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PRESENCE OF HYDROCARBONS IN UNDERGROUND WATER SOURCES AND THEIR ENSUING IMPACT ON SOLUBILITY OF SOME SUBSTANCES

OBECNOŚĆ WĘGLOWODORÓW W WODACH PODZIEMNYCH I ICH WPŁYW NA ROZPUSZCZALNOŚĆ NIEKTÓRYCH SUBSTANCJI

Summary. Presence of organic substances produced as by-products of synthesis is equal to that of naturally produced substances in industrialized countries. Global consumption of mineral oils alone is over 3 mln t per year. Pollution of soil and underground water sources by substances such as detergents, mineral oils, dyes, pesticides, etc., is chronic due to the daily use of these hydrocarbons. They are a peril to soil quality, drinking water quality and certainly to human beings because they are not appropriately discarded. Presence of these substances in water provokes chain of redox reactions (eg. nitrate to nitroso, Fe (+3) to Fe (+2), etc. or *vice versa*). Under these circumstances the effect on solubility and transport of some elements and their salts in soil stratum and water sources is evident. We think that the very soil, as well as drinking water, quality is deteriorated and indirectly also the flora and fauna living on that soil, as through the food chain, traces of hydrocarbons, their metabolized and some elements of by-products will affect also human beings. The paper indicates ultimate consequences of this pollution, the methods used for analysing the contaminants in the waters, and makes suggestions for protection and recovery of hydrocarbons in the soil stratum.

Key words: hydrocarbons, pollution, underground water sources

Introduction

For the purposes of monitoring and classification activities of the state of underground water, a preventive reconstruction of hydro-geological site is necessary, that is to say:

- identification and parameterization of main aquifers,
- definition of the mode of alimentation-defluction-delivery,

- identification of the relationships between surface and underground waters,
- identification of the water points (wells, springs, etc.),
- determination of the hydro-chemical characteristics,
- identification of the characteristics of how the water is used.

The hydro-geological model must be periodically updated on the basis of new experimental information, and the monitoring activities must refer to the individual aquifers.

The environmental condition of the underground waters is defined on the basis of the quantitative and chemical state of the waters according to the criteria indicated by ANPA (Ital. *Agenzia Nazionale per Ambiente*, National Agency for Environment) and published in the Legislative Decree n° 152 of 11.05.1999 and subsequent updates (DECRETO LEGISLATIVO... 1999), Legislative Decree n° 219 of 10.10.2010 (DECRETO LEGISLATIVO... 2010), implementing Directive 2008/105/EC on standard quality environment in the field of water policy, amending and subsequently repealing Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC, as well as amending Directive 2000/60/EC and transposition of Directive 2009/90/EC laying down, pursuant to Directive 2000/60/EC, technical specifications for chemical analysis and the monitoring of water status (COMMISSION DIRECTIVE... 2009).

Depending on the soil condition petrol can act in different ways. If the fraction is fine, petrol can be captured in the ground, and when such sites are detected the contaminated soil can be removed. When the fraction is medium or coarse the hazard grows because the pollutants migrate down towards the water table and threaten drinking water.

The main sources of contamination is tank leakage at petrol stations, spills on motorways and uncontrolled petrol waste disposals (MACIOSZCZYK 1987). Different processes related to soil and groundwater contamination are shown in Figure 1.

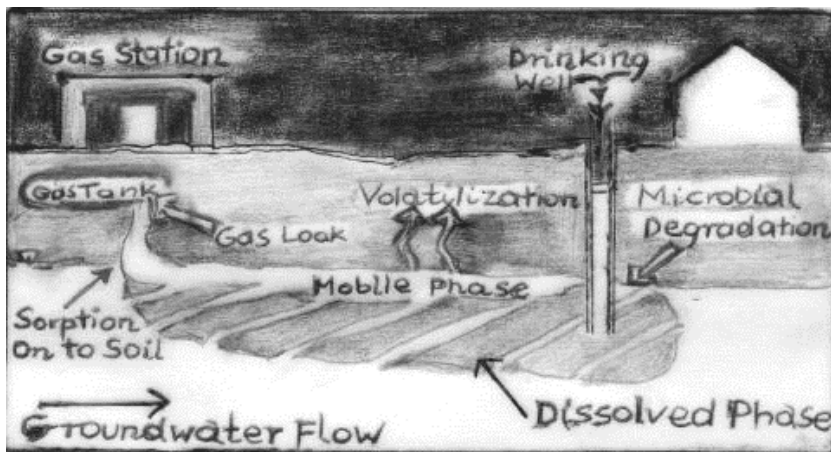


Fig. 1. Different processes related to soil and groundwater (MACIOSZCZYK 1987)

Rys. 1. Niektóre procesy dotyczące gleby i wód gruntowych (MACIOSZCZYK 1987). Groundwater flow – przepływ wód podziemnych, sorption on to soil – sorpcja na glebie, dissolved phase – faza dysolwencji, mobile phase – faza ruchoma, gas tank – zbiornik gazowy, gas leak – źródła gazu, volatilization – ulatnianie, microbial degradation – degradacja mikrobiologiczna, gas station – stacja benzynowa, drinking well – źródło wody pitnej

Aromatic hydrocarbons are the basic compounds occurring in oil and its products. Share of these compounds can achieve level of several percent in oil fractions. The general name for these compounds is BTEX, a shortage for: benzene, ethyl benzene, toluene and xylene (MACIOSZCZYK and DOBRZYŃSKI 2002). Aromatic hydrocarbons are a group of toxic compounds occurring in small amounts in groundwater. Usually, their presence is an effect of environmental contamination caused by petrochemical industry, chemical industry or by traffic.

Aromatic hydrocarbons pollute natural environment by penetrating the atmosphere from where they are distributed *via* snow and precipitation to soil, as well as surface and groundwater.

Contaminants classification

Contaminants can be classified depending on their physical and chemical properties. For liquids, the pertinent properties are their solubility in water, their density, and their vapour pressure. Some liquids such as low molecular weight alcohols, are miscible with water in all proportions. Others have sufficient solubility to be taken completely into solution at the concentrations that occur in groundwater systems. Still others have low solvency so that they separate from water to form immiscible phases separated from the water. The density of immiscible liquids is a key parameter. If the immiscible phase is less dense than water, it will float on the water surface. If it is denser, it will sink. The third variable is the vapour pressure which separates organic liquids into a “volatile organic compound (VOC)” class and a “semi-volatile organic compound (SVOC)” class. The boundary vapour pressure is set at an arbitrary 5 Torr (0.667 kPa). Although the boundaries in solubility and vapour pressure are set by arbitrary choices, it is important to remember that no compound is completely insoluble and no compound is completely non-volatile (NYER 1998).

With these and other aspects of contaminants in mind, it is possible to construct the following classification:

1. Water soluble compounds. These are materials which would be taken completely into solution at all concentrations likely to be found. Water soluble inorganic compounds include ammonia and the nitrate ion, mostly derived from human and animal wastes and perhaps the most widespread of inorganic contaminants. Also water soluble are other inorganic ions such as chloride and sulfate, as well as some highly toxic species such as cyanide ions derived from some industrial wastes. Some organic compounds are also water soluble such as alcohols, carboxylic acids, phenols and some agricultural chemicals.

Soluble contaminants move with the water. In underground aquifers the water from diffuse infiltration through the soil, from sinkhole drains, and from sinking streams ultimately makes its way into the conduit system. The concentration of soluble contaminant should be determined by the concentration at the source adjusted for dilution by other water sources merging in the main conduit (CURRENS 2000).

2. Light, slightly soluble organic compounds. Light, non-aqueous phase liquids (LNAPLs) are those that will float on water (EWERS et AL. 1992). Gasoline, diesel fuel, home heating oil and related petroleum hydrocarbons are the most common examples. Gasoline is a complex mixture of low molecular weight, relatively volatile, saturated

hydrocarbons. It is the presence of percentage quantities of aromatic hydrocarbons, benzene, toluene, ethyl benzene, and xylene (BTEX) that gives gasoline its toxicity.

Solubility of some light aromatic hydrocarbons is presented in Table 1.

Table 1. Solubility in water of some light aromatic hydrocarbons (FETTER 1993)

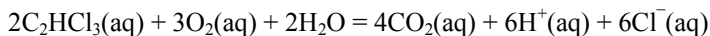
Tabela 1. Rozpuszczalność w wodzie niektórych lekkich węglowodorów aromatycznych (FETTER 1993)

Compound – Składnik	Solubility – Rozpuszczalność (mg/l)
Benzene – Benzen	1 780
Toluene – Toleuen	500
o-Xylene – o-Ksylen	170
Ethyl benzene – Benzen etylowy	150

Local pollution can also be caused by run-off from streets. Their toxic influence on organisms has been proved so their presence in drinking water must be regulated. It is characteristic that the thresholds for aromatic compounds are often on a lower level than the concentrations permitted in groundwater.

3. Heavy, slightly soluble organic compounds. Dense, non-aqueous phase liquids (DNAPLs) will sink in water. Mostly these are chlorinated (or brominated) compounds (KNAUSS 1999). They include such low molecular weight, relatively volatile compounds as methylene chloride, CH_2Cl_2 , trichloroethylene, C_2HCl_3 (TCE) (NYER 1998), and percioroethylene, C_2Cl_4 (PCE) which are widely used as solvents, de-greasers, and dry cleaning agents. These materials are transported in tank car quantities and are often stored in underground tanks. The transport of DNAPL shows both similarities and differences to the transport of LNAPLs. The processes in common include also volatilization, although this process is not as effective because in the case of DNAPL, the overlying water provides a protective blanket. Either type of contaminant can be degraded in the carbonate water environment. The effectiveness of this process is strongly dependent on the specific DNAPL being considered. DNAPLs have a finite solubility (Table 2) and are also gradually stripped away by the continuing flow of fresh water in the conduit. DNAPLs and other compounds are slowly degraded by chemical reaction with the water.

The actual kinetics of this process have been measured by KNAUSS et AL. (1999) for the specific example of trichloroethylene. The degradation reaction is:



The end products of the breakdown process are CO_2 and HCl. Because the reactant H_2O is the solvent, it is present in excess supply and the aqueous oxidation rate equation has the simple form:

$$r_{\text{TCE}} = dC/dt = -kC_0^n$$

where: r_{TCE} is the rate, C is the concentration of TCE, C_0 is the concentration being the initial concentration, n is the reaction order (found $n = 0.85$), k is the rate constant ($k = 5.77 \pm 1.06 \times 10^{-7} \text{ s}^{-1}$ at 100°C) (KNAUSS et AL. 1999).

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Table 2. Vapour pressures and solubility in water of some chlorinated hydrocarbons (FETTER 1993)
Tabela 2. Ciśnienia par i rozpuszczalność w wodzie niektórych chlorowanych węglowodorów (FETTER 1993)

Compound – Składnik	Vapour pressure in 20°C (kPa) Prężność pary w 20°C (kPa)	Solubility – Rozpuszczalność (mg/l)
Methylene chloride Chlorek metylenu	45.60	20 000
Carbon tetrachloride Czterochlorek węgla	11.97	800
Vinyl chloride Chlorek winylu	353.78	1.1
Trichloroethylene (TCE) Trichloroetylen (TCE)	7.98	1 100
Tetrachloroethylene (PCE) Tetrachloroetylen (PCE)	1.76	150

Other DNAPLs include a family of compounds called pesticides chlorinated and polychlorinated biphenyls (PCBs) (NEFF 1985, EDWARDS 1993, GERHARD et AL. 2001).

The action of pesticides, not in itself absolutely selective, leads to damage of various types and extents, also to those non-parasitical species present in the soil and underground waters. In the USA it has been calculated that less than 0.1% of pesticides reach their target parasite, the remainder becoming a potential contaminator of the land and water resources. The processes of biological degradation (mineralization, co-oxidation) represent a key mechanism in preventing accumulation of pesticides in soil. Most reactions of degradation of pesticides, in fact, are due to microbicidal enzymes. Attention must be paid, however, to the fact that the intermediate stages of degradation may have characteristics of persistence and of toxicity which are completely different from those of the initial composite; often toxicity is much higher compared to the initial composite.

PCBs are among the most stable chemical compounds and of low degradability. Their good isolation characteristics and low combustibility led to their use in condensers and high tension transformers. Other uses of PCBs are in lubrication, impregnation, and flame retardant materials.

PCBs in high concentrations cause liver, spleen and kidney damage. Severe poisonings cause organ damage and cancer.

Some PCB manufacturers and applications have been subject to legal restrictions on their manufacture and use since 1989.

4. Pathogens. Viruses, bacteria, Protozoa, and larger organisms are easily transported into underground aquifers because of the absence of filtering from the soil. Most widespread of these are the fecal coliform group of organisms and the fecal streptococcus bacteria. The presence of these organisms is the most common indicator of pollution from sewage or animal waste. Of most concern among Protozoa is *Giardia lamblia* which is released in a cyst form in animal feces and is present in many surface waters (CHESS 1987).

5. About two-thirds of the elements on the periodic table are metals. Most of these do not impose environmental problems because they are rare in nature and are rarely used in commercial products. We deal here only with a limited set of metallic elements, which, however, cover much of the range of behaviour of the remaining elements. Two metals, iron and manganese, make up most of the natural background. These metals occur widely in sedimentary rocks and their oxides and hydrated oxides are common in cave deposits. Nickel and chromium appear in waste from chrome-plating and other non-ferrous metals industry and are typical representatives of the iron-group or transition-group elements. Both are toxic and both have been implicated as carcinogens. Zinc ores occur in carbonate rocks. Zinc and the chemically similar but more toxic cadmium occur widely as “galvanized” coatings on utensils, building materials, and other objects (MCLEAN and BLEDSOE 1992, DOJLIDO 1995, HISCOCK 2005).

Metal in underground aquifers can be divided into three categories: alkaline earth metals derived from the carbonate rock – mainly calcium and magnesium with minor amounts of strontium and barium, heavy metals occurring as part of the natural background – mainly aluminum, iron, and manganese with trace amounts of many other metals, and contaminant metals introduced into the aquifer through human influences. The chemistry of the alkaline earth metals in underground waters is the subject of water geochemistry and is not of concern here (DOJLIDO 1995). However, the concentrations and transport of other metals in underground water systems has received much less attention.

Unlike organic composites, metals cannot be metabolized. The degassing of soils and underground waters contaminated by metals consists primarily in modifying the mobility of the contaminating metals in order to obtain a soil and body of water with acceptable levels in terms of metallic content. The mobility of metals is dependent upon a number of factors. The mobility of metals in a soil-underground waters system depends upon the type and nature of the soil, of the waters, of the concentration of significant contaminants, of the concentration and type of ions that can be considered competitive, of the binders compounds, both organic and inorganic, of the pH level, and of the state of oxidation. However, such data must be site-specific.

Let us consider the characteristics of some metals that are commonly found as contaminants of underground waters. Cd, Cr(III) and Pb are normally present in cationic form (BENJAMIN and LECKIE 1980). These species are generally not mobile in the environment but tend to remain where initially deposited. The capacity to absorb cationic metals increases with increases in pH, in the capacity for cationic exchange, and in the content of organic carbon. In conditions of neutrality, these metals are strongly absorbed by the clayey fraction and may be absorbed by hydrated oxides of iron, of manganese, and of aluminum already present on the site concerned. The cationic metals will precipitate as hydroxides, carbonates or phosphates. In sandy or acidic soils, the cationic metals are more mobile. In atypical conditions for a soil (i.e. pH lower than 5 or greater than 9; higher concentrations of oxidative or reductive substances; high concentrations of colloidal substances or organic and inorganic soluble compounds), which meet as a result of the processes of degassing, the mobility of these metals may be greatly increased.

In some experiments in which solids are present on the surfaces of oxides (g-FeOOH, a-SiO₂, g-Al₂O₃), a competitive absorption between the metals has been noted. In many experiments the absorption of Cd diminishes with the addition of Pb or Cu in the case of each of these three solids (NEFF 1985, KNAUSS et AL. 1999). Competition for

the available surface sites may be observed when a small percentage of the site is occupied. As, Cr(VI) and Hg behave differently from Cd, Cr(III) and Pb. As and Cr(VI) exist in nature in an anionic form. The compounds of chrome have a different toxicity as a consequence of the state of oxidation of the element. It is generally accepted that the oxide of Cr(III) has a low toxicity on account of its low solubility. In reality this valuation does not take into account the fact that Cr(III) may be mobilized at soil level and oxidized into Cr(VI) in an oxidizing atmosphere, generating a species which is recognized as being significantly toxic. Hg presents a high vapour pressure, in fact, at 24°C, an atmosphere saturated in mercury contains approximately 18 mg/m³. This level is 360 times higher than the average (0.05 mg/m³) recommended for exposure. In nature there are three different states of oxidation (RENNEBERG and DUDAS 2001).

Mercury will interact with soil minerals by adsorption-desorption and precipitation-dissolution reactions. Soil texture and mineralogy are important in determining interaction of mercury with soil minerals (RENNEBERG 2000). When soils are contaminated with both mercury and hydrocarbons the two contaminants can interact, resulting in a phase containing both contaminants. The interactions between mercury and hydrocarbons are expected to be similar to the interactions between mercury and natural soil organic matter. Hydrocarbons have S- and O-containing active sites that may bind mercury. Hydrocarbons present in soil at high concentrations can coat soil particles. Mercury can then undergo partition into these non-aqueous liquid phases. The absorption of composites of mercury in the soil is probably the dominating process which determines its presence in the terrestrial environment. The mercury not absorbed in the soil passes into the underground waters and is probably absorbed by plants. Its absorption is dependent upon numerous factors such as chemical structure, the quantity and chemical nature of the soil and waters, level of pH, and its potential for reduction (HOGG et AL. 1978).

Conclusions

Contaminants to underground waters have been broadly classified into water soluble organic and inorganic contaminants, slightly soluble organic contaminants, metals, and pathogens. For each of these categories, there are multiple, complex, and interacting mechanisms for the transport, storage, eventual release of the materials and their passage through the food chain in human organisms.

Environmental contamination problems are becoming more and more severe due to technological and industrial development. In spite of this, as a consequence of strict legal provisions at the regional, European and international level, pollution of water bodies is under control and constantly monitored, as demonstrated by the report of the United States (Rio, June 19, 2012 – STATUS REPORT... 2012), and by the proceedings of the most recent Rio conference (June 20-22, 2012) on sustainable development (RIO+20...). In its turn, technological innovation in the field of chemical analysis (COMMISSION DIRECTIVE... 2009) requires that the states of water bodies be specifically monitored.

In Italy, control of water bodies is under the jurisdiction of ANPA (Ital. *Agenzia Nazionale per Ambiente*, National Agency for Environment), APPA (Ital. *Agenzia Provinciale per Ambiente*, Provincial Agency for Environment), ARPA (Ital. *Agenzia Regio-*

nale per Ambiente, Regional Agency for Environment), and IRSA (Ital. *Istituto di Ricerca sulle Acque*, Water Research Institute).

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Streszczenie. Ilość substancji organicznych otrzymywanych jako produkty uboczne syntezy jest równa ilości substancji naturalnych produkowanych w krajach uprzemysłowionych. Obecnie samo globalne zużycie olejów mineralnych wynosi ponad 3 mln t rocznie. Zanieczyszczenie gleby i licznych źródeł wód podziemnych przez substancje takie, jak detergenty, oleje mineralne, barwniki, pestycydy itp., jest spowodowane ich stałym stosowaniem w życiu codziennym. Stanowią one zagrożenie dla zdrowia, pogarszają jakość gleby i wód podziemnych. Obecność tych substancji w wodzie może również wywołać łańcuch reakcji redoks (np. NO_3^- do NO_2^- , Fe^{+3} do Fe^{+2} itp.). W tych okolicznościach wpływ węglowodorów na rozpuszczalność i transport pierwiastków i ich soli w warstwie wód podziemnych jest oczywisty, jak również to, że jakość gleby oraz źródeł wody pitnej pogorszą się. Wykonane analizy wykazały ślady węglowodorów i innych substancji zanieczyszczających gleby i wody podziemne w całym łańcuchu pokarmowym. Praca wskazuje ostateczne konsekwencje tego zanieczyszczenia, metody analizy substancji zanieczyszczających wody podziemne oraz propozycje dotyczące ochrony gleb i wód podziemnych.

Słowa kluczowe: węglowodory, zanieczyszczenie, wody podziemne

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