



Influence of particle size and organic carbon content on distribution and fate of aliphatic and aromatic hydrocarbon fractions in chalks



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HIGHLIGHTS

- We examine the influence of chalk pore sizes on the fate of hydrocarbon fractions.
- We determine distribution coefficient (K_d) for each hydrocarbon fraction.
- We simulate the fate and transport of aliphatic and aromatic hydrocarbons in chalk.
- Monte Carlo and sensitivity analyses identified key parameters and uncertainties.

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ABSTRACT

Although great efforts had been devoted to investigate the fate and transport of various hydrocarbon sources in major aquifers, there is still a need to better understand and predict their behaviour for robust risk assessment. In this study, the fate and distribution of the aliphatic and polycyclic aromatic hydrocarbons (PAHs) of diesel fuel in chalk aquifer was investigated using a series of leaching column tests and then modelled using the Contaminant Transport module of the Goldsim software. Specifically the influence of chalk particle size on the behaviour and fate of the hydrocarbons was investigated. Distribution coefficient (K_d) between the water and chalk solid phase according to chalk particle sizes was determined for each hydrocarbon group. The larger sizes of chalk particles have higher K_d values. After 60 d of leaching using a water flow of 45 mm d^{-1} , most of the aliphatic and aromatic hydrocarbon compounds of the diesel were retained within the top 5 cm chalk layer and none of the targeted hydrocarbons were detected in the leachate from the four particles sizes chalk. Further to this, the results showed that the chalk is capable of holding more hydrocarbons than sand and chalk can limit their migration of hydrocarbons. The numerical results and the Monte Carlo analysis showed that the migration of the alkanes and PAHs is greatly retarded by the organic carbon in chalk. It is also observed that the initial mass of the alkanes and PAHs and their respective partition coefficients are important for the decaying of the source at the surface immediately after the spill and the rate-limited

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dissolution is responsible for entrapping the hydrocarbons in the top layer of the chalk. Overall these results can help to better inform risk assessment and help decision for the remediation strategy.

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1. Introduction

Chalk is found extensively in Northwest Europe and it is the most important aquifer in the UK, which accounts for 60% of the groundwater used for drinking water in England (MacDonald and Allen, 2001). Chalk is a white limestone constituted of 99% of calcium carbonate (CaCO_3) with minor marl and flint layers and very low mineral oxide content (mainly MnO_2 , $\text{Fe}(\text{OH})_3$) with a high porosity (between 35% and 45%) and low effective permeability (Hardman, 1982). It consists of matrix blocks bounded by interconnected fractures and its hydraulic conductivity is approximately 10^{-8} m s^{-1} . The hydraulic conductivity of chalk is within the range of clays, which reflects chalk has a low ability to transmit water (Clapp and Hornberger, 1978). It is widely publicized in UK that the quality and quantity of available groundwater from the chalk aquifer is under increasing pressure due to various environmental and anthropogenic activities including among other pollution due to agriculture, diesel spill from retailing fuel station, increasing population and water consumption, increasing land development (Boyle and Boyle, 2013; Ashley et al., 1994; Michel et al., 2009; Wealhall et al., 2001).

Between 2003 and 2009, it was reported that approximately 100,000 L diesel was spilled into chalk aquifer in Southern England. However limited information is available on the fate and transport of the hydrocarbon components of diesel in chalk (Boyle and Boyle, 2013). A study carried out by Ashley et al. (1994), showed that the hydrocarbon components of diesel 10 years after a spill occurred remained in the top layer of the chalk subsoil (Ashley et al., 1994). Indeed the low hydraulic conductivity of the chalk acts a barrier for the contaminants to migrate into the groundwater.

In the meantime, chemical and mechanical weathering contributes to the formation of carbonic flows which react with the calcite of the chalk and the formation of fractures and fissures (Mathias et al., 2006). While the fractures represent only 1% of the blocks, they have a higher hydraulic conductivity and can contribute to solutes and chemicals to migrate and being transported more quickly—so there is a risk that pollutants such as hydrocarbons can reach the groundwater (Mathias et al., 2006). Therefore, the chalk is not 100% impermeable for all the contaminants. Additionally, whether the main components of diesel, comprising aliphatic and aromatic hydrocarbons can leach into the groundwater still needs to be studied.

Many authors have taken an interest in the transport of diffuse pollution (mainly pesticides from agriculture and MTBE for diesel or gasoline) and solute transport across the unsaturated zone of chalk (Wealhall et al., 2001; Shah et al., 2009; Lapworth and Goody, 2006; Witthüser et al., 2003). They often used lysimeter approach to recreate in the laboratory, with some simplification, phenomena likely to be encountered in the field and leaching tests to study the fate and transport of contaminants in solid matrices (Van den Daele et al., 2007; Grathwohl and Susset, 2009). The finding of several studies is in agreement with a slow transport velocity and therefore the unsaturated zone of the chalk will significantly delay the contaminants before reaching the groundwater. However, if there is presence of fractures in the unsaturated zone, the water and solutes from the surface can infiltrate through the chalk and reach the groundwater very quickly, and so there is a potential risk of rapid transport of contaminants (Barhoum et al., 2014).

The migration of hydrocarbons in subsoil is influenced by the physicochemical properties of soil and chemicals, including: (1) soil matrix characteristics such as field capacity, porosity, bulk density, texture composition and particle size distribution; (2) chemicals properties such as solubility, vapour pressure, Henry's law constant, K_{oc} and K_{aw} . Dondelle derived partition coefficient (K_d values) for PAHs in sediment, sandy and silty soils using batch equilibrium experiment and showed that even small amounts of anthropogenic activated carbon-type organic matter in the contaminated soil samples can contribute to higher sample-specific K_d (Dondelle and Loehr, 2002). However, to the best of the author's knowledge, studies on the determination of K_d values for aliphatic and aromatic fractions in chalk are rarely conducted.

Several studies have shown that rainwater and solute transport mechanisms through the unsaturated zone are partly controlled by the soil as well as the type and thickness of the superficial layers, which act as a buffer that extends the drainage period and reduces the proportion of fracture flow (Barhoum et al., 2014; Ireson and Butler, 2011). However, the role of superficial formations and clay-with-flints in particular, is not well known.

Although great efforts had been devoted to investigate the fate and transport of petroleum hydrocarbons in the subsurface, the experimental methods are time consuming and highly dependent on the soil and hydrocarbons properties. Some environmental uncertainties, such as the spatial distribution of contaminants, are difficult to be investigated using experimental approach (Karapanagioti et al., 2003). Accordingly, environmental modelling provides a science-based structure to develop the fate and transport of hydrocarbons. It makes the hydrocarbons migration process more intuitively and helps to find out the main influencing factor(s) more easily. Over the last two decades, much more information on volatile organic compounds (VOCs), especially BTEX (Benzene, Toluene, Ethylbenzene and Xylenes) and ether oxygenates, non-aqueous phase liquids (NAPL) and PAHs in the unsaturated zones has become available from laboratory and numerical simulations (Karapanagioti et al., 2003; Rivett et al., 2011; Kamaruddin et al., 2011; Panday et al., 1997; Shah et al., 2009). Specifically, Karapanagioti et al. (2003) critically reviewed models and codes dealing with the description of contaminant

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