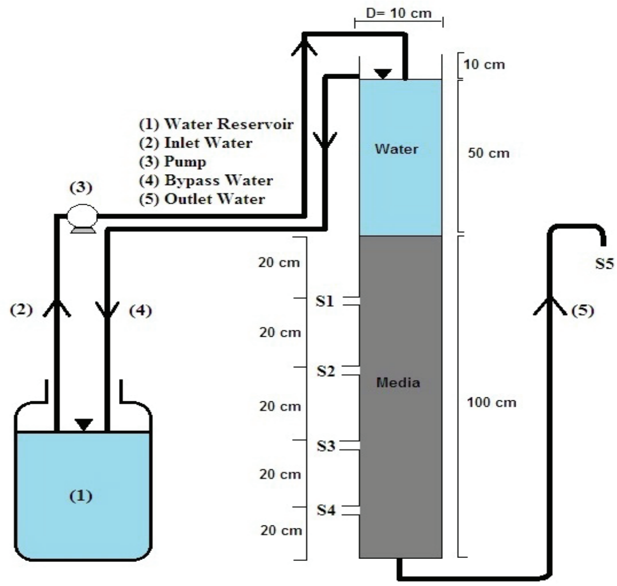


Fig. 1. The schematic of slow sand filter in this study.

2.3.2. Operation and sampling

For the removal of lindane in water, product water from municipal water treatment plant that passed through screening, coagulation–flocculation, sedimentation and filtration treatment methods was used in this study. The characteristics of the raw water are listed in Table 3. Lindane was spiked into the water at the concentration of 20–200 μg/L and then pumped into the filter with fixed water rate of 0.1 m/h. The effects of water turbidity, microbial quality and Schmutzdecke type were also investigated on the removal of lindane from water by SSF. For this purpose, the pilot study was performed in three stages as follows: in the first step, the lindane-containing water with concentration of 200 μg/L was pumped into the filter media without the Schmutzdecke layer and the adsorption capacity of the filter media was assessed for the removal of lindane. In the second step, the effect of filter media containing Schmutzdecke layer was evaluated for the lindane removal (20, 100 and 200 μg/L) in the presence and absence of water turbidity and bacteria. In the second step, first no-lindane water was passed into the filter for 30 d to form a Schmutzdecke layer and then water containing the abovementioned concentrations of lindane was passed through the filter column for 5 d. The third step was conducted such as the second step, except that the modified (acclimated) Schmutzdecke layer was applied on the filter media. The later step was performed for lindane concentration of 200 μg/L. In all the steps, the sampling of water (10 mL) for the lindane analysis was carried out along the filter media at five sampling points (S_1 to S_5).

2.3.3. Preparation of microorganisms

In one part of the experiments (third step in section 2.3.2), modified Schmutzdecke was used on the filter media and the effect of the media was assessed on the removal of lindane.

Table 3
Characteristics of raw water used in this study

Parameter	Value
Turbidity (NTU)	0.2
pH	8.0
Total dissolved solids (mg/L)	356
Electrical conductivity (μS/cm)	558
Total organic carbon (mg/L)	2.6
Hardness (mg/L $CaCO_3$)	261.6
Alkalinity (mg/L $CaCO_3$)	132.6
Sulphate (mg/L)	119.1
Chloride (mg/L)	10.5
Nitrate (mg/L NO_3-N)	8.5
Nitrite (mg/L NO_2-N)	0.005
Fluoride (mg/L)	0.4
Calcium (mg/L)	68.2
Magnesium (mg/L)	21.9
Sodium (mg/L)	8.3
Potassium (mg/L)	0.2
Iron (mg/L)	0.05
Manganese (mg/L)	0.002

For this purpose, the Schmutzdecke of SSF was acclimated in lindane solution in laboratory. Therefore, the amount of Schmutzdecke was poured in a nutrient solution containing the mineral materials and also lindane as source of carbon. The composition of the nutrient solution was made with 1 L of tap water as follows: $(NH_4)_2SO_4$ (2.36 g/L), KH_2PO_4 (2.09 g/L), NH_4Cl (1.91 g/L), $Na_2HPO_4.7H_2O$ (0.27 g/L), NaCl (0.5 g/L), $CaCl_2.2H_2O$ (0.03 g/L), $MgSO_4.7H_2O$ (0.2 g/L), $ZnSO_4.7H_2O$ (0.03 g/L), $FeCl_3.7H_2O$ (0.03 g/L), $MnSO_4$ (0.03 g/L) [29]. 5 g of the wet Schmutzdecke were placed in the abovementioned solution containing 10 μg/L of lindane. The suspension was then shaken on a rotary shaker (100 rpm) for 24 h at room temperature. After the mixing time, the suspension was filtered and poured in a similar solution containing 25 μg/L of lindane. The acclimation of the Schmutzdecke was conducted according the above method for the lindane content of 50, 100 and 200 μg/L. The modified Schmutzdecke was finally separated in the solution and then placed onto the filter media for the removal of lindane.

2.4. 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 Korea) equipped with ^{63}Ni electron capture detector (ECD) system and split–splitless injector was applied to measure lindane concentration (with detection limit of 0.005 μg/L) 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 × 5 μm) 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. The recovery values and the coefficients of variation in this study were 97.4%–104.6% and lower than 5% by GC/ECD, respectively.

Turbidity of water was measured by turbidimeter (2100P, Hach, USA). Microbial analysis (heterotrophic plate count, HPC) was also carried out by pour plate method (# 9215B), briefly as follows; 1 mL of the water sample was poured into 10 cm petri dish and 10 mL of culture medium (R2A agar with temperature of 45°C) was also placed into the plate. The sample and culture were mixed and then incubated at 35°C for 48 h [30].

3. Results and discussion

3.1. Adsorption potential of filter column

In the experiments, the adsorptive capacity of the filter media in the absence of the Schmutzdecke layer was investigated for the lindane removal. The lindane-containing water with concentration of 200 μg/L was pumped into the filter media. The sampling was taken up from five locations along the media during research time of 0 (zero time equals the water discharge of S_1 to S_5 for the first time) to 24 h. The results are shown in Fig. 2. As seen, the removal efficiency of lindane was increased by increasing time up to 6 h and after that time, the performance of the media was decreased due to the media saturation. The findings showed that the removal efficiency of the media was 91.24% for the run time of 6 h and this value was decreased to 45.34% for 24 h. The results of batch sorption experiments for filter media showed that the sorption capacity of the sand was 1.96 μg/g (data not shown). Fig. 2 also indicates that by increasing the filter depth, the removal efficiency of lindane was enhanced, so that, the concentration of lindane in the various media depth was as $S_1 > S_2 > S_3 > S_4 > S_5$ for any sampling run.

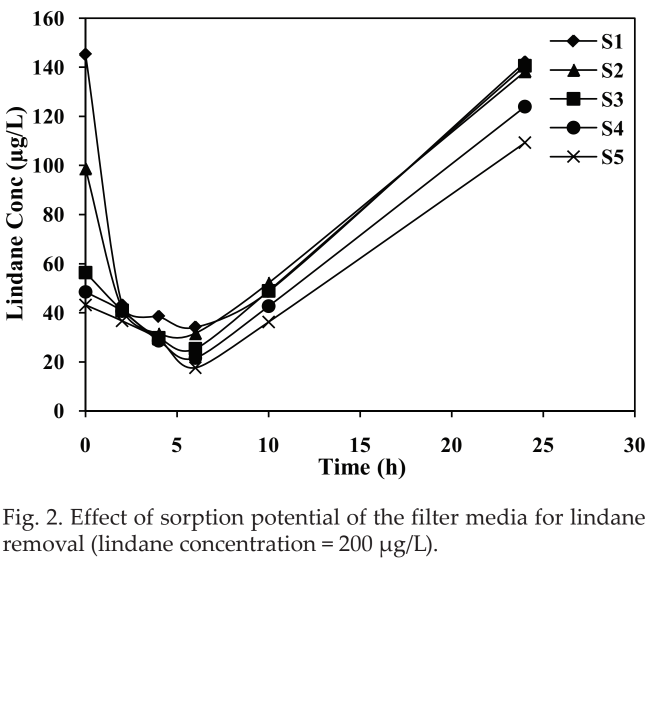


Fig. 2. Effect of sorption potential of the filter media for lindane removal (lindane concentration = 200 μg/L).

3.2. Effect of filter column covered by Schmutzdecke layer

For the production of Schmutzdecke layer on the filter, the water without lindane was passed through the filter for 5 d. The lindane-containing water was then treated by the filter column. Figs. 3(a) and (b) indicate the removal percent of lindane by filter media covered by Schmutzdecke layer in water with and without turbidity. The concentration of lindane in water was 200 μg/L. The results of Fig. 3(a) show that the filter caused the lindane removal by 85.4% over period of 1 d and this efficiency was nearly constant by increasing time up to 5 d. In a study conducted by Hedegaard and Albrechtsen [26] on the removal of some pesticides by sand filtration in water, they reported that the filter media could decrease MCPP (metabolites mecoprop), bentazone, glyphosate and *p*-nitrophenol to 42%–85%, 15%–35%, 7%–14% and 1%–3%, respectively, within operating time of 6–13 d. Hedegaard and Albrechtsen [26] also proposed that sand filtration can be used for treatment of pesticide in contaminated groundwater. Feld et al. [25] also expressed that the performance of sand filtration in

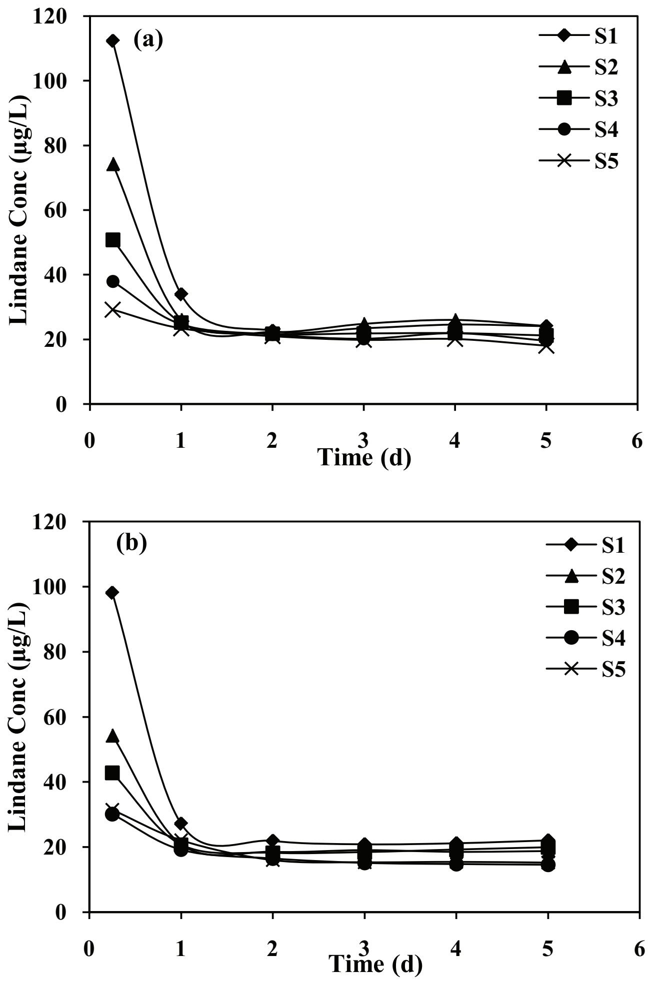


Fig. 3. Removal of lindane by filter media covered by Schmutzdecke layer in water (a) without turbidity (lindane concentration = 200 μg/L) (b) with turbidity (lindane concentration = 200 μg/L, turbidity = 5 NTU).

the removal of phenoxypropionate herbicides of DCPP, MCPP and 4-CPP was 15%–30%. Vandermaesen et al. [22] studied on the removal of 2,6-dichlorobenzamide (BAM), bentazone and 2-methyl-4-chlorophenoxyacetic acid (MCPA) in water by sand filter showed that 65%, 24% and 57% of the pesticides removal occurred for BAM, bentazone and MCPA, respectively. Nakamoto et al. [31] stated that up to 98% of methaldehyde pesticide was removed from water by SSF. In a study by Phu [32] on the removal of various pesticides in tertiary treated wastewater by SSF, the results showed that the mean removals of pesticides of atrazine, diazinon, 2,4-D, metolachlor, chlorpyrifos and bromacil were 40.84%, 58.58%, 51.69%, 38.26%, 44.78% and 41.20%, respectively. Fig. 3(b) also presents the effect of water turbidity on the lindane removal by filter media with Schmutzdecke layer in the lindane-containing water (200 μg/L). As seen, water turbidity up to 5 NTU had a negligible effect on the removal of lindane in comparison with the absence of turbidity in Fig. 3(a). Filter depth had a positive effect on the removal of lindane up to the first day (Fig. 3), it may be due to the sorption capacity of the sand for lindane. After first day, the filter depth had no significant effect on the removal of lindane, because of the saturation of the sand by lindane in lower layers of the filter. The reason that lindane only was substantially removed in the upper layer of the filter (20 cm upper the filter) can be due to the availability of Schmutzdecke layer and subsequently biological activity for the degradation of the pesticide in this part. The results also showed that the filter media decreased water turbidity and HPC from 5 to 0.1 NTU (removal efficiency of 98.0%) and from 700 to 12 CFU (removal efficiency of 98.3%), respectively (data not shown in the figure). Figs. 4(a) and (b) also present lindane removal by the filter contains Schmutzdecke layer for various concentrations of inlet lindane (100 and 20 μg/L). As can be seen, the removal percentage of lindane by the filter for inlet lindane concentrations of 100 and 20 μg/L was higher than lindane concentration of 200 μg/L. The removal efficiency of the filter media for inlet lindane of 100 and 20 μg/L was 95.4% and 100% over 1 d operation. It can be concluded from Figs. 3 and 4 that SSF containing Schmutzdecke layer cannot decrease lindane concentration in drinking water below the WHO guideline (2 μg/L) when lindane concentration in inlet water was higher than 100 μg/L. But, SSF can meet the WHO guideline for lindane-containing water up to 20 μg/L.

3.3. Effect of filter column covered by modified Schmutzdecke layer

The effect of filter media covered by the modified (adapted) Schmutzdecke layer for the removal of lindane (200 μg/L) is depicted in Fig. 5. As presented, 96.3%–97.8% of the inlet lindane was decreased only over the operation time of 1 d and after that the removal increased to 98%–99% up to the time of 5 d. The removal efficiency of lindane by the SSF modified Schmutzdecke layer was nearly 15% more than that of SSF with the natural Schmutzdecke layer. The adaptation of the Schmutzdecke with different concentration of lindane in laboratory was effective for the removal of lindane in the column.

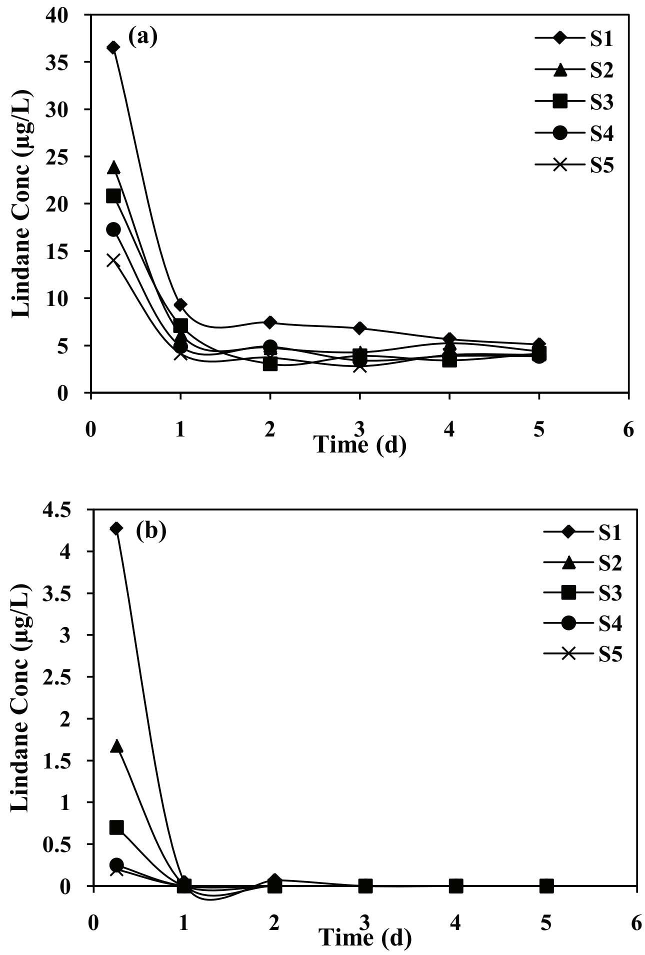


Fig. 4. Effect of filter media covered by Schmutzdecke layer for the removal of turbidity containing water (turbidity = 5 NTU) and (a) lindane concentration of 100 μg/L and (b) lindane concentration of 20 μg/L.

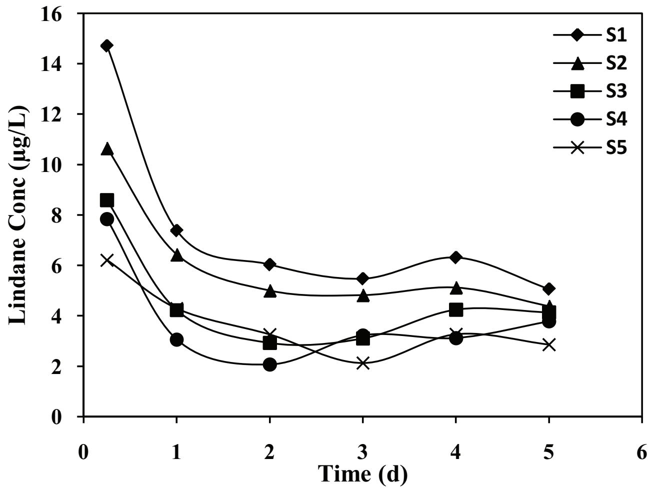


Fig. 5. Effect of filter media covered by modified Schmutzdecke layer for the removal of lindane-containing water of 200 μg/L and turbidity (5 NTU).

4. Conclusion

This study was performed to remove lindane in water by SSF. The influences of different factors such as lindane concentration, turbidity, filter bed depth and Schmutzdecke type were determined on the removal of lindane. The results indicated that the filter media caused the removal of 85.4%, 95.4% and 100% lindane with inlet concentration of 200, 100 and 20 μg/L, respectively, over 1 d after operation. The increasing water turbidity (from 0.2 to 5 NTU) and filter depth more than 20 cm had no significant effects on the removal of lindane. SSF with the modified Schmutzdecke could remove lindane 15% more than that of SSF with the natural Schmutzdecke. On the basis of the results, SSF can meet the WHO guideline for lindane in water up to lindane concentration of 20 μg/L.

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