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SOILS AND SUB-SOILS CONTAMINATION: MIGRATION OF PETROLEUM TO THE GROUND SURFACE ACCOUNTING FOR VARIABLE PERMEABILITY COEFFICIENT IN COMPRESSIBLE ROCKS.

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ABSTRACT

We have investigated the pollution of soils and sub-soils by the petroleum during it's upwards motility to the ground surface. Our analysis has taken into account the compressibility of the rocks and the variability of the permeability coefficient. We have demonstrated that the system is governed by a strongly non-linear equation of diffusion. This equation has been solved numerically using the DOPRI5 fourth-and-fifth-order Runge-Kutta variable step integrator. Three types of rock were involved in this survey, namely intact rock, clay rock and sandy rock. We have obtained that the type of the rock, as well as the type of the oil, plays a crucial role in the migration process. So, in the case of the sandy rock, the maximum distance attainable by the oil is approximately 1430 m. This value turns into 1850 m when we consider rather the clay rock for the same period of observation. Taking into account light oil, we have reached a distance of about 1200 m, whereas extra-heavy oil let us to only 1000 m. Seeking for the impact of the coefficient of permeability, we have considered two different values of the permeability: $k = 10^{-14} m^2$ and $k = 10^{-12} m^2$. And a real gap was recorded between the rate of oil diffusion..

Key words: Petroleum diffusion; soils contamination; compressibility coefficient of rocks; porous media; DOPRI5 – RK method

1 - INTRODUCTION

The problem of the pollution by the petroleum has retained the attention of a number of scientists over the past decades. Studies focused mainly on the petroleum contaminated soils and sub-soils. In general, they were interested in situations where petroleum s are spilled on some sites on the ground surface. The reader can refer to the following contributions (Kang et al 1996, Labieniec et al 1996, Bai et al 1996, Simo et al 2019a; Simo et al 2019b). It is a fact that petroleum are thrown, here and there, in the four corner of the world. Such phenomena could be accidentally or naturally produced. However, they could also result from harmful actions of some people ill-intentioned. Researches related to this topic of soil contaminants include the following papers (Haapkylal 2007, Das et al 2011, Verbruggen et al. 2011, Wolfson 2012, Aerospace 2005).

It should be recalled that, oil spills is not the only form of pollution by petroleum. Soils and sub-soils can also be contaminated during the tertiary migration of petroleum to the ground surface. The formation of petroleum comprises two main steps, namely the diagenesis and the catagenesis. The process of diagenesis leads to the formation of kerogen and bitumen. Thereafter, the kerogen undergoes further change and gives rise to hydrocarbons. The catagenesis is the step during which hydrocarbon chains are formed after the thermal degradation of kerogen. In this process of the formation of the petroleum, we are interested at what happen after the catagenesis: The upward movement toward the ground surface. In the present work, we address the problem of hydrocarbon mass movement from the oil reservoir to the surface of the earth. This process is known as the tertiary migration. It is a migration to the surface, either from a reservoir or source rock.

Hydrocarbons released by the source rock during the primary migration continue their race toward the reservoir rock. This process is known as the secondary migration (Walters, 2017). If during their motion they don't encounter any obstacle (impermeable rock), they pursue their progressive motion toward the surface of the earth. This paper attempts to dig inside the process of migration of hydrocarbons toward the ground surface.

Furthermore, let us emphasize that so far as the oil confined in the reservoir rock is concerned, a large portion of hydrocarbon trapped is not perfectly sealed. Subsequently, the stored volume leaks to the surface over time. Surface manifestation of oil and gas can be divided into two broad categories namely: the macroseepage and the microseepage (Etiope, 2015; Richers et al., 1982; Asadzadeh and de Souza Filho, 2017). This issue is important because natural oil seep is a worldwide phenomenon that contributes to the pollution of the environment by petroleum than all other sources combined (Stout and Wang, 2016). In this sense, it should always be considered when investigating the origin or the forms of contamination. As a consequence of various seeps, dangerous petroleum compounds contaminate sub-soils and eventually soils., leading to major detrimental effects on the environment. In the Gulf of Mexico, there are more than 600 natural oil seeps that leak between one and five million barrels of oil per year, equivalent to roughly 80 000 to 200 000 tonnes (Board,2003). Petroleum seeps are quite common in many areas of the world. An example of remigration of oil related to uplift events is the case of the Barents Sea region, resulting in dry wells with paleo-oil saturation (Henriksen et al., 2011; Nyland et al., 1992; Ohm et al., 2008).

Because of these unpredictable process, physical and chemical characteristic of soils and subsoils are affected in some parts of the world. And the process of contamination is triggered. There is contamination because these petroleum components find themselves in a place where they should not be or at concentrations above background (Chapman, 2007).

This problem of contamination of sub-soils and soils by the petroleum released by the source rock or by the reservoir rock is the main objective of this contribution. The petroleum that migrates from source or reservoir rock toward the ground surface includes light oil, heavy oil and extra-heavy oil. It should be noted that, contaminated soils have negative impacts on the environment and human health. Petroleum is composed of compounds called petroleum hydrocarbons. It is worth noting that, many petroleum hydrocarbons are highly mobile. Thus, if they are in soil they can be readily transported to farms. It is detrimental and very dangerous to agricultural activities. Many vegetables can absorb contaminants as they grow. An other harmful effect of soil pollution may comes from direct contact with polluted soil or from contact with other resources, such as water, that have come in direct contact with the polluted soil (WHO, 2010). It is important that people don't be exposed to contaminated soils.

The paper is organized as follows: We present the model in the next section. Section three stands for the implementation of the DOPRI5 fourth-and-fifth-order Runge-Kutta variable step integrator for the numerical investigations. In this section, computational results are followed by discussions and commentaries. Section four is devoted to the conclusion of the paper.

2. THE MODEL

As we mentioned earlier, this work is based on sites potentially contaminated by oil leaks from the reservoir rock or coming directly from the source rock. It is very important to emphasize that these petroleum contaminants shall not remain indefinitely on the subsoil. They shall inevitably migrate in the soil following the well-known diffusive model of compounds in porous media. Three types of rock shall be considered, according to their coefficient of compressibility: rock intact, clay rock and sandy rock. Several researches related to this topic of porous and fractured media flow have been carried out. The reader can refer to the following papers (Darcy 1856, Forchheimer 1901, Carmen 1937, Fand et al. 1987, Kececioglu and Jiang 1994, Barree and conway 2004, Liu et al. 2009, Lai et al. 2012).

In order to derive the equation of diffusion of petroleum into the ground, we combine Darcy's law and the conservation equation. In its most general, three-dimensional form, the equation of conservation of mass can be written as

$$\frac{\partial \phi \rho}{\partial t} = -\nabla.(\rho \vec{v}) \qquad (1)$$

where ϕ and ρ are porosity and oil density, respectively. $\vec{v}$ is the fluid velocity which is given by the Darcy's law. Generalized Darcy's law for liquid transport in unsaturated soil has been proposed as (Richards, 1931; Bear, 1979):

$$\vec{v} = \frac{-k}{\mu} \nabla p \tag{2}$$

Here, $\vec{v}$ is the velocity, κ is the permeability, p is the pressure and μ is the viscosity. The compressibility of the rock, C_R and the compressibility of the fluid, C_f are defined, respectively, by the following expressions

$$C_R = \frac{1}{\phi}\frac{\partial \phi}{\partial p}, \qquad C_f = \frac{1}{\rho}\frac{\partial \rho}{\partial p} \tag{4}$$

The integration os Eqs. (4) within the boundary limit of $\rho = \rho_0$ and $\phi = \phi_0$ at $p = p_0$, yields

$$\rho = \rho_0 e^{[C_f(p-p_0)]}; \qquad \phi = \phi_0 e^{[C_R(p-p_0)]} \tag{5}$$

Where ρ_0 and ϕ_0 are the initials oil density and rock porosity, respectively. p_0 is the initial pressure. We assume that the change in pressure is not very significant. In such context, the above approximations can be made

$$\rho \cong \rho_0\left[1 + C_f(p - p_0)\right] \qquad \phi \cong \phi_0\left[1 + C_f(p - p_0)\right] \tag{6}$$

Substituting equations (2), (4) and (6) into equation (1) yields the master equation of the system

$$\phi_0 \frac{\partial \rho}{\partial t} = \frac{1}{\mu \rho_0 C} \nabla . (k \rho \nabla \rho) , \tag{7}$$

where

$$C = C_R + C_f \tag{8}$$

represents the total compressibility coefficient.

In this paper, we restrict ourselves on one-dimensional diffusion equation of oil. In one dimensional approximation, this equation of diffusion of the pollutant (7) is reduced to :

$$\frac{\partial \rho}{\partial t} = \frac{1}{\mu \rho_0 \phi_0 C} \frac{\partial}{\partial z}\left(k \rho \frac{\partial \rho}{\partial z}\right) . \tag{9}$$

Here, z measures the depth of the petroleum in the soil.

In what follows, we assume that the coefficient of permeability does no remains constant during the process. In this paper, we focus our attention on the case where this coefficient is expressed as a function of density. Following Greer (Greer, 2016), we assume that the permeability coefficient is a linear function of density. The reader can also refer to the paper (Simo, 2019a and Simo, 2019b).

$$k = a\rho + b \tag{10}$$

The constants a and b being approximated by the values :

$$a = 3{,}5.10^{-8}; b = 5{,}1.10^{-7} \quad (11)$$

This expression, (10), results from a great agreement between a numerical method (Greer, 2016) and an experimental method (Wang, 1951).

In this context, the master equation of the system, (9), is transformed into:

$$\frac{\partial \rho}{\partial t} = \frac{1}{2\mu\phi\rho_0 C}\frac{\partial}{\partial z}\left[(a\rho + b)\frac{\partial}{\partial z}(\rho^2)\right] \quad (12)$$

As one can see, this equation is highly non-linear. An analytical approach is questionable. So, we opted for a numerical resolution of the latter.

3 NUMERICAL COMPUTATIONS OF THE PETROLEUM DIFFUSION EQUATION BY THE METHOD OF RUNGE-KUTTA OF THE FOURTH-AND-FIFTH-ORDERS USING THE DOPRI5 CODE AS THE INTEGRATOR.

3.1 Technique of computation

The nonlinear equation of diffusion (9) describing the migration of the petroleum in the soil does not have an obvious analytical solution. In this context, we opted for a numerical computation. So, the master equation governing the system shall be solved computationally, making use of numerical analysis methods. Let's remain that only a quite stable numerical analysis could give rise to physically acceptable solutions. In this vein, numerical errors such as those errors generated by round off should not be amplified. The approximate solution should also remain bounded.

Investigations connected to linear problems with constant coefficients can be easily approach making good use of all the mathematical tools necessary for such study of stability. One can refer, for instance, to the Von Neumann method (Charner et al 1950, Isaacson et al 1994, Crank et al 1947, Süli et al 2003). The situation is quite different in the case of nonlinear problems. Such topics are more difficult to analyze and may require a stronger form of stability. So, it is crucial to use appropriate technique to solve the problem. It is what currently justifies the choice of the Runge Kutta method using DOPRI5 code (Smith 1985, *Journal of Computational 1986*, Hairer et al 1987, Atkinson 1988, Schatzman 2002, Simo et al 2017, Simo et al 2019a, Simo et al 2019b) as the integrator for accurately describing the nonlinear diffusion of the petroleum in porous materials such as soils and sub-soils. In the frame of this technique, the spatial part of the operator is discretized, whereas the temporal part is keeping as such. This solver enables us to control the local error by varying the time step. Thanks to this approach, we highlight a significant increase in the precision of the approximation in time and space independently and easily.

As we mentioned earlier, the petroleum involved in the ascending movement could be provided by the reservoir rock or directly by the source rock. We assume that these rocks are supplied continuously and naturally according to the migration process. So, we suppose that oil concentration takes a constant value ρ_0, at these origin points. The main principle states that: At an initial time t = 0, an amount of petroleum is released from the origin point. Immediately, it starts its migration process towards the ground surface. The present study aims to establish the distribution of oil in the ground in the course of the time. Typical values of the coefficient of compressibility are shown in the following table:

Rock type	**C (1/ Pa)**
Clay rock	10^{-6} - 10^{-8}
Sandy rock	10^{-7} - 10^{-9}
Intact rock	10^{-9} - 10^{-11}

Table 1: Input data related to different types of rocks under consideration.

3.2 Results and discussions

Our numerical investigations focus on three different types of petroleum. Namely, light oil, heavy oil and extra-heavy oil. We consider three types of rock according to their coefficient of compressibility as we mentioned earlier. At the beginning of the process, all the pores of the soil are supposed to be empty; so far as the petroleum is concerned.

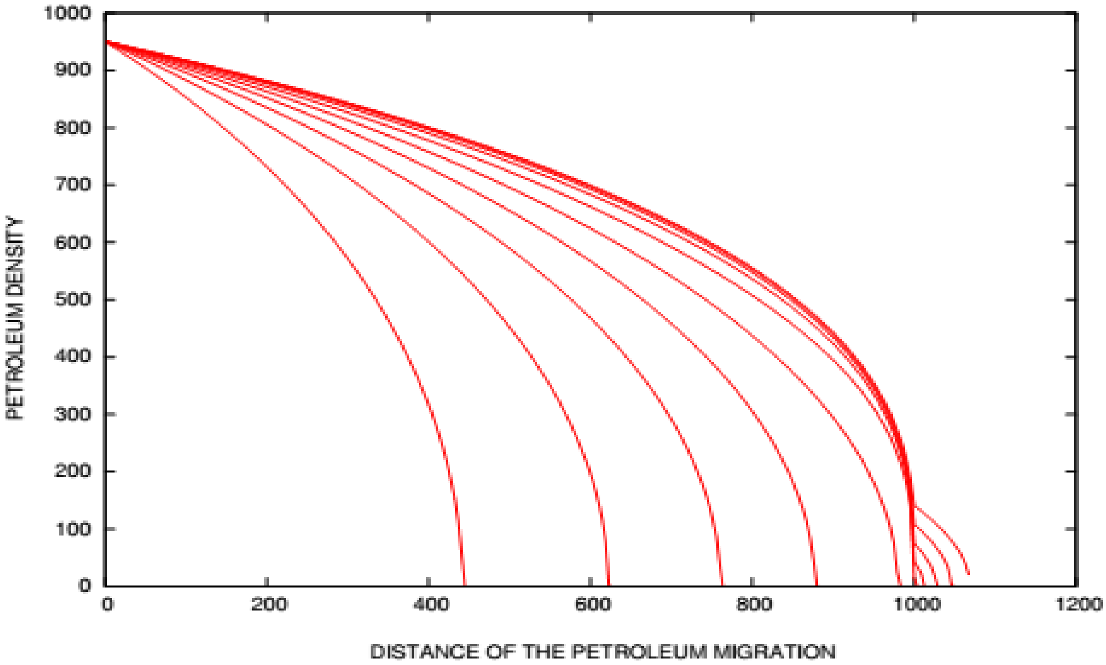

Figure 1: Oil content in intact rock in term of *the distance from a point considered as the origin of the flow of* a heavy oil of density 20 ° API. *The* compressibility is chosen to be $C = 10^{-10} Pa^{-1}$. The distances are expressed in meters (m) and the petroleum density in Kg/m^3.

At the beginning of the process, the petroleum penetrates slowly and progressively in the soil. The petroleum does not cross the soil thickness under consideration instantaneously from one point to the other. Petroleum takes some time to cover the all thickness. The rate of penetration can be observed in Fig. (1). This figure clearly indicates the positions reached by the petroleum in the soil at the given periods. So, the contamination of the entire portion of the ground necessitates some time. The petroleum moves gradually and slowly through the porous media involved in this study. For instance, for a five months period of observation, the petroleum has affected the soil only on a distance of 425 m. The density decreasing from its initial value, 934 Kg/m^3, to the zero value within 5 months. On the other hand, when we consider rather a period of 30 months, we realize that during this period, the petroleum has covered a distance of 1000 m. Figs. (1) and (2) shows that as the petroleum moves through the ground, its amplitude gradually decreases from its initial value at the origin to the zero value recorded at the highest position reached by the pollutant. It should be noticed that in Fig. 1, curves are drawn at different time periods of observation. The distribution of the oil in the rock keeps the same profile for different observation periods.

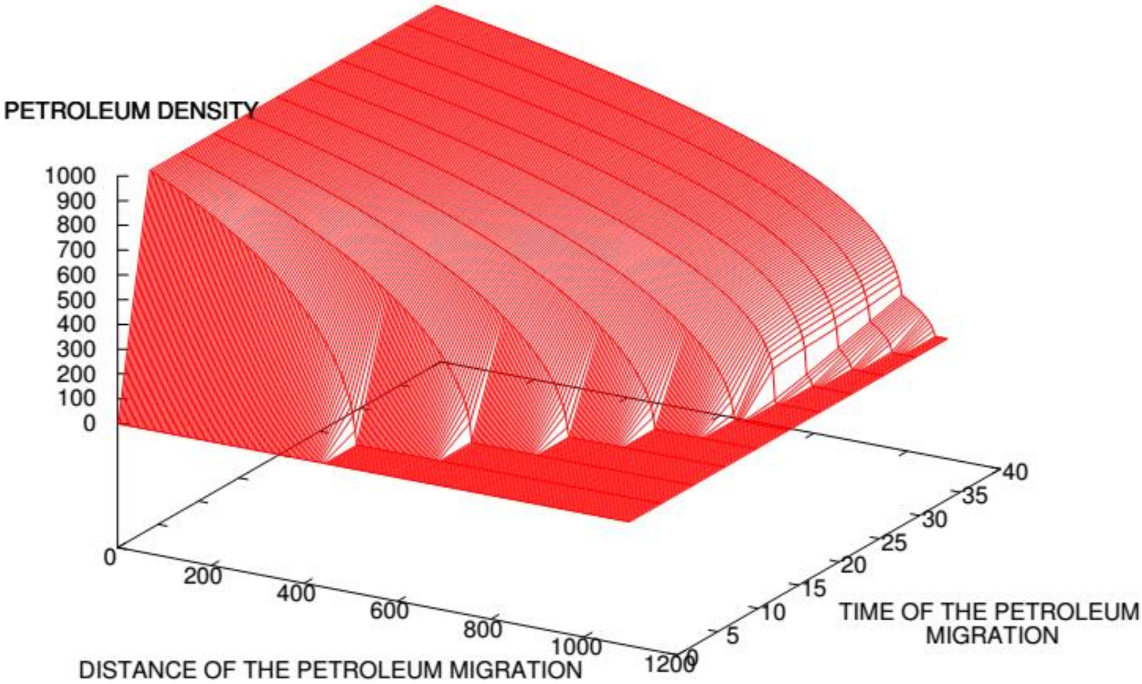

Figure 2: *Time evolution of the* **oil content in** intact rock. Calculations are performed in the same conditions as in Figure (1). Here the time is expressed in months.

Now, let's seek for the impact of the compressibility coefficient on oil migration towards the surface. The compressibility coefficient being directly linked to the type of the rock. To achieve these aims, we perform our computations with the three different types of rocks involved in this survey. This work is based on a heavy oil of density 20 ° API. We considered the same initial density of the petroleum during the computation process; i.e. $\rho_0 = 934\, Kg/m^3$. We obtain the following results: In the case of the sandy rock, the maximum distance reachable by the oil is approximately 1430 m. In the case of the clay rock, the maximum distance reaches by the pollutant is 1850 m. All these results are visible in Figs. 3, 4, 5 and 6. The general trend of these curves tells us that higher values of the coefficients of compressibility are associated with higher distances reachable by the pollutant during its upwards motility. As a bit of explanation, we can emphasize that: Within compact rocks with large cracks, the circulations of hydrocarbons are faster and disordered. So, when the compressibility coefficient of the rock increases, the oil diffusion rate also increases. The values of the compressibility coefficients of rocks are given in Tab.1.

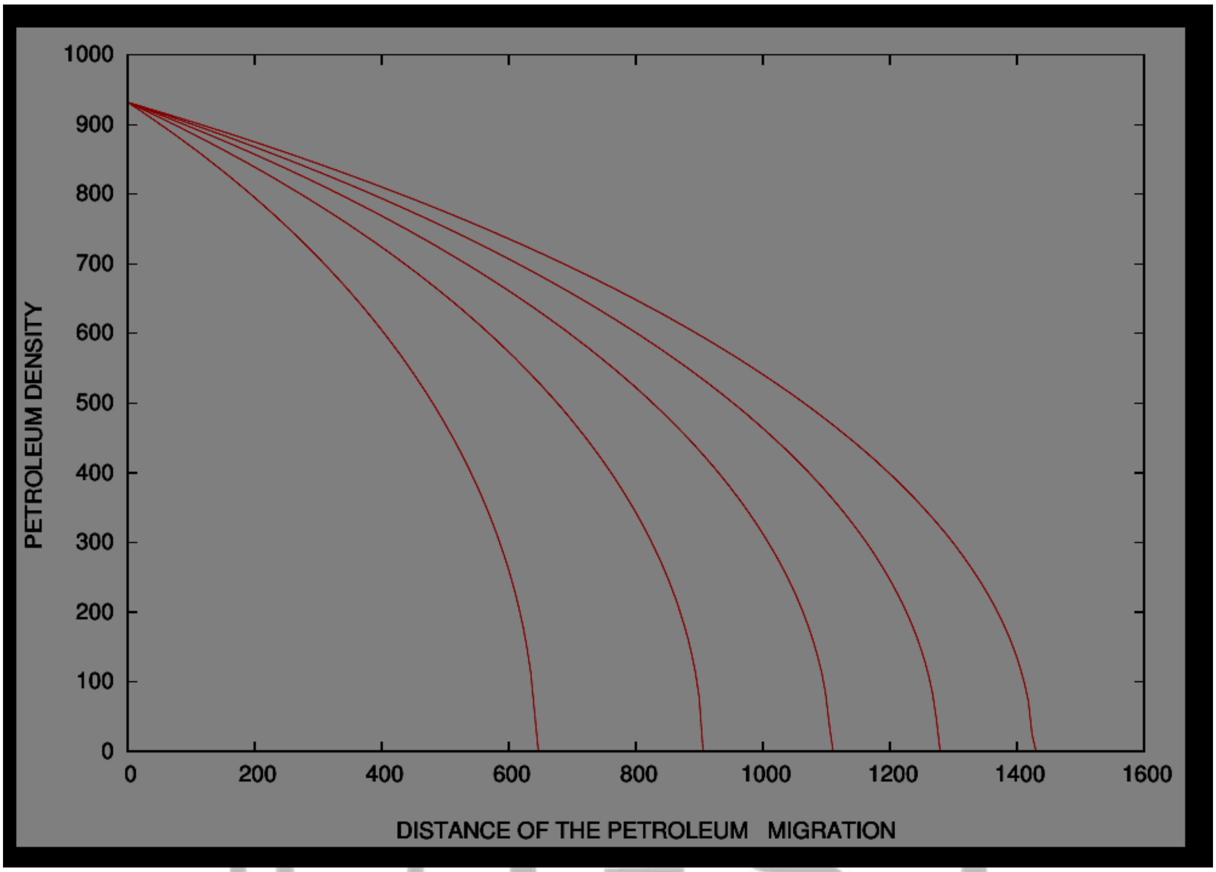

Figure 3: Oil content in sandy rock in term of *the distance from a point considered as the origin of the flow of* a heavy oil of density 20 ° API. *The* compressibility is chosen to be $C = 10^{-8} Pa^{-1}$. The distances are expressed in meters (m) and the petroleum density in Kg/m^3.

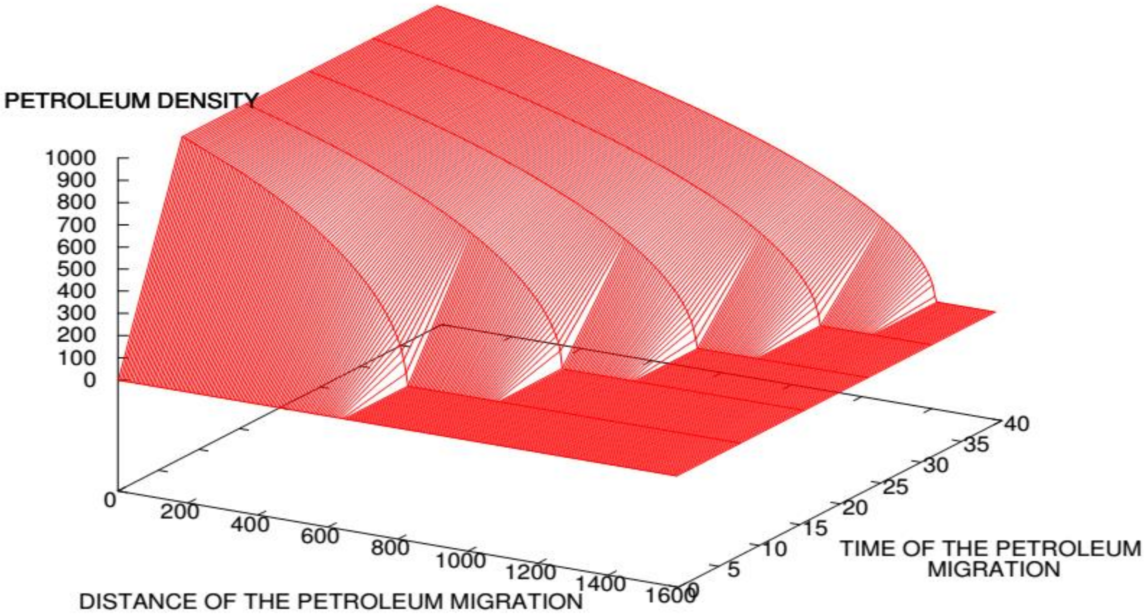

Figure 4: *Time evolution of the* oil content in sandy rock. Calculations are performed in the same conditions as in Figure (3). Here the time is expressed in months.

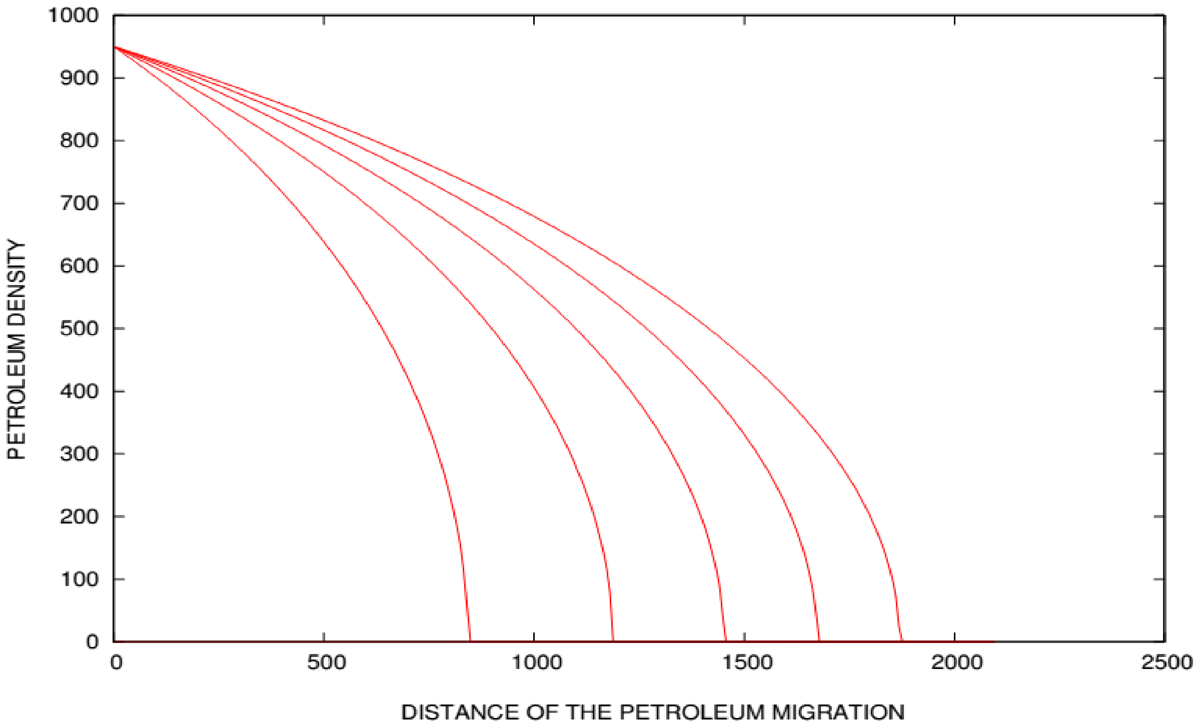

Figure 5: Oil content in clay rock in term of *the distance from a point considered as the origin of the flow of* a heavy oil of density 20 ° API. *The* compressibility is chosen to be $C = 10^{-6}Pa^{-1}$. The distances are expressed in meters (m) and the petroleum density in Kg/m^3.

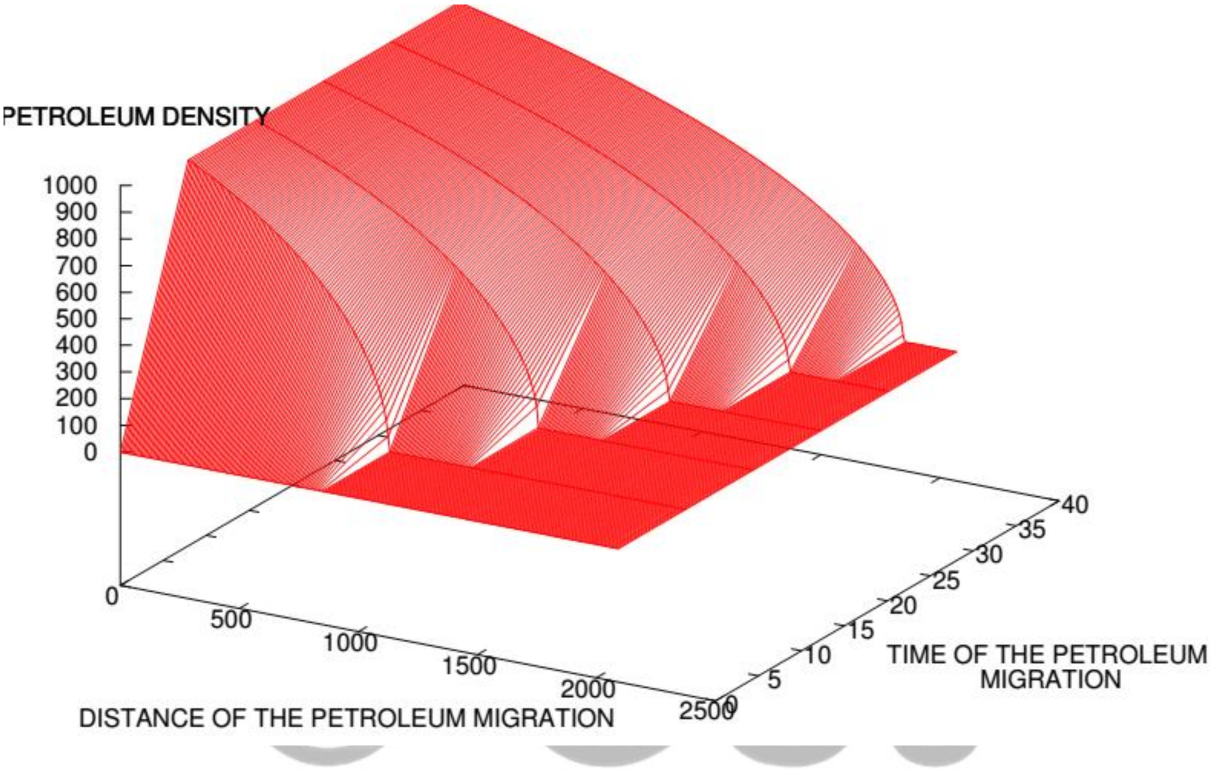

Figure 6: *Time evolution of the* oil content in clay rock. Calculations are performed in the same conditions as in Figure (5). Here the time is expressed in months.

We want to examine the impact of the density of the oil on the distance covered during its upwards motion. Calculations are performed for three different values of density, i.e. d = 25 °, d = 20 ° and d = 9 °, respectively. We obtain that, oil with small density moves faster than oil with higher density. As we can see in figure (7), as denser the oil, more it is difficult to migrate to the surface. For the same period of observation, the maximum heights reached by the pollutant are different. So, extra-heavy oil reaches a distance of about 1000 m, whereas light oil reaches a distance of approximately 1200 m.

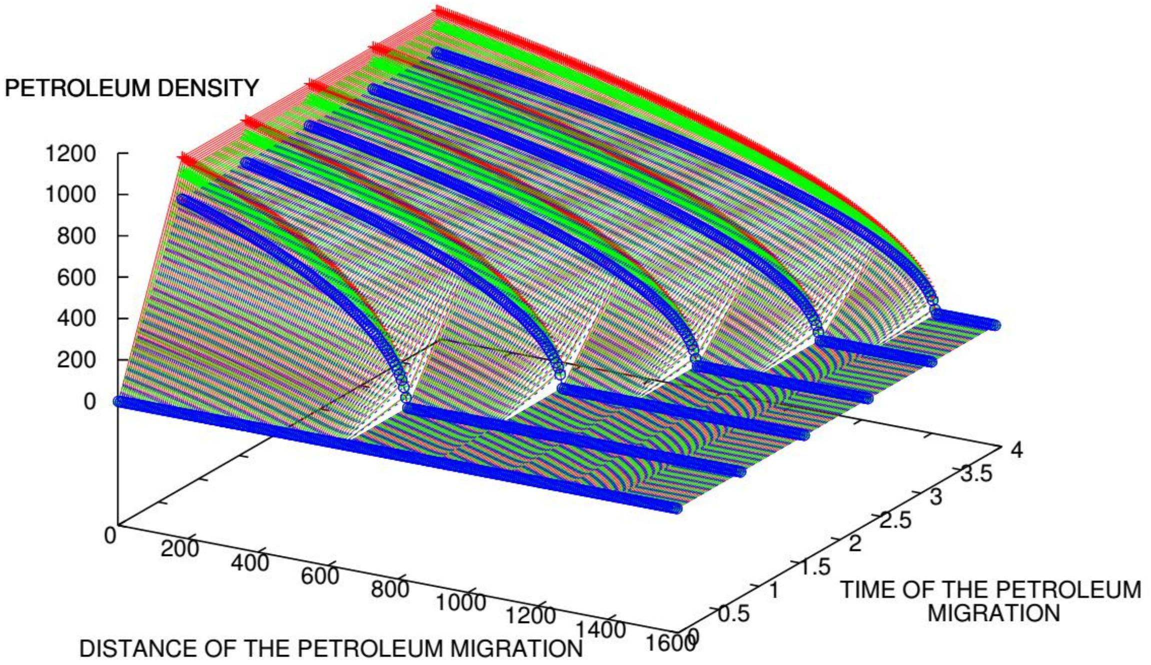

Figure 7: *Time evolution of the* oil content in clay rock with a compressibility coefficient $C = 10^{-6}Pa^{-1}$. Calculations are performed for three types of oil: light oil, d = 25 ° (curve in blue), heavy oil, d = 20 ° (curve in green) and extra-heavy oil, d = 9 ° (curve in red). Here the time is expressed in months and the petroleum density in Kg/m^3.

Now, we focus our attention on the impact of the permeability on oil migration. Fig(8) clearly indicates that the rate of penetration of oil in the rock is affected by the permeability. Here, me consider two different values of the permeability: $k = 10^{-14}m^2$ and $k = 10^{-12}m^2$. This figure exhibits a gap between the curves representing the two cases. The green curve corresponding to high permeability coefficient grows faster than the one associated to small permeability coefficient (green curve). This result was awaited. In fact, the permeability coefficient corresponds to the speed at which oil circulates within the rock.

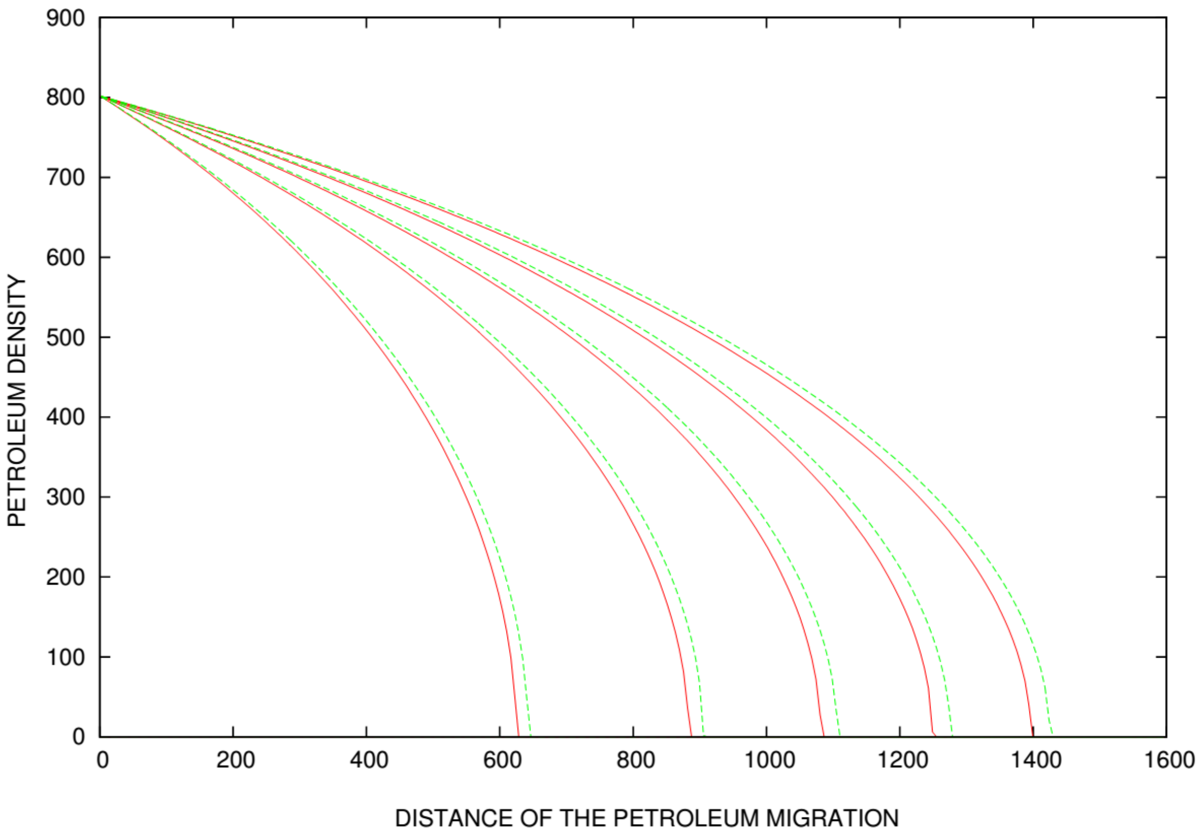

Figure 8: Oil content in clay rock in term of *the distance from a point considered as the origin of the flow of* a heavy oil of density 20 ° API. *The* compressibility is chosen to be $C = 10^{-6} Pa^{-1}$. Computations are performed for two different values of the permeability coefficient : $k = 10^{-14} m^2$ (red curve) and $k = 10^{-12} m^2$ (green curve). The distances are expressed in meters (m) and the petroleum density in Kg/m^3.

CONCLUSION

This work deals with the problem of pollution of sub-soils and soils by petroleum involved in the tertiary migration. In this contribution, we are interested by what happen after the catagenesis: The step during which the cracking of kerogen leads to the formation of petroleum and natural gas. As soon as it is formed, the petroleum triggers his upwards motility to the surface of the earth. In this context, sub-soils and soils could be contaminated by petroleum arising either from the source rock or from the reservoir rock. We have investigated the diffusion process of these petroleum contaminants. We have demonstrated that the system is governed by a strongly non-linear equation.

We have realized that the main equation of diffusion derived here could not be solved analytically. Therefore, to gain some understanding of the system behavior, we opted for a numerical approach. The technique consisted to introduce the fourth-and-fifth-order Runge-Kutta method using DOPRI5 code as the integrator. The discretization of space followed the finite volume technique. Computations have been done taking into account the nature of the oil as well as the type of the rock. In this vein, three types of rock were considered, namely intact rock, clay rock and sandy rock. Calculations also implicated three types of oil: light oil, heavy oil and extra-heavy oil. We assumed that the permeability coefficient is not constant and we introduced a density-dependent function. Greer analysis was used to approximate the linear density dependence of the permeability. Our work is described as follows: As hydrocarbons pursue their migration to the surface, we evaluate their densities at different depths below the surface.

For the different types of oil under consideration, we have plotted a number of curves to show the distribution of contaminants in the ground and their degree of penetration. In various conditions, the maximum height of hydrocarbon migration through the sub-soil or soil has been computed.

We have demonstrated that, oil migrates slowly and progressively to the ground surface. The petroleum evolves slowly and progressively toward the surface of the earth. It takes a considerable time period to cover a given distance. The distribution of the petroleum in the rock keeps the same profile for various configuration analyzed in this paper. We have obtained that as the petroleum penetrates into the rock, during its upward movement, its density decreases gradually. We have demonstrated that, an increase of the compressibility coefficient of the rock is followed by an increase of the rate of the oil diffusion. So, the type of soil also plays a role in the pollutant distribution. For example, contaminants may reach ground surface more easily in clay rock than in intact rock. Calculations have shown that light oil moves faster than heavy oil. The permeability coefficient has a real impact in the migration process: The distance covered by the pollutant increases with the permeability coefficient.

Throughout this work, we were mainly concerned with the problem of sub-soils and soils contaminated by the petroleum. From our point of view, such a problem deserves to be addressed because contaminated soils, obviously, have a number of harmful effects on human health. When soil is contaminated with petroleum, they can hurt the native environment. Live is very risky in contaminated soils. Here are some edifying examples: Indeed, the practice of agriculture should be questionable in such sites. Petroleum are just as toxic to plants as they are to humans. Many vegetables can absorb contaminants as they grow. So, If you grow food in contaminated soil, there is a risk that your food will also be contaminated. Otherwise, breathing in contaminated soil dust may cause physical or chemical damage to humans. Humans can be exposed to petroleum hydrocarbons by inhaling the fumes from contaminated soil. An other point that can be emphasized is that: water supply in such environment can be exposed to pollution because the petroleum can trickle through the soil and reach it easily. Let us list just these dangerous impacts among others.

Thus, care should be taken, as far as this matter of soils contamination is concerned. All agricultural activities must be preceded by prior analysis of the soil, for instance. Besides, if you want to build a house somewhere and construct your life there, cautions must be taken to make sure that the environment is safe. Verifications should be made to confirm that soil is not contaminated by the petroleum. Please, offer to your children an area where they can play safely without inhale dangerous products.

Finally, we believe strongly that people should be aware of the existence and the manifestation of this other form of sub-soils and soils contamination by the petroleum.

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Nomenclature

ρ density of the petroleum [Kg/m^3]

ρ_0 initial density of the petroleum [Kg/m^3]

C total compressibility [$1/p_a$]

k rock permeability [m^2]

p_0 initial pressure of the rock [p_a]

p rock pressure [p_a]

μ fluid viscosity [$p_a.s$]

c_f fluid compressibily [$1/p_a$]

$c_\emptyset$ rock compressibility [$1/p_a$]

$\emptyset$ porosity of the rock [-]

$\emptyset_0$ initial porosity of the rock [-]

v flux [m/s]

$\acute{t}$ normalized time parameter [-]

t_0 units of the measurement of time [-]

z vertical coordinate [m]

z_0 depth of reservoir rock [m]

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