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Modeling of petroleum hydrocarbons by indigenous microorganisms in the region under Nitrate reduction conditions

A.A.Daya[1*],A.Agah[2]

1. Associate Professor of Department of Mining, University of Sistan and Baluchestan

2. Assistant Professor of Department of Mining, University of Sistan and Baluchestan

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ABSTRACT

A two-dimensional model was developed for the decomposition of petroleum hydrocarbons by local microorganisms under the conditions of nitrate reduction (denitrification). To apply this model, a group of anaerobic mobile microbes with low concentration in the environment was considered. Therefore, after this time, there will be no leakage from the ground surface. The model was implemented with the assumption that the sources of pollution will be removed from the surface of the earth after 36 years. Also, it is assumed that the microbial population is not absorbed into the solid phase of the aquarium environment and nitrate is the only electron acceptor in the environment. Numerical solutions were obtained using the finite volume method. The results showed that the concentration of petroleum hydrocarbons in some areas of the site is higher than the standard, which can cause health risks. In addition, the results of field studies showed that nitrogen accumulation in the soil of Ray Industrial Area and Tehran Oil Refining Company is high. This issue shows that the natural dilution processes alone are not able to completely clean the studied site from oil pollution, and for the complete cleaning of the water system, a suitable improvement strategy should be considered. To be designed according to the conditions of the site.

1.Introduction

Petroleum hydrocarbons are widely released into the environment as a result of human activities (Mustapha et al. 2018; Gupta and Sharma 2019; Gupta et al. 2019; Gupta 2020a, b; Gupta and Yadav 2020). Today, one of the environmental concerns is the pollution of aquatic environments by petroleum hydrocarbons. The movement of LNAPLs in unsaturated porous media is sufficiently complex when only two liquid phases, air and water, are present. When a third fluid phase is involved, such as an immiscible organic liquid, the flow becomes more complicated. The movement of LNAPLs in the subsurface is dominated by the mechanisms of convective and hydrodynamic processes (Yadav et al., 2012). Diffusion and dispersion mechanisms lead to the transfer or spread of the pollution plume in a porous medium and cover a large area. On the other hand, the absorption of the constituents of the solution leads to the division of the species between the solid and aqueous phases, and as a result, the pollutant mass in the solution is reduced (Menkiti et al. 2021; Lee et al. 2021). However, sorption is the cause of soil contamination in the soil phase (MacQuarrie et al. 1990). Biodegradation of organic contaminants by an indigenous subsurface microbial population is a process by which mass can be removed from an aquifer without physically extracting the contaminant. Environmental conditions such as temperature, soil moisture, nutrient supply, water level fluctuations make the distribution of LNAPLs plumes dynamic in the water and soil system. Water level or water content affects the spatial distribution of LNAPLs in an aquifer (Dobson et al., 2007). The rise of the water level causes the upward movement of the plume and the increase of the plume cover. Fluctuation of water level causes the trapping mechanisms of LNAPLs in soil pores (Dobson et al., 2007; Lee and Chrysikopoulos 1995). The complex nature of these pollutants often causes remediation efforts to fail or be less effective than intended.

Considering the importance of the protection of underground water sources, it is necessary to use low-cost and effective methods for accurate prediction of the contaminated area due to hydrocarbon spills. Models can be a powerful tool for predicting the transformation and transfer of pollutants, including petroleum hydrocarbons, in groundwater. The transport processes that can be more

*Corresponding author's email : aliakbardaya@eng.usb.ac.ir
DOR: 10.22111/jhe.2023.45886.1097

or less dominant in the different stages of the evolution of oil pollutant spills in the underground water include the processes of expansion, dispersion, dissolution from the non-aqueous phase liquid (NAPL), surface adsorption to aquifer materials, and decomposition. Biological (Abreu et al., 2009; Ng et al. 2014; Ng et al. 2015). Considering all these processes can be effective in the comprehensive evaluation of the natural dilution of hydrocarbon compounds in underground water or to design a suitable strategy for cleaning the underground water table from hydrocarbon compounds. In order to make a more realistic and comprehensive modeling of the processes involved in the pollution of the water table with petroleum hydrocarbons, sufficient data and a correct conceptual model that shows the effective mechanisms in Contaminant transport, pollution plume development and also hydrogeochemical reactions are involved (Liu et al. 2016; Teramoto and Chang 2019). In presenting a conceptual model, knowing the characteristics of the aquifer, geochemistry, determining the boundaries of the study area and the purpose of modeling is essential. Modeling the biodegradation of NAPLs in the subsurface is a research area in many environmental applications. Examples of biological degradation and natural attenuation of contamination in groundwater aquifers (Volkman et al., 1984; Christensen and Larsen, 1993; Peters and Moldowan, 1993; Blanc et al., 1996; Prommer et al. al. 1999; Prommer et al. 2002; Agah et al., 2011; Agah et al., 2012; Blagodatsky and Smith, 2012; Miller et al., 2013; Agah and Doulati Ardejani, 2015; Garg et al., 2017 ; Hidalgo et al 2020) and penetration of degradable volatile organic compounds through the vadose zone in buildings (Parker, 2003; Knight and Davis, 2013; Mustafa et al 2014; Akbariyeh et al., 2016; Membere and Johnson 2019). The goals of this study are to model the decomposition of petroleum hydrocarbons by local microorganisms under nitrate reduction conditions. To perform this modeling, a numerical finite volume model has been developed by modifying a general-purpose commercial package called PHOENICS (Spalding 1981). Adjustments related to mathematical expressions related to chemical and biological reactions were made by creating a PHOENICS input file (Q1) and providing additional code in FORTRAN in the GROUND subroutine (Doulati Ardjani et al., 2004; Doulati Ardjani et al., 2014; Aghaz and Davalit Ardjani., 2014). These FORTRAN encodings were used for all non-standard calculations by the PHOENICS solver during the solution process.

2. Materials and Methods

2.1.Geology and topography

From the point of view of geology, almost all the parts around the city of Tehran include tuff, andesite, tuffified andesite and agglomerate related to the Eocene period. The eastern part includes conglomerate of the Devonian period and carbonate rocks of the Mesozoic period. The southern part is covered with tuff and andesite from the Eocene period and is lower than the sea level. The urban area of Tehran is located on the alluvium resulting from extensive erosion of the Alborz Mountains (Figure 1).

The topographical situation in the urban area of Tehran consists of two parts: the plain and the mountainous area. The northern parts of the urban area of Tehran are located in the mountainous region and are located as a part of the Alborz Mountain range with a length of 300 km from east to west, where all kinds of folded features can be seen. This area is covered by older alluviums, but new alluviums are also partially found.

The topography around the Tehran Oil Refining Company shows that the height of the area decreases slowly from north to south and hardly slopes in the east to west direction. The topography of the area of Tehran Oil Refining Company fluctuates within less than 7 meters and

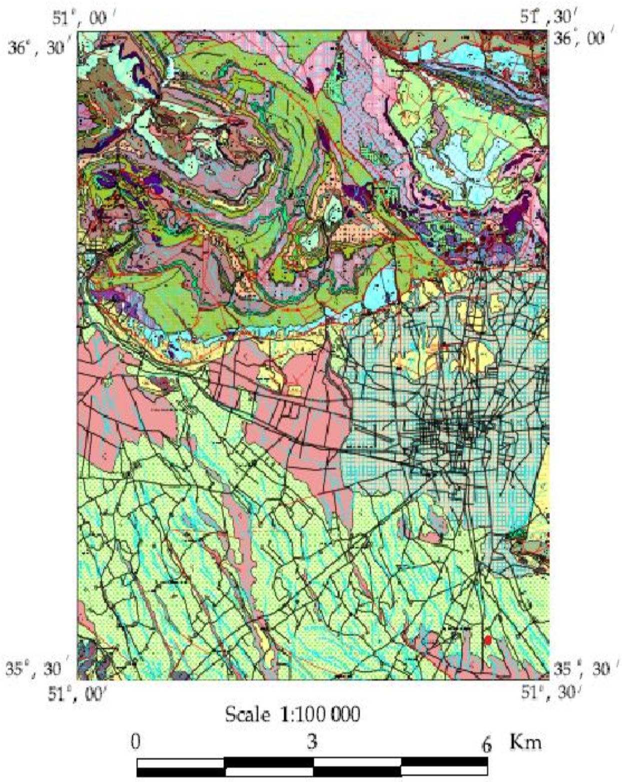

Fig. 1. Geological map of studied area

the average height is approximately 1030 meters above the surface of open seas.

There are two faults in Ray Industrial Area including Tehran Oil Refining Company. One of them is located near Azim-Abad, which extends towards Shahrah-Abad in the west. But this fault is old and filled with clay, silt and alluvial deposits. In the southern part of Ray district, south of New Castle, a fault with a length of 18.5 km stretches from east to west. This fault is also old and its activity has stopped.

2.2.The transport equation of soluble pollutants in underground water

The two-dimensional diffusion-diffusion equation with delay, irreversible adsorption, and biodegradation kinetics using the Mund model for petroleum hydrocarbons, bacteria, and nitrate in the saturated flow regime can be written as follows:

$$R\frac{\partial C_{org}}{\partial t}=\frac{\partial}{\partial x}\left(D_{ij}\frac{\partial C_{org}}{\partial x}\right)-\frac{\partial}{\partial x}\left(v\,C_{org}\right)-k_1C_{org}-\frac{\mu_{\max}}{Y}\left(B+\frac{\rho_b B_s}{\theta}\right)\left(\frac{C_{org}}{K_{org}+C_{org}}\right)\left(\frac{C_{NO_3^-}}{K_{NO_3^-}+C_{NO_3^-}}\right)$$

$$\frac{\partial B}{\partial t}=\frac{\partial}{\partial x}\left(D_{ij}\frac{\partial B}{\partial x}\right)-\frac{\partial}{\partial x}\left(v\,B\right)-K_AB+K_D\frac{\rho_b}{\theta}B_s+\mu_{\max}B\left(\frac{C_{org}}{K_{org}+C_{org}}\right)\left(\frac{C_{NO_3^-}}{K_{NO_3^-}+C_{NO_3^-}}\right)-k_{dec}B$$

$$\frac{\partial B_s}{\partial t}=K_A\frac{\theta}{\rho_b}B-K_DB_s+\mu_{\max}B_s\left(\frac{C_{org}}{K_{org}+C_{org}}\right)\left(\frac{C_{NO_3^-}}{K_{NO_3^-}+C_{NO_3^-}}\right)-k_{dec}B_s$$

$$\frac{\partial C_{NO_3^-}}{\partial t}=\frac{\partial}{\partial x}\left(D_{ij}\frac{\partial C_{NO_3^-}}{\partial x}\right)-\frac{\partial}{\partial x}\left(v\,C_{NO_3^-}\right)-\frac{\mu_{\max}f}{Y}\left(B+\frac{\rho_b B_s}{\theta}\right)\left(\frac{C_{org}}{K_{org}+C_{org}}\right)\left(\frac{C_{NO_3^-}}{K_{NO_3^-}+C_{NO_3^-}}\right) \quad (1)$$

Where in:

t stands for Time; X stands for Spatial coordinates; ν stands for Pour water velocity in the x direction; D_{ij} stands for hydrodynamic dispersion tensor; θ stands for the porosity of the table material, which is assumed to be constant; ρ_b stands for dry block density of soil; C_{org} stands for pollutant concentration in the aqueous phase; $C_{N\sigma}$ stands for aqueous nitrate concentration; B stands for concentration of suspended bacteria in the aqueous phase; B_s stands for the mass of bacteria attached to the solid phase per unit mass of the solid phase; f stands for nitrate consumption coefficient; μ_{max} stands for maximum specific growth rate of bacteria; Y stands for bacterial efficiency factor; k_{dec} stands for bacteria death rate constant; k_1 stands for pollutant degradation rate constant; R stands for pollutant delay factor; $K_{NO_3^-}$ stands for nitrate half-saturation constant; K_{org} stands for pollutant semi-saturation constant; K_A stands for Bacteria adhesion rate coefficient to the solid phase; K_D stands for coefficient of separation rate of bacteria from the solid phase.

2.3.Setting the model and input data

The results of field studies showed that nitrogen accumulation in the soil of Ray Industrial Area and Tehran Oil Refining Company is high. This issue can indicate the decomposition of petroleum hydrocarbons under denitrification conditions in the region. Therefore, a two-dimensional model was designed to simulate the decomposition of petroleum hydrocarbons under denitrification conditions in the entire Ray industrial area. Because a variety of oil pollutants have been identified in the underground water table in the mentioned area, the total amounts of hydrocarbons are considered in the modeling. The simulation was performed as a view from above.

The dimensions of the mentioned two-dimensional model are 3000 meters along the horizontal and 5000 meters along the vertical, which includes 15360 control volumes. The underground water flow system was assumed to be stable. Concentration values are presented in mg/L. The input data of the model are given in table 1.

Table 1. Input parameters used in this simulation.

Spatial and temporal discretization	**Value**	**Reference**
Length of model in X direction	3000 meters	Choi et al., 2009)
Width of model in Y direction	5000 meters	
The number of volumetric elements	15360	Choi et al., 2009)
Duration of simulation	50 years	
Number of time steps frequency	100	
Flow and transmission parameters	**value**	**Reference**
Hydraulic steering along x	8.07 $*10^{-6}$ m/s	
Hydraulic steering along y	$8.07*10^{-6}$m/s	
Dry density of soil	1.7 gr/cm^3	
porosity	0.14	
Longitudinal dispersion coefficient α_L	15	
Transverse scattering coefficient α_T	1 meters	
Groundwater flow rate	$7.16*10^{-8}$ m/s	
Microbial concentration	1.86 mg/L	(Choi et al., 2009)
Initial concentration and nitrate input	18 mg/L	
Pollutant delay factor R	2	
First order rate constant of pollutant decomposition (K1)	$1.33*10^{-3}$ min^{-1}	(Choi et al., 2009)

The following initial and boundary conditions were determined for the model:

- Zero concentration was considered as the initial condition. This value describes the distribution of the dissolved pollutant in the flow domain before modeling $C(x,0)=0$, $x\geq0$.

• The constant concentration was considered equal to the concentration of the pollutant at the inlet boundary of the model C(0,T)=0 , t>0.

• The constant concentration equal to zero was determined at the output boundary of the model $C(\infty,t) = 0$, $t\geq0$.

In this simulation, it is assumed that petroleum hydrocarbons are biologically decomposed under anaerobic nitrate reduction conditions. For example, the denitrification reaction of toluene is expressed as follows (Molson, 2000):

$$C_7H_8+6NO_3^- \longrightarrow 7CO_2+3N_2+4H_2O \qquad (2)$$

3.Results and discussion

The first scenario: two-dimensional reaction model including the processes of dispersion, diffusion, absorption and biological decomposition under the conditions of denitrification, assuming that after 36 years, the sources of pollution will be removed from the surface of the earth.

In this simulation, the model provided by Choi et al. (Equations 3-1) was used to describe the process of biological decomposition under denitrification conditions including microbial growth. To apply this model, a group of anaerobic mobile microbes with low concentration in the environment was considered. The model was implemented with the assumption that the sources of pollution will be removed from the surface of the earth after 36 years. Therefore, after this time, no leakage occurs from the ground surface, but no cleaning method is implemented for contaminated soil and underground water. Also, it is assumed that the microbial population is not absorbed into the solid phase of the aquarium environment and nitrate is the only electron acceptor in the environment. With these assumptions, the model was run for 50-time steps. The modeling results in two-time steps of 36 years and 50 years are shown in Figures 2 and 3.

Comparison of figure (2) with figure (3) indicates a good agreement between numerical modeling results and field data.From the comparison of figures (2) to (3), it can be seen that no biological decomposition reaction occurs in area (a) due to the high concentration of the pollutant (above 500). As a result, nitrate is not consumed in that area and its concentration remains equal to the input concentration. On the other hand, in the mentioned area, the microbial population lacks growth and only degrades, so its concentration decreases to zero.

In region (b), due to the decomposition of petroleum hydrocarbons under denitrification conditions, all the amount of nitrate is consumed and its concentration reaches zero. As seen in figure (3), the occurrence of denitrification reaction in the mentioned area also causes the growth of the microbial population, which is determined by the increase in concentration in that area

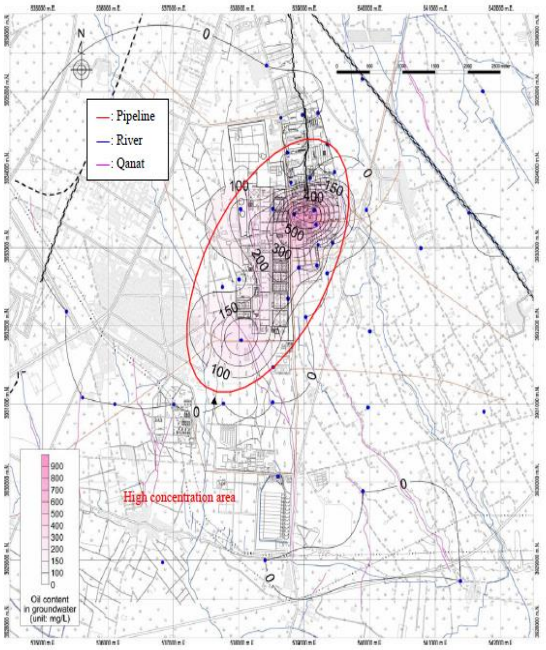

Fig 2. Distribution of oil content in underground water in the entire industrial area of Ray (Fusione Techno Solutions Co., 2006).

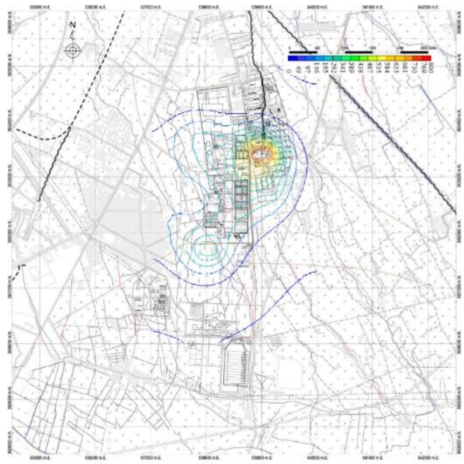

Fig 3. distribution of petroleum hydrocarbons concentration in a period of 36 years. In this scenario, the pollution sources were removed from the surface of the earth after 36 years

The concentration of petroleum hydrocarbons in area (c) is zero, therefore, no biological reaction takes place in this area that causes nitrate consumption and microbial population growth. On the other hand, the microbial population is destroyed under first-order kinetics, therefore, its concentration decreases in the area (c).

3.1.Second scenario: two-dimensional reactive model including the processes of dispersion, diffusion, absorption and biological decomposition under the conditions of denitrification with the assumption that after 36 years the sources of pollution will be removed from the surface of the earth and in addition, the contaminated soil will also be cleaned.

In this scenario, in addition to the removal of pollution sources on the surface of the earth, the soil contaminated with petroleum hydrocarbons above the groundwater level is also cleaned. Therefore, after a period of 36 years, the pollution entering the aquifer is significantly reduced by different processes of dispersion, surface absorption and biological decomposition under denitrification conditions (Figure 4).
However, despite this, the concentration of petroleum hydrocarbons in some areas of the site is higher than the standard, which can cause health risks. This issue shows that the natural dilution processes alone are not able to completely clean the studied site from oil pollution, and for the complete cleaning of the water system, a proper improvement strategy should be considered. To be designed according to the conditions of the site.
As seen in figure (5), in some areas of the site, the concentration of petroleum hydrocarbons reaches zero. This decrease in concentration and change in the shape of their plume is caused by the processes of hydrodynamic dispersion and advection.
By comparing figures (4) to (5), it is clear that the denitrification reaction does not occur in the areas shown with the name (a) due to the absence of petroleum hydrocarbons. As a result, in these areas, the concentration of nitrate remains at the limit of the input concentration and the concentration of the microbial population reaches zero due to destruction.
In region (b), petroleum hydrocarbons and nitrates are consumed in the denitrification reaction. This causes a decrease in the concentration of petroleum hydrocarbons and the complete consumption of nitrates and the growth of the microbial population in this area.
The decrease in the concentration of petroleum hydrocarbons in area (c) due to the hydrodynamic dispersion process causes a decrease in the rate of nitrate consumption and the growth rate of the microbial population due to the denitrification reaction.
Figure (5-6) shows the activity area of denitrification reaction along with its reaction rate in this scenario. As can be seen, the denitrification reaction rate has decreased slightly compared to the first scenario. This is related to the decrease in the concentration of petroleum hydrocarbons in this scenario.

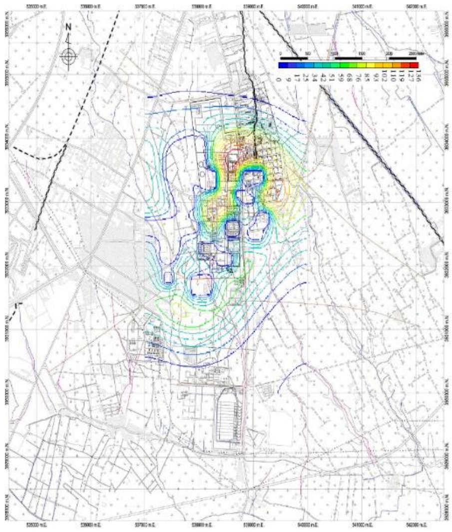

Fig 4. Distribution of petroleum hydrocarbons concentration during 50 years. In this scenario, the pollution sources were removed from the surface of the earth after 36 years and the contaminated soil was also cleaned.

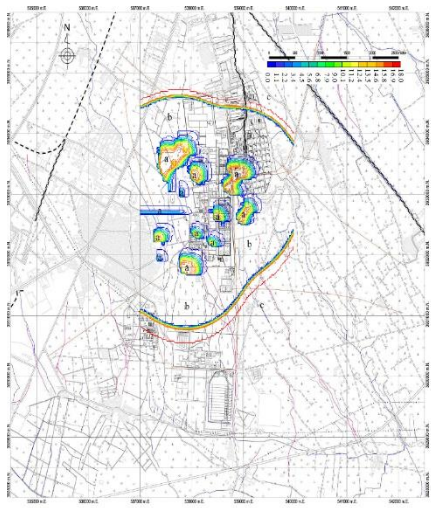

Fig 5. Nitrate concentration distribution over a period of 50 years. In this scenario, the pollution sources were removed from the surface of the earth after 36 years and the contaminated soil was also cleaned.

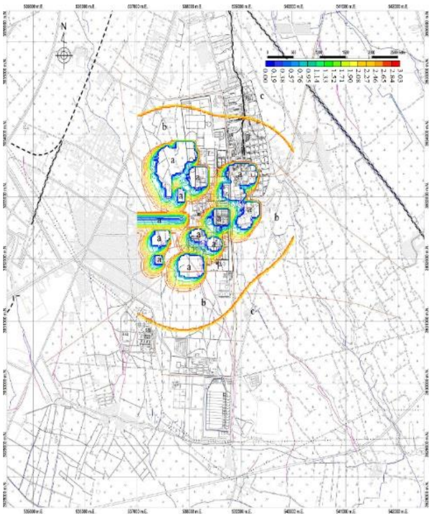

Fig 6. Distribution of microbial population concentration over a period of 50 years. In this scenario, the pollution sources were removed from the surface of the earth after 36 years and the contaminated soil was also cleaned.

4.Conclusions

The findings of field studies indicated that there is a high level of nitrogen accumulation in the soil of Ray Industrial Area and Tehran Oil Refining Company. This suggests that petroleum hydrocarbons are decomposing under denitrification conditions in the region. To further understand this process, a two-dimensional model was created to simulate the decomposition of petroleum hydrocarbons under denitrification conditions throughout the entire Ray industrial area. Due to the detection of various petroleum pollutants in the underground water table in this area, it was observed that the concentration of petroleum hydrocarbons in certain parts of the site exceeds the standard limit, posing potential health risks. This highlights the fact that natural dilution processes alone are insufficient for effectively removing oil pollution from the studied site. Therefore, it is necessary to develop an appropriate improvement strategy based on the site conditions to achieve complete cleansing of the water system.

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