RESEARCH PAPER

Application of Physical and Biological Methods to Remove Heavy Metal, Arsenic and Pesticides, Malathion and Diazinon from Water

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Abstract

Environmental pollution caused by heavy metals and pesticides have increased concerns over potential toxicity to a range of species, and their removal from water is of unquestionable importance. The present study was attempts to remove heavy metal, Arsenic and pesticides, Malathion and Diazinon from water using physical and biological methods. The physical remove of the pollutants was carried out through a particle trapping systems, contained straws to trap the fine and large particles. The system received 3 g montmorillonite as the absorbent media each 24 h during 10 days. A media containing algae, *Anabaena* sp. was also used for the biological treatment of the pollutants. The results showed that the montmorillonite particles in the particle trapping system were able to reduce the initial concentrations of pollutants. The removing efficiency of 78% and 58% were measured respectively for initial concentrations of 1 and 2 g/L Arsenic in the biological system. The biological treatment also reduced the initial levels of pesticides with the reduction efficacies of 68.3% and 71.6% for Diazinon and 40.5% and 67.8% for Malathion, respectively. In the both systems, pH increased gradually with the removal of contaminants. The results are discussed with regard to potency of particle trapping system supplemented montmorillonite and algae removal of pollutants from water solutions.

Introduction

Large quantities of toxic effluents have been generated in response to increased use of organic and inorganic compounds in the process industries and agricultural areas. Due to non-degradable and persistence nature, pollution of the aquatic environment is of the main problems in relation to ecosystem, animals and human health (Reddy & Behera, 2006). Pesticides such as Diazinon (Dz) and Malathion (Mt), and heavy metals like Arsenic (Ar) have been recognized as major contaminants of surface and underground waters (Real, Benitez, Acero, & Gonzalez, 2007). These pollutants affect adversely many aspects of life including growth, immune system, and reproduction in human and aquatic organisms (Ullah & Zorriehzahra, 2015). More than 6,000 tons of Dz is used annually in the United States. Also Mt composes 32-44% of total organophosphorus pesticide in the US (Zhang & Pagilla, 2010). In Iran, a Dz concentration range of 77.6-101.6 μg/L with a maximum concentration of 768.9 μg/L has been measured in surface water of the Babolrood River. Furthermore, a concentration of 506.6 μg/L has been also reported for Mt (Fadaei *et al.,* 2012). Among heavy metals, Ar is one

of the most common elements in the earth's crust (Nriagu *et al.,* 2007). Ar is a toxic and carcinogenic substance and cause damage to the nervous system, liver and skin, leading to cancer and death (Sharma, Singh, & Siddiqi, 2014). Chronic exposure to Ar through drinking water is the most important cause of Ar toxicity (Kapaj, Peterson, Liber, & Bhattacharya, 2006). Therefore, environmental protection agencies and national standards have declared an acceptable level of <10 μg/L for concentration of Ar in drinking water (Bhowmick *et al.,* 2014). Due to the serious effects of water pollution on aquatic life, it is essential that simple, quick and economical methods be develop to remove contaminants including pesticides and heavy metals.

At present, different techniques have been used to remove contaminants from water. Absorption, oxidation, flocculation/sedimentation, filtration and bio-accumulation are of the main methods to remove pesticides (Moussavi, Hosseini, & Alahabadi, 2013). Also, various methods including sedimentation, absorption, bio-absorption, reverse osmosis and ion exchange are available to reduce Ar levels in water (Mondal, Bhowmick, Chatterjee, Figoli, & Van der Bruggen, 2013). Among these methods, absorption is

generally more suitable due to the simplicity, high efficiency and cost-effectiveness (Mohan & Pittman, 2007). Low cost and high abundance in nature caused widely use of clay minerals as the absorbents (Hu & Luo, 2010). Being widespread in nature, montmorillonite (MON) has a size of about 10 Å which forms the major phase in bentonite. Due to the high potential of absorption and negative charge at the surface, and ionic exchange ability it can absorb organic cations (pesticides, phenols, etc.) and inorganic cations such as heavy metals (Yin & Deng, 2015).

In addition to the chemical and physical methods, biological methods have been suggested as an efficient strategy to remove pollutants from water. Yeasts, bacteria, fungi and algae species can absorb and accumulate pollutants in their body. Among the biological methods, using algae is very effective, reliable and predictable in removing pollutants from water (Salman & Abdul-Adel, 2015). Microalgae, such as blue-green algae, can decompose organophosphorus compounds (such as pesticides) as source of phosphorus and carbon (Salman & Abdul-Adel, 2015). Also, cheapness and low sludge production are the main advantageous of biological treatment of heavy metals (Ungureanu, Santos, Boaventura, & Botelho, 2015). *Anabaena* sp. algae have a lot of potential to use as absorbent in contaminated water (El-Sheekh, El-Shouny, Osman, & El-Gammal, 2005). Removal of heavy metals and nutrients by the *Anabaena* and some other Cyanobacteria algae is strongly recommended as a powerful technique for the removal of pollutants from wastewater (Fawzy & Issa, 2016; Cepoi, Donţu, Şalaru, & Şalaru, 2016). Therefore, they may play a major role in the removal of organic and inorganic pollutants from water.

In the present study, we have applied a physical and a biological method for removing Ar, Dz and Mt from water. In the physical method, MON was used as the absorbent media. In the biological method, algae,

Anabaena sp. was used as the absorbent. *Anabaena* has a lot of potential for removing different kinds of pollutants (El-Sheekh *et al.,* 2005; Salman & Abdul-Adel, 2015).

Materials and Methods

Physical Method for Removing Ar, Dz and Mt

The physical remove of the pollutants was carried out using a particle-trapping system (PTS). The system consisted of two containers of 30 × 20 × 25 cm which was partitioned into three parts including 1- water entrance, 2- particle-trapping part (PTP) contained PVC-based straws, and 3- pumping part. Water was introduced from the water entrance, and after passing through the PTP, it entered into the third part of the container, where it was flooded back to the first part (water entrance) using an electric pump (capacity of 700 L/h) (Figure 1). The depth of water in the PTP was always constant. Two forms of PTS were used that differed in the capacity of PTP to remove particles with a diameter of more (large-PTP) or less (fine-PTP) than 50 μg.

Stock solutions of Ar ($AsCl_3$; 1 and 2 g/L), and Dz and Mt (60% purity, Raja-Shimi Co., Iran; 10 and 1000 mg/L) were prepared and added to the PTS filled with 10 L doubly distilled water. Then, the pump was turned on and the system start to circulate water. In order to identify the actual concentration of metal, 100 ml of water from PTS was sampled 2 min after complete mixing of water in three parts of the PTS. Ten days removal experiment was considered in which 100 ml of water was collected from PTS to assay the pollutants and water pH every 24 hours. A mixture of clay, MON (3 g; Pasargad Chemical Co. Iran; cationic exchange capacity: 0.958 meqg^{-1}; 500 μm diameter), was added to the PTS after each sampling time (Chen et al., 2016).
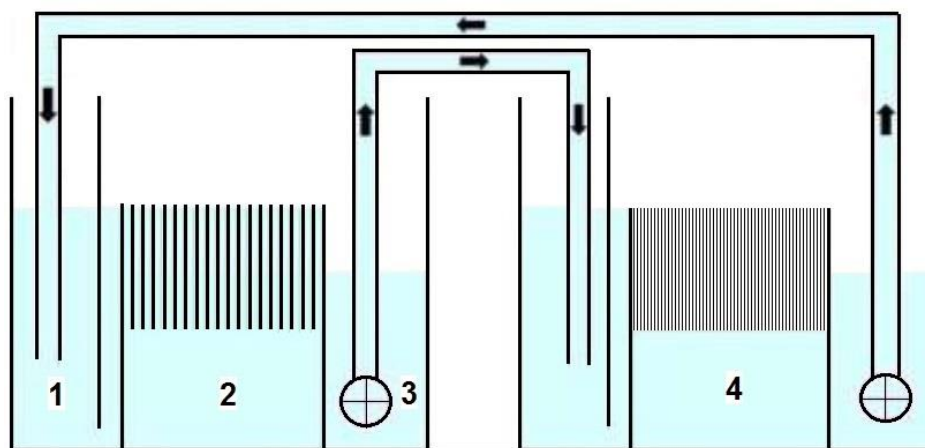
Figure 1. Schematic of the particle-trapping system (PTS). 1: Water entrance part, 2: particle-trapping part (PTP) of the first container, large-PTP; 3: pumping part, and 4: PTP of the second container, fine-PTP.

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Biological Method for Removing Ar, Dz and Mt

Algae, *Anabaena* sp., were used to biologically remove Ar and pesticides from water. To this end, a system consisted of a container of 30 × 20 × 30 cm was established for each pollutant (Figure 2). The containers were partitioned into three parts including 1- water entrance, 2- algal media (PTP; contained halm-based straws), and 3- pumping part. The initial concentrations of Ar and water flow through the system were similar to the physical treatment experiment; however, 6 and 600 mg/L pesticides were used in biological treatment. Algae media included strips of 20 × 4 cm in size which were covered with *Anabaena* (100 g; obtained from Limnology Laboratory of University of Tehran) to make a bio-media (Figure 2). Furthermore, a light source including a fluorescent lamp of 40 W was placed at a distance of 15 cm from the surface to stimulate photosynthesis in the algae.

Concentration of the Pollutants and Water pH

Concentrations of As (Luengo, Puccia, & Avena, 2011) and Dz and Mt (Venugopal, Sumalatha, & Bonthula, 2012) were spectrophotometrically measured with UV/Vis 2100 spectrophotometer (Unico, USA), and a pH-meter (Hanna instruments, Italy) was used to measure pH throughout the study.

Statistical Analysis

All the experiments were performed in triplicates. Data analysis was carried out by SPSS software (IBM statistical). Before analysis, data normality was evaluated by Kolmogorov-Smirnov test. Then, Multivariate Analysis of Variance (MANOVA) was applied to compare the means. When significant F-ratios were calculated by MANOVA, the Duncan test was used to determine which means were different at $P<0.01$.

Results

Physical Treatment

Remove of Ar

The results indicated a significant effect of the initial concentration of Ar and the duration of the process in removing Ar ($P<0.01$) in PTS. The interaction of these two factors also had a significant effect on the amount of Ar removal ($P<0.01$).

The initial concentration of Ar (1 and 2 g/L) reduced to 43 ± 2.51 mg/10L and 142 ± 8.04 mg/10L equal to 57% and 29% during 10 days treatment, respectively (Figure 3). This concentration was recorded as the minimum level in this experiment. The removal efficiencies of 45% and 27% were measured during first 5 days of the experiment for initial concentrations of 1 and 2 g/L Ar, respectively. These efficiencies were 14% and 5% for the second 5 days of the experiment.

Remove of Dz and Mt

Results showed the significant effects of initial concentrations of pesticides ($P<0.001$) and duration of the experiments ($P<0.001$) on the amount of treatment; however, no significant effect was observed for the type of the pesticides ($P = 0.084$). Also, initial concentrations × exposure time to absorbent ($P<0.01$), initial concentration × type of pesticides ($P<0.05$), and initial concentrations × exposure time to absorbent × type of pesticides ($P<0.05$) interactions were significant, but exposure time to absorbent × type of pesticides was not significant ($P = 0.072$) on the amount of removal.
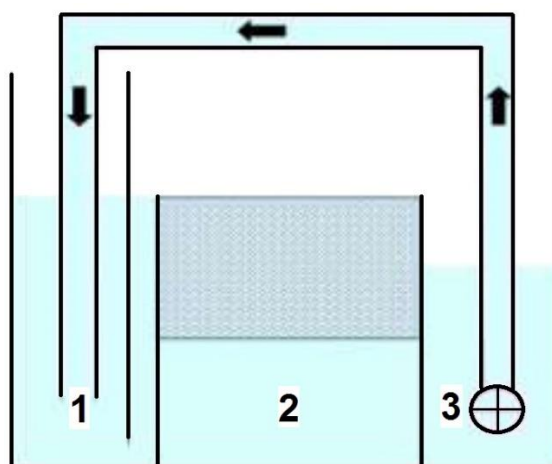
Figure 2. Schematic representation of the biological system. 1: Water entrance part, 2: strips covered with algae media; 3: pumping part.

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In PTS, the initial concentration of Dz (10 mg/L and 1000 mg/L) reduced to 5.2 ± 0.001 mg/L (48% decrease) and 360± 0.003 mg/L (64% decrease) during 10 days experiment respectively (Figure 4). Also, the initial concentration of Mt (10 mg/L and 1000 mg/L) decreased to 5.87 ± 0.001 mg/L (41.3% decrease) and 313 ± 0.002 mg/L (68.7% decrease), respectively (Figure 4).

Biological Treatment

Remove of Ar

The results of MANOVA showed the significant effect of algae in reduction of Ar in water (Figure 5). In this regard, the initial concentration of Ar (1 and 2 g/L) reduced to 22 ± 2.07 mg/10L (78% decrease) and 83.1

± 4.002 mg/10L (58.5% decrease) during 10 days experiment. Also, the removal percentage of 61% and 45% were measured during the first 5 days of experiment for initial concentrations of 1 and 2 g/L Ar, respectively. These efficiencies were 45% and 24.5% for the second 5 days of the experiment.

Remove of Dz and Mt

Significant effects of the initial concentrations (P<0.001), duration of exposure to absorbent (P<0.001) and type of the pollutant (P<0.01) were significant on the remove of pesticides from water. All of the interactions, except for the exposure time × type of the pollutant (P=0.095), were statistically significant (P<0.01).

The results indicated the significant effect of
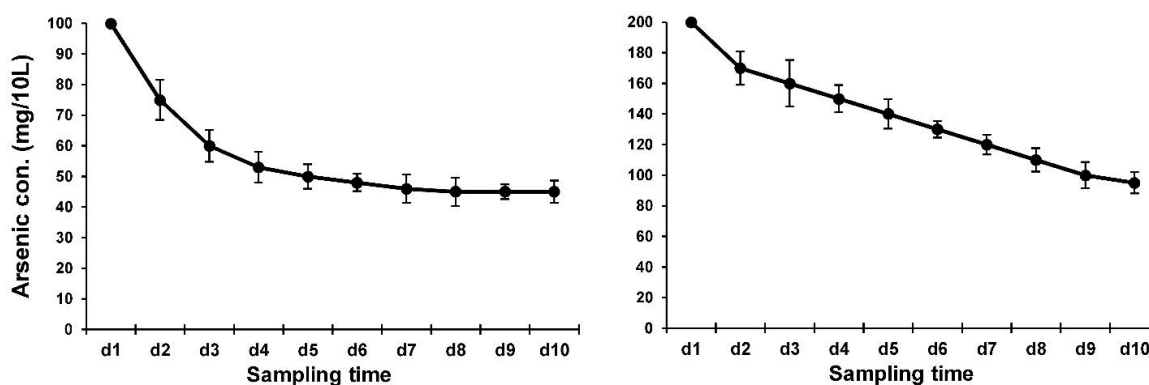
Figure 3. The changes in the concentration of Arsenic (Ar) during 10 days removing treatment by montmorillonite as adsorbent in the particle-trapping system. The values of Ar are presented as mean ± standard deviation in each sampling.
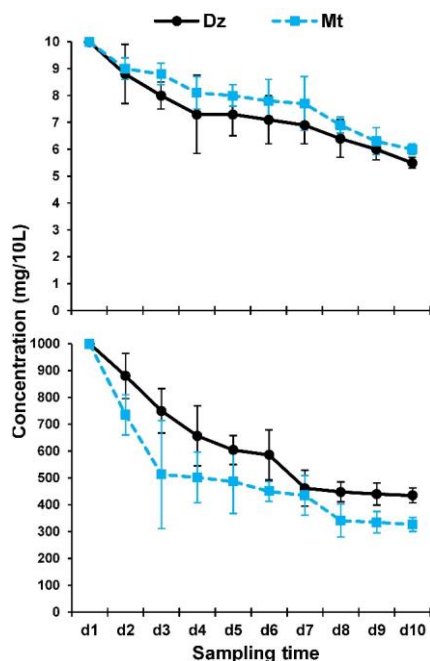
Figure 4. The changes in the concentration of diazinon (Dz) and malathion (Mt) during 10 days removing treatment by montmorillonite as adsorbent in the particle-trapping system. The values are presented as mean ± standard deviation in each sampling.

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algae in reduction of Dz and Mt in water (Figure 6). In this regard, the initial concentration of diazinon (6 and 600 mg/L) reduced to 3.1 ± 0.002 mg/L (71.6% decrease) and 190 ± 4.005 mg/L (68.3% decrease) during 10 days treatment. The initial concentration of Mt (6 mg/L and 600 mg/L) decreased to 1.93 ± 0.002 mg/L (67.8% decrease) and 356.8 ± 0.004 mg/L (40.5% decrease) during 10 days experiment, respectively.

Water pH Changes

Changes in water pH in all the experiments were showed in Figure 7. In physical method, the water pH value increased over time due to the activity of the MON adsorbent, so that the duration of the experiment, the initial concentration of Ar, and the interaction of the two factors had significant effects on water pH.

The initial concentration of Ar had no significant effect on the pH value when *Anabaena* was used (P=0.19). However, the duration of experiment had a significant effect on the water pH (P<0.05). Interaction of initial concentration and duration of experiment also had no significant effect.

The initial concentration of Dz had a significant effect on the water pH in the biological treatment system (P<0.01), so that the pH of the water containing 600 mg/L was always lower than that of 6 mg/L treatment. However, the duration of experiment and interaction of Dz initial concentration × duration of experiment were not significant. For Mt, different concentrations of pesticide did not have a significant effect on water pH, but the effect of the duration of the experiment was significant (P<0.05). The interaction of the concentrations of Mt and duration of the experiment on water pH was not significant.
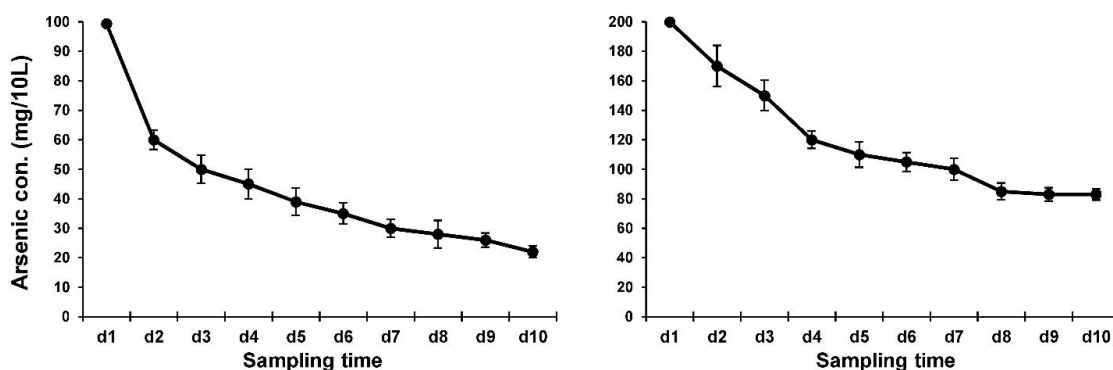

Figure 5. The changes in the concentration of Arsenic during 10 days removing treatment by algae, *Anabaena* as adsorbent. The values are presented as mean ± standard deviation in each sampling.
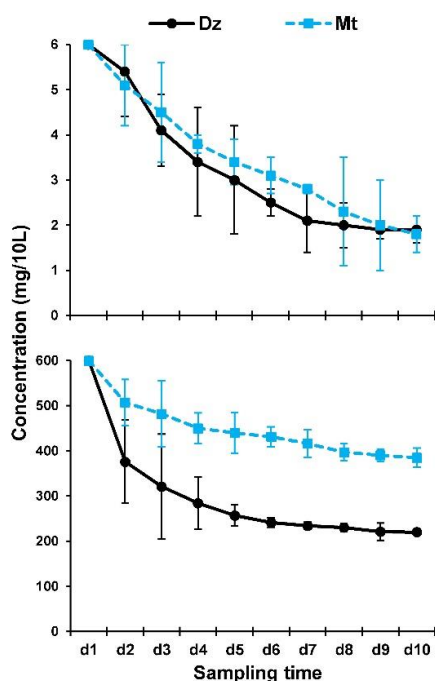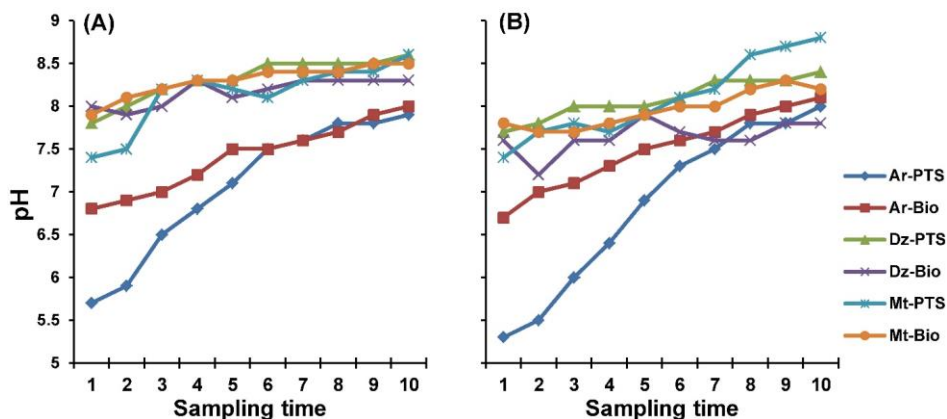

Figure 6. The changes in the concentration of diazinon and malathion (Mt) during 10 days removing treatment by algae, *Anabaena* as adsorbent. The values are presented as mean ± standard deviation in each sampling.

Figure 7. The changes in the water pH during 10 days removing of arsenic, diazinon and malathion by montmorillonite in particle trapping system or algae, *Anabaena* in biological system as adsorbents. (A) low initial concentration and (B) high initial concentration. The values of pollutants are presented as mean ± standard deviation.

Discussion

One of the practical ways in dealing with water scarcity is the reuse of sewage and wastewater. Sewage types such as oily, blood, dairy, and even surface wastewater contain many suspended particles that are sometimes found to increase during primary and secondary purification processes, which creates a problem in water purification and removal of turbidity. In previous approaches, by adding chemical substances in the secondary and third stages of treatment, more bulky and heavier compounds create to settle and filter in the next stage (Esplugas, Bila, Krause, & Dezotti, 2007). In "particle trap treatment system" according to the physical laws and conditions of forces and movement of water molecules and streams of microlamines and microturbolence, and simultaneous use of polyethylene media and algae-bacteria culture, we could carry out biological purification and simultaneous removal of turbidity from sewage. This approach, do not need to use chemicals (which are costly), and also the environmental problems associated to the previous systems do not exist.

The present study showed that arsenic and pesticides (malathion and diazinon) can be removed from the water in a short time by means of physical (PTS) and biological treatment (algae media) methods. Pollution removal efficiency was more than 50% in the most cases. Also, the absorption rate was higher in the early days of the experiment and gradually decreased to a steady state as reported previously (Habila, ALOthman, Al-Tamrah, Ghafar, & Soylak, 2015). Similar to the results of physical treatment in this study, various studies have shown that MON has a high efficiency in the removal of various pollutants (Jiang & Kim, 2008; Jiang & Ashekuzzaman, 2012). Li *et al.* (2012) used TiO$_2$-enriched MON and concluded that this material is an effective adsorbent for the removal of Ar from water. However, no study has yet been conducted to evaluate the simultaneous performance of clay particles and trapping systems for the treatment of heavy metals.

The results of this study showed that the use of algae as a biological method have significant effects in reducing the pesticides and Ar in water. Algae absorb significant amounts of nutrients especially high levels of nitrogen and phosphorus for the synthesis of the proteins (60-40% dry weight of algae), nucleic acids and phospholipids (Munoz & Guieysse, 2006). In addition, using micro-algae for removing the pollutants of water is an environmentally favorite process (Ungureanu *et al.,* 2015). There are many studies regarding the remove of Ar from water by aquatic plants such as *Callitriche lusitanica, C. brutia, Lemna minor, Azolla caroliniana* and *Fontinalis antipyretica* (Jasrotia, Kansal, & Mehra, 2017). It has been indicated that single-cellular green-blue Algae, *Synechocysis* sp. can efficiently remove Ar from water (Yin, Wang, Bai, Huang, & Sun, 2012). *Anabaena subcylindrica* has been used to remove heavy metals from sewage. This algae resulted 12.5%-81.8% reduction in copper concentration in water (El-Sheekh et al., 2005). Generally, the biological removal efficiency depends on the type of absorbent, physiological conditions of the cells, availability and concentration of metal and chemical composition of the aqueous solution (Kumar, Dahms, Won, Lee, & Shin, 2015). Based on above studies and the results of the present study, it can be concluded that the use of algae can efficiently remove Ar from water.

In the present study, the absorption efficiency of pesticide increased with increasing of pesticide concentration which can be due to the more exposure of pesticides with absorbent. Similar results were reported when activated carbon was used for absorption of malathion (Habila *et al.,* 2015). In the biological method, however, different results obtained where the absorption efficiency decreased with increase of pesticide concentration. This result can be attributed to the rapid saturation of algae cells from pesticide at higher concentrations (Salman & Abdul-

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Adel, 2015). Therefore, it is suggested that large amount of *Anabaena* algae are required to remove high concentrations of pesticides. As well, it should be noted that micro-algae have a high sensitivity to environmental pollutants and therefore the loss of biological function in high concentrations of pesticides can be due to the gradual death of algae (Safonova *et al.,* 2004). Unlike pesticides, low Ar concentrations were eliminated more efficiently which can probably be attributed to more exposure of Ar with absorbent surface in low concentrations. Heavy metals in high initial concentrations usually saturate the absorbent surface which consequently reduces the absorption efficiency (Salman & Abdul-Adel, 2015).

The duration of treatment with adsorbent materials is one of the most important factors affecting the absorption efficiency of pollutants (Singh *et al.,* 2010). In the present study, a period of 10 days was considered for each experiment which was suitable for gradual removing the pollutants. In both methods used, the removing efficiency of the pollutants in the first 5 days of the experiment was more than the second 5 days. These results were not surprising since rapid absorption of heavy metals at the beginning of the treatment is associated with a higher concentration gradient and more sites available for absorption by MON and algae cells (Li *et al.,* 2012; Kumar *et al.,* 2015; Ungureanu *et al.,* 2015).

It is well known that pH is a critical factor in the adsorption of Ar by MON and algae (Li *et al.,* 2012). Our results showed that with adsorption of Ar the water pH increased during 10 days experiment. Therefore, higher adsorption of Ar occurred at lower pH levels. In accordance with our results, some studies showed the higher removal of heavy metals in acidic pH (Ramesh, Hasegawa, Maki, & Ueda, 2007; Luengo *et al.,* 2011; Uddin, 2017). Also, it was also observed that water pH increased during 10 days absorption of pesticides, indicating cation exchange by MON and its active role in absorption. Similar results have been reported by Hassan, Abed, & Ismael (2009). In the present study, the increase in water pH was observed when algae were used as absorbent. This case can be attributed to photosynthesis and consumption of CO_2 by algae, indicating good efficiency of biological method (Molleda, Blanco, Ansola, & de Luis, 2008).

In general, the results of the present study showed that the use of clay montmorillonite in the PTS was effective in removing heavy metal Ar and pesticides Dz and Mt from water over a period of 10 days. The results of the algae purification also showed the ability of *Anabaena* sp. cells to treat the pollutants. According to the results of the present experiment, it can be concluded that the PTS along with montmorillonite has high efficiencies for removing high levels of the pollutants, and the biological treatment system may apply to remove low concentrations of them from water solutions.

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