



## MODELING THE TRANSPORT OF CONTAMINANT BY WASHING PROCESS IN THE SANDY SOIL

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### **ABSTRACT**

The aim of this study is modeling the transport of industrial wastewater in sandy soil by using finite element method. A washing technique was used to remove the industrial wastewater from the soil. The washing technique applied with an efficient hydraulic gradient to help in transport of contaminant mass by advection. Also, the mass transport equation used in modeling the transport of industrial wastewater from soil includes the sorption and chemical reactions. The sandy soil samples obtained from Al-Najaf Governorate/Iraq. The wastewater contaminant was obtained from Al-Musyiebelectricity power plant. The soil samples were synthetically contaminated with four percentages of 10, 20, 30 and 40% of the contaminant and these percentages calculated from the distilled water used in the soaking process. The soaking process continued for 30 days. The contaminated soil samples were washed by using distilled water applied with a hydraulic gradient of 0.5. A laboratory physical model was designed to study the removal efficiency of contaminant from the soil after 10 days of remediation. The percentages of removal efficiency of the contaminant from the soil are (97.63, 96.79, 96.58, and 93.87) %. A computer program presented by Smith and Griffiths (P8.8) was developed by taking into consideration both effects of adsorption and chemical reactions in solving mass transport equation. The results obtained from the developed computer program well agreed with those obtained experimentally in pattern and magnitudes. The effects of adsorption and chemical reactions are slight and have no effects on the quantity of contaminant mass transported by advection.

**Keywords:** modeling, wastewater, sandy soil, contamination, mass transport, washing.

### **INTRODUCTION**

The developments of modern life especially due to the connection of industries with other developments increase the opportunities for soil contamination.

External contaminants penetrating a soil body through a wet or dry climate that behave variously with regard to each soil type accordingly to the absorption properties, texture, density, humidity (Wuana and Okieimen, 2011). There are many methods used in the remediation of contaminated soils. The general vicinity impacts, the removal efficiency of contaminant, and the time required for remediation are the control factors in selecting the remediation technique. Deane *et al.* (1999) examined the transport and destiny of hydrophobic organic chemicals (HOCs) in soaked and consolidated soil sediments. The results showed high agreement between the experimental results and numerical modeling outcomes. In spite of the long time of tests, the results showed that the HCB in the steady-state synthetic balance more than consolidated sediment.

Belardi *et al.* (2000) used the soil-washing remediation method to treat silty sand soil contaminated by total petroleum hydrocarbons (TPH). The hydrogeological characterization of contaminated soil samples showed the presence of a gravel particles in the sandy/silty soil. The particles provide preferential paths to the water circulation. The results of study showed that the soil samples contaminated with TPH ranged between 2700 and 7000 ppm can be reduce the TPH concentration below the acceptable limits, where the soil returned to the site was around 90%. Conteet *et al.* (2005) studied the remediation of highly contaminated site around the chemical plant of

ACNA in Northern Italy. Two soils samples from the ACNA site, differing in texture and amount and type of organic contaminants were subjected to soil washings by using different washing solutions. The extraction of pollutants by sonication and soxhlet was conducted before and after the soil washings. Sonication resulted more analytically efficient in the fine-textured soil. Clean-up by water was unable to exhaustively remove contaminants from the two soils, whereas all the organic surfactants revealed very similar efficiencies (up to 90%) in the removal of the contaminants from the soils.

Moutsatsou *et al.* (2006) studied the washing of highly contaminated sandy soil by mining and metallurgical activities. Contaminant consists of slags, sulfur compound waste, and various low-grade lead condensates. The soil was washed with different solutions. The result of study showed high removal efficiency was obtained when hydrochloric acid even at the lowest concentration. Akpoveta *et al.* (2012) evaluated the efficiency of using sodium dodecyl sulfate (SDS) in remediating crude oil contaminated soil through surfactant enhanced soil washing technique. Crude oil contamination adversely affected the geotechnical properties of soil samples. The results showed that; the surfactant method was found to be very quick and effective with removal efficiency of 88.32% after seven hours. Antonucci *et al.* (2013) developed a one-dimension model to simulate the chelation process of metal (lead). Flushing remediation technique was applied to the soil. The flow and transport equations were solved simultaneously. The model was capable to estimate particularly the residual concentration



of lead in the soil at different depths and the concentration of the different types of species along the column of soil.

Karkush and Resol (2017a,b) studied the remediation of contaminated sandy soil by using washing process. The soil samples contaminated with four percentages of industrial wastewater. This contaminant was disposed as by product of the fuel used in the feeding of the electricity power plant. The results of experiments showed high removal efficiency of industrial wastewater from the soil. Karkush and Altaher (2017) studied experimentally the removal efficiency of total petroleum hydrocarbons from the clayey soil by using the washing-electokinetic technique. Different types of washing liquid used in this study. The results showed a removal efficient ranged between 17-53%. However, the present study is focusing on the numerical modeling of contaminant transport in the sandy soil remediated by using washing process. The mass transport equation solved numerically by using finite element method and finite difference method by adding the effects of chemical reactions to this equation.

### **Soil washing**

There remediation techniques can be classified into two principal classes: i) ex-situ and, ii) in-situ. The ex-site is related to the remediation of the soil off-site. In-situ clean-ups are often elect since they are generally made at low cost. However, they generally require long time to make the concentration of contaminants within allowable limits. On the other hand, excavating a contaminated area (ex-situ technique) and treating the material on the same site, this is called ex-situ, on-site or transporting it to another site for cleaning so that this is called ex-situ, off-site. The selection criteria for such technologies are based on the soil type, contaminant type, and concentration level of contaminant (USEPA, 1995; Reddi and Inyang, 2000; Sharma and Reddy, 2004). Three approaches improved for soil remediation. These approaches are categorized into thermal, physic/chemical, and biological approaches (USEPA, 2004). Also, it is practical to use combined techniques for successful effective remediation of some contaminants, to reduce the concentrations of contaminants to the limited levels (USEPA, 1995). If industrial waste comes in contact with the granular soils, the most popular soil remediation technique is soil washing (Riser-Roberts, 1998).

The process of soil washing may be defined as a physico/chemical treatment procedure that utilizations fluids, typically water, to flow throughout pores of soils. The contaminants typically sorbs to the surfaces of the sediment or soil particles instead of the coarser sand or rock particles. The purpose of soil washing is to separate the fine silt and clay particles from the coarser sand and gravel particles and then to transport and transfer of these chemical contaminants from the soil surface to the washing solution, so it's necessary to treat the solution used in soil washing by using different techniques. The concept of reducing soil contamination through the use of particle size separation is based on the finding that most organic and inorganic contaminants tend to bond, either

chemically or physically, to soil particles. Most silt and clay particles are become larger particles like sand and gravel. Washing separates the small particles from the large ones by breaking the adhesive bonds between particles.

### **EXPERIMENTAL WORK**

The soil samples are obtained from Al-Kufa district in Al-Najaf governorate in the south-west of Iraq. The soil samples are classified as sandy soil with organic matters (NTI Report, 2008). The groundwater level was recorded at levels of 2.0-2.50 m below the natural ground surface. Hand drilling was used to obtain soil samples from a depth of 1.75 m below the natural ground surface. Then, the soil samples were put in plastic containers and labelled with necessary data. The contaminant used in the present work is an industrial wastewater disposed from an electricity power plant in the middle of Iraq. The soil samples have been contaminated artificially with four percentages of (10, 20, 40 and 100) % of the wastewater calculated from the weight of the distilled water used in the soaking process, which will be termed later contaminant solution. The soil samples covered with contaminant for 30 days. The chemical and physical properties of contaminant are given in Table-1.

**Table-1.** Chemical and physical properties of contaminant.

Property	Value	Property	Value
EC, %	0.396	K, %	0.0084
Ni, %	0.0002	NO <sub>3</sub> , %	0.02
Ca, %	0.2101	Zn, %	0.0071
Cr, %	0.0001	Na, %	0.0228
Fe, %	0.1517	Mg, %	0.004
pH value	2.65	SO <sub>3</sub> , %	0.1172
Pb, %	0.0	Density, kg/m <sup>3</sup>	1001.3

The soil sample used in each test weighs 90kg soaked with 60kg of contaminant solution. Distilled water and industrial wastewater composed the contaminant solution. Four percentages of contamination solution are used in this study. The soil samples are designated as in the following:

- F0: intact soil sample.
- F1: soil sample contaminated with 10% of wastewater and 90% of distilled water.
- F2: soil sample contaminated with 20% of wastewater and 80% of distilled water.
- F3: soil sample contaminated with 40% of wastewater and 70% of distilled water.
- F4: soil sample contaminated with 100% of wastewater.

After 30 days of covering the contaminant solution to the soil sample, the soil samples are well mixed



in the container to insure good distribution for the contaminant in the soil samples. Then, the contaminated soil samples are air dried and remolded by adding the natural moisture content to get the field unit weight. The setup of proposed remediation technique and more details about the testing procedure are available in Karkush and Resol (2017a,b).

### Mass transport modeling

In this section, the mechanisms of contaminant migration in the soil and verifying the efficiency of selected remediation technique is discussed. This done by comparing the results of experimental work with a theoretical solution of the mass transport equation. To obtain a general equation of contaminant mass transport in the soil (Reddi and Inyang, 2000; Sharma and Reddy, 2004):

$$-\frac{\partial J_x}{\partial x} - \frac{\partial J_y}{\partial y} - \frac{\partial J_z}{\partial z} + r = \frac{\partial(nC)}{\partial t} \quad (1)$$

Where;  $J$  = mass transition of solute per unit area of cross-section;  $r$ =rate of large-scale manufacturing/utilization given by the dynamic model of response or compound response deliver amongst contaminant and soil,  $n$  = porosity, and  $C$  = is the solute concentration expressed as mass per unit volume of solute.

The flux of mass,  $J$  which resulted from dispersion and advection can be expressed as:

$$J \text{ due to advection} = v_i n C \quad (2)$$

$$J \text{ due to dispersion} = -n D_i \frac{\partial C}{\partial i} \quad (3)$$

Where;  $v_i$ = pore liquid velocity in the  $i$ -direction. Note that  $D_i$  in Equation 3 incorporate the two parts of dispersion and mechanical dispersion. Summing up the contributions from advection and dispersion, the mass fluxes in the three dimensions become:

$$J_x = v_x n C - n D_x \frac{\partial C}{\partial x} \quad (4.a)$$

$$J_y = v_y n C - n D_y \frac{\partial C}{\partial y} \quad (4.b)$$

$$J_z = v_z n C - n D_z \frac{\partial C}{\partial z} \quad (4.c)$$

Substitution of Equation 4 (a,band c) in Equation 1 yields:

$$-\left[ \frac{\partial}{\partial x} \left( v_x n C - n D_x \frac{\partial C}{\partial x} \right) \right] - \left[ \frac{\partial}{\partial y} \left( v_y n C - n D_y \frac{\partial C}{\partial y} \right) \right] - \left[ \frac{\partial}{\partial z} \left( v_z n C - n D_z \frac{\partial C}{\partial z} \right) \right] + r = \frac{\partial(nC)}{\partial t} \quad (5)$$

The velocity is assumed as uniform and the scattering coefficients don't shift in space, Equation 5 may be rearranged to:

$$\left[ D_x \frac{\partial^2 C}{\partial x^2} - v_x \frac{\partial C}{\partial x} \right] + \left[ D_y \frac{\partial^2 C}{\partial y^2} - v_y \frac{\partial C}{\partial y} \right] + \left[ D_z \frac{\partial^2 C}{\partial z^2} - v_z \frac{\partial C}{\partial z} \right] + \frac{r}{n} = \frac{\partial C}{\partial t} \quad (6)$$

The major reaction is sorption (reduction or consumption) and the minor reaction is chemical reactions. Chemical reactions include ions exchange and rate of reaction due to continuing the washing of the sample during the remediation process. These reactions happened due to the use of the acidic solution in the contamination of soil samples (Reddi and Inyang, 2000; Sharma and Reddy, 2004).

### Sorption

Sorption is a term utilized to incorporate every one of the procedures in charge of the mass exchange retention, adsorption, and particle trade. Sorption reactions are in general classified as either sorbent driven or solvent driven (Weber, 1991). When they are sorbent driven, the solid phase is the active one attracting the chemical, perhaps because of the presence of cation exchange sites on the solids. When they are solvent driven, the chemicals (usually nonpolar and organic) are hydrophobic in nature and therefore tend to associate with the nonpolar phase such as the organic content of the porous matrix (Reddi and Inyang, 2000; Sharmand Reddy, 2004).The theoretical equation used to fit the experimental linear isotherm of sorption is:

$$S = K_d C \quad (7)$$

Where  $S$  represents the value of sorbet mass (mg/g) on the surface,  $C$  represents the equilibrium of solution concentration,  $K_d$  represents the linear sorption isotherm slope and defined as the coefficient of distribution (ml/g).

### Chemical reactions

The existing of different chemicals in the soil matrix will response with each other to accomplish equilibrium, may exchange from one stage to the next among the three stages solids, fluids, and gases (Grove, 1977). An imperative thought in transport demonstrating is the rate at which the different transfer forms happen in soils. The period is required for a particular exchange process in respect to that of diffusion conditions and advection procedures will represent the major action in transport modeling. However, the diffusion and advection actions can be neglected in the modeling of mass transport equation. The main two principles of chemical reactions that happen in contamination issues are balance controlled particle trade responses with a direct adsorption isotherm and irreversible first-arrange rate responses. Mathematically, the descriptions of these reactions are direct or linear and permit the utilization of more straightforward numerical arrangement systems.

### Rate of reactions

The subsurface disposal of radioactive products is an example where a first-order irreversible rate reaction



occurs. This reaction is the radioactive decay of the species, adsorbed or in solution. The rate constant can be derived in the following manner. The disappearance of a species by a first order irreversible reaction is given by the following equation (Grove, 1977).

$$\frac{\partial C}{\partial t} = -kC \quad (8)$$

Where  $k$ = constant of the rate ( $T^{-1}$ ).This equation can be integrated with the limits chosen as the time necessary for the initial concentration to decrease by one-half.

$$\int_{C_0}^{C_0/2} \frac{dC}{C} = -k \int_0^{t_{1/2}} dt \quad (9)$$

Where, $t_{1/2}$ represents the half-life of species (T) and  $C_0$ represents the initial concentration ( $ML^{-3}$ ). The general solution of Equation 9 for k is:

$$k = \frac{0.693}{t_{1/2}} \quad (10)$$

The chemical reactions terms can now be incorporated into the mass-transport equation in the following manner. Assume the transporting of a chemical media is disappearing by the first order irreversible rate of reaction and it is starting to exchange reversible reaction by balancing-controlled procedure in which the exchange isotherm is straight. For 2D problem, Equation (6) becomes:

$$\left[ D_x \frac{\partial^2 C}{\partial x^2} - v_x \frac{\partial C}{\partial x} \right] + \left[ D_y \frac{\partial^2 C}{\partial y^2} - v_y \frac{\partial C}{\partial y} \right] - k \left( C + \frac{C\rho_b}{n} \right) = \frac{\partial C}{\partial t} + \frac{\rho_b \partial C}{n \partial t} \quad (11)$$

Where

$\rho_b$  represents the density of solid matrix,  $ML^{-3}$ ,  $\bar{C}$ represents the adsorbed concentrationin species, and C represents the dissolved concentrationin species. Equation 10 can be rearranged in the form.

$$\left[ D_x \frac{\partial^2 C}{\partial x^2} - v_x \frac{\partial C}{\partial x} \right] + \left[ D_y \frac{\partial^2 C}{\partial y^2} - v_y \frac{\partial C}{\partial y} \right] - kC \left( 1 + \frac{\rho_b k_d}{n} \right) = \frac{\partial C}{\partial t} \left( 1 + \frac{\rho_b k_d}{n} \right) \quad (12)$$

The retardation factor R is defined as:

$$R = 1 + \frac{\rho_b k_d}{n} \quad (13)$$

$$\left[ \frac{D_x \partial^2 C}{R \partial x^2} - \frac{v_x \partial C}{R \partial x} \right] + \left[ \frac{D_y \partial^2 C}{R \partial y^2} - \frac{v_y \partial C}{R \partial y} \right] - kC = \frac{\partial C}{\partial t} \quad (14)$$

### Mass transport solution by finite element

The finite element method is a technique used for solving boundary value problems in partial differential equations by first discretizing these fields in their space dimensions. Pinder (1973) used a Galerkin finite-element

technique with the use of isoperimetric quadrilateral elements to solve ground-water contamination problems. The resulting set of linear equations is solved by matrix technique. In the present formulation, the application includes introducing point sources or sinks and the contaminant is conservative in that chemical reaction involving the solute are absent. The element used in the present study is four nodded quadrilateral elements. The basic differential equation governing the mass transport with sorption and chemical reaction in two dimensions contains the variable C, its first and second spatial derivatives with respect to the coordinates x andy-axes. Also, it depends on the rate of change of C with time. Equation 14 can be re-written in the finite element matrix equation by using Galerkin approach.

$$\iint [NN^T d_x d_y] \frac{\partial C}{\partial t} = \left[ \iint \left[ \frac{D_x \partial^2 C}{R \partial x^2} - \frac{v_x \partial C}{R \partial x} \right] + \left[ \frac{D_y \partial^2 C}{R \partial y^2} - \frac{v_y \partial C}{R \partial y} \right] - kC \right] d_x d_y \quad (15)$$

The presence of the first order spatial derivative; i.e. the advection term, leads to asymmetrical components of the stiffness matrix (Smith and Griffiths, 2004).By using Green-Gauss theorem, Equation 15 can be re-written in matrix format as follows:

$$[k_T] = \left\{ \frac{\partial C}{\partial t} \right\} = [k_D + k_V + k_R]\{C\} \quad (16)$$

where  $[k_T]$ is the rate of concentration changes with time matrix,  $[k_D]$  is the stiffness matrix resulting from dispersion and diffusion,  $[k_V]$  is the stiffness matrix resulting from advection and  $[k_R]$ is the stiffness matrix resulting from sorption and chemical reaction.

$$[k_T]_{4*4} = \iint NN^T d_x d_y \quad (17)$$

$$[k_D]_{4*4} = -\iint \left[ D_x \frac{\partial N}{\partial x} \frac{\partial N^T}{\partial x} + D_y \frac{\partial N}{\partial y} \frac{\partial N^T}{\partial y} \right] d_x d_y \quad (18)$$

$$[k_V]_{4*4} = -\iint \left[ v_x N \frac{\partial N^T}{\partial x} + v_y N \frac{\partial N^T}{\partial y} \right] d_x d_y \quad (19)$$

$$[k_R]_{4*4} = -K \iint NN^T d_x d_y \quad (20)$$

The matrix  $[k_V]$  is unsymmetrical due to presence of the first order partial derivatives; while the matrix  $[k_D]$  related to the dispersion part of flux is symmetrical. To integrate the matrix equation containing the partial derivatives of the variable needs evaluating with respect to the time, there are several methods (Smith and Griffiths, 2004).

### RESULTS AND DISCUSSIONS

The soil washing remediation technique is controlled by aphysicochemical process such as desorption, complexion, dissolution, and oxidation-reduction where at the end of soil washing, the majority of the contaminant is transferred into the washing solution. However, some of



contaminants may remain in the soil and this can be represented mathematically as (Sharma, 2004).

$$C_{si}M_s = C_{sf}M_s + V_l C_l \quad (21)$$

Where

$C_{si}$  = initial contaminant concentration in the soil (mg/kg),

$M_s$  = total dry mass of the soil (kg),

$C_{sf}$  = final contaminant concentration of the soil sample (mg/kg),

$V_l$  = total volume of washing solution (L), and

$C_l$  = contaminant concentration of the washing solution (mg/L).

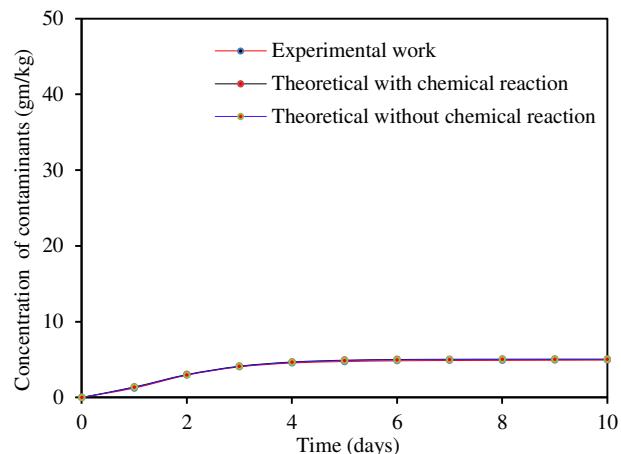
If the washing process continued for a sufficient time, an equilibrium condition will be reached and the contaminant concentration in washing solution ( $C_l$ ) and in the soil ( $C_{sf}$ ) can be related by the distribution coefficient of contaminant between the soil and the washing solution ( $K_d$ ) as  $C_{sf} = K_d C_l$ . When this equation is substituted in Equation 21, Equation 22 or 23 will be used to determine the removal efficiency of contaminant from soil.

$$\text{Removal efficiency (\%)} = \frac{1}{1+V_l(K_d M_s)} * 100 \quad (22)$$

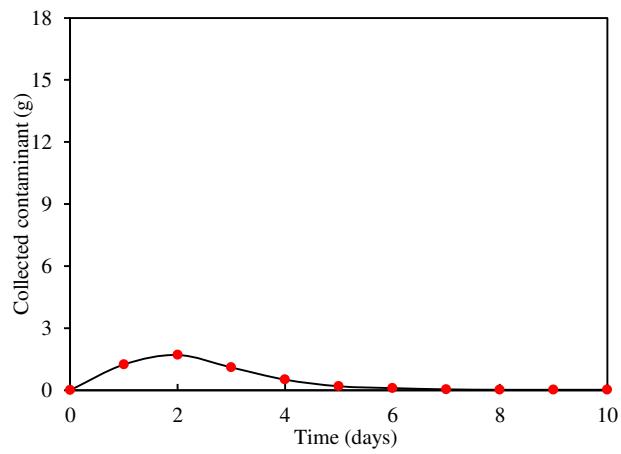
or

$$\text{Removal efficiency (\%)} = \frac{C_{si} - C_{sf}}{C_{si}} * 100 \quad (23)$$

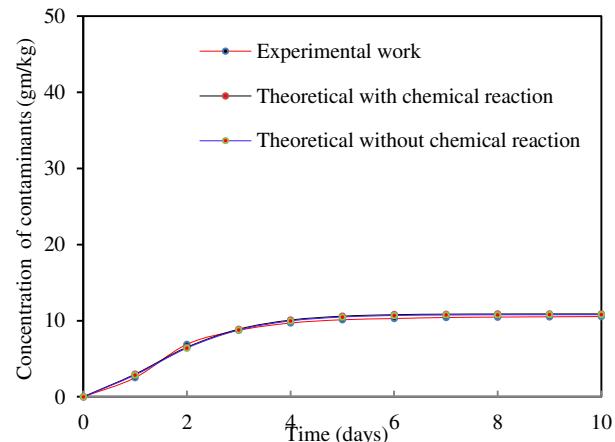
On the other hand, the contaminant removal efficiency is calculated by the quantity of the collected solute which leached from the contaminated soil samples and verified the results obtained from equations 22 or 23. The numerical solution of mass transport equation taking into consideration the effects of chemical reactions by finite element method was used. A computer program P8.8 written in FORTRAN 95 presented by Smith and Griffiths (2004) has been developed by adding the effects of chemical reactions into the partial differential equation of mass transport as sink action due to dissolving the contaminant in distilled water. The results of experimental work have been used to verify the results of theoretical modeling. The results of concentration of contaminant in the soil samples and collected contaminant versus time obtained from the modified program are compared with those obtained experimentally as shown in Figures 1 to 8. The concentration of contaminant reaches its maximum value after four hours of starting washing process in the tested soil samples.



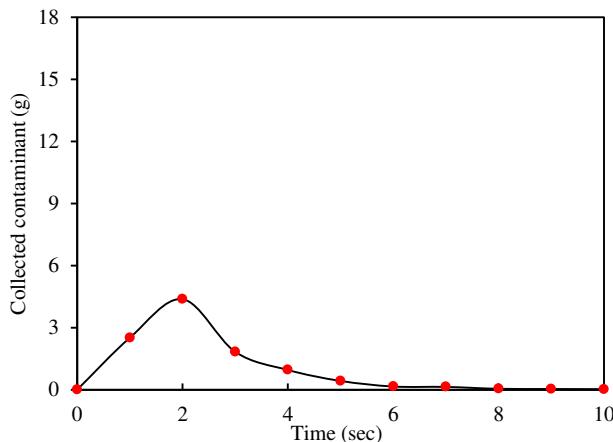
**Figure-1.** Accumulated concentration of contaminant versus time of soil sampleF1.



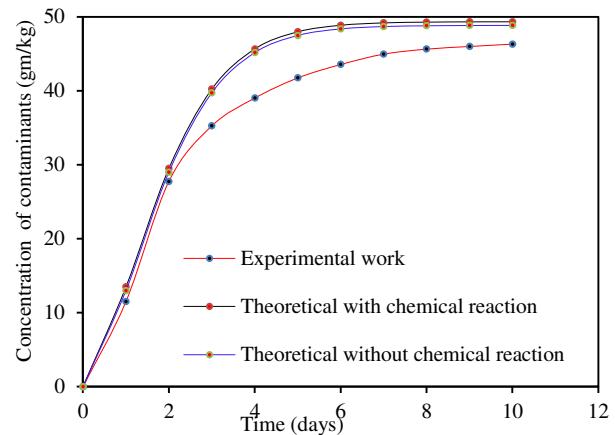
**Figure-2.** Collected contaminant versus time of soil sampleF1.



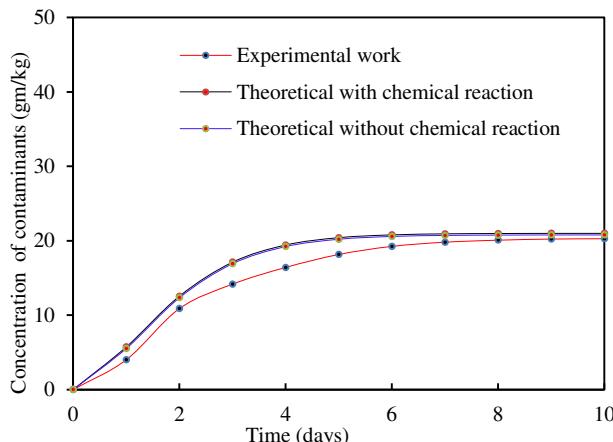
**Figure-3.** Accumulated concentrationof contaminant versus time of soil sampleF2.



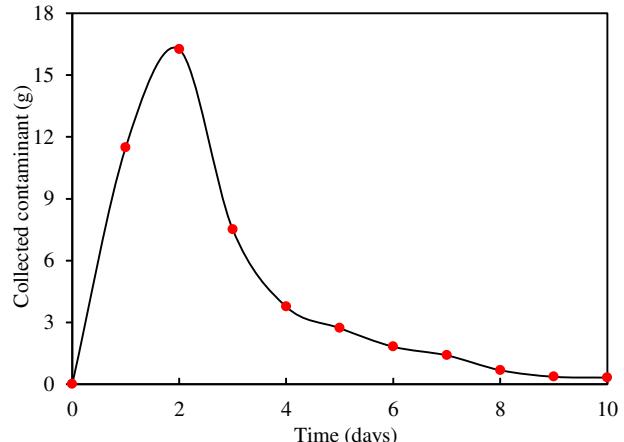
**Figure-4.** Collected contaminant versus time of soil sampleF2.



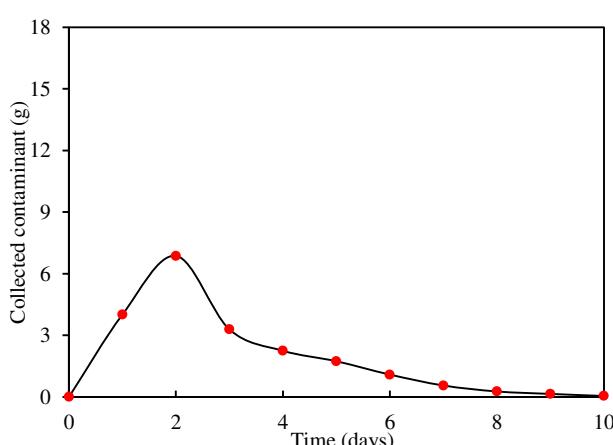
**Figure-7.** Accumulated concentrationof contaminant versus time of soil sampleF4.



**Figure-5.** Accumulated concentrationof contaminant versus time of soil sampleF3.



**Figure-8.** Collected contaminant versus time of soil sampleF4.



**Figure-6.** Collected contaminant versus time of soil sampleF3.

The results obtained from FORTRAN program agreed well with those obtained from experimental work for different percentages of contamination. The same pattern of relationship between the percentage of collected contaminant and time for both experimental and theoretical work are noticed for the tested soil samples. There is noticeable difference between experimental and theoretical work results in the first hour of washing, but after that, the theoretical curve approaches from the experimental one. The sink term added to the differential equation of mass transport has slight effects on the variation of contaminant concentration with time. The efficiency of the washing technique is determined depending on Equation 23. The removal efficiency of the contaminants from soil samples are given in Table-2. The values of removal efficiency prove the adequacy of suggested remediation technique of sandy soil contaminated with industrial wastewater like that used in the present study. The suggested technique of remediation is considered very important for sandy soil analysis because it's cheap, practical, time-saving and does not need for equipment.



**Table-2.**The removal efficiency of remediation process.

Soil sample	Removal efficiency, %
F1	97.63
F2	96.79
F3	96.58
F4	93.87

## CONCLUSIONS

According to the results of experimental work and numerical finite element analysis applied in this present work, the following conclusions can be drawn out:

- The chemical properties and electrical conductivity of sandy soil are slightly affected by different percentages of industrial wastewater used in the artificial contamination of soil samples. This action is resulted from the inert coarse particle of sandy soil used in this study.
- Reducing the hydraulic conductivity of contaminated soil samples will affect the mass transport of contaminant from the soil.
- The results of finite element method are well agreed with those obtained experimentally for the different percentages of contamination.
- Adding the chemical reactions to the mass transport equation has slight effects on the transported mass of contaminant from the soil samples.
- The proposed remediation technique "soil washing" is efficient, economical and time consuming for sandy soils. The removal efficiencies were (97.63, 96.79, 96.58, and 93.87) of soil samples F1, F2, F3, and F4 respectively.
- Increasing the concentration of contaminant requires increasing the period of remediation to get acceptable percentage of contaminant removal efficiency.

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