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G. Matthess/S.S.D. Foster/A. Ch. Skinner

Theoretical Background, Hydrogeology and Practice of Groundwater Protection Zones

**Volume 6
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International Contributions to Hydrogeology

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PREFACE

Groundwater protection against contamination by man's activities is one of the most important tasks of modern hydrogeology on a world wide scale. However, public acceptance of the need for groundwater protection varies in both developed and developing countries. As a result measures are taken very often at a stage where groundwater is already or will soon be severely contaminated. As a consequence of groundwater residence times of tens to thousands of years, a contamination in groundwater will last for very long times. Its clean-up usually causes tremendous costs. Furthermore, the time lag between introduction of a contaminant into the hydrological cycle and its appearance in groundwater may deceive the public concerning its real threat for the groundwater quality and the water supply.

Therefore, preventive protection of groundwater is necessary. The ecological, economical and social consequences of contamination exceed many times the costs of a suitable protection system. Groundwater protection is a complicated problem, the solution of which needs an interdisciplinary approach. Groundwater protection must be integrated into the processes of planning, investigation, development, use and management of groundwater and other natural resources.

The catchment areas of groundwater systems used for public water supply need special protection. For this purpose groundwater protection areas with zones of graded inhibitions of hazardous activities decreasing with growing distance from the water works prove to be a suitable tool. The regulations in the protection areas restrict or even exclude competing land use. Thus, their correct dimensioning is a demanding task, especially for hydrogeologists.

The International Association of Hydrogeologists (IAH) has discussed aspects of groundwater protection at congresses and symposia since the early 70's and, being aware of its importance, has established a Commission for Groundwater Protection in 1979. During the IAH Regional Meeting at Basle in 1979, a temporary Working Group "Drinking Water Protection Areas in Humid Climates" was constituted to compare the various national protection systems and to define criteria for a scientifically based system for groundwater protection zones. The officers of this working group were A.S. Kleczkowski (Poland) (Chairman), G. Mattheß (Co-chairman) and R. Blau (Switzerland) (Secretary).

Results of the work of this group on the scientific basis and on the practical aspects of a system for the assessment of groundwater protection areas were presented at the IAH/UNESCO workshop at Koblenz in September 1, 1983. This workshop was a part of the second phase of the International Hydrological Programme (IHP II Project A.3.7.).

Thanks to moral and financial support of UNESCO, IAH, and of the governments of some countries and to the international cooperation of hydrogeologists and other scientists concerned with groundwater, this publication was prepared which summarizes the scientific background, the practical experience and knowledge and the recent, sometimes controversial, discussion on protection zones for water supply systems.

Jaroslav Vrba Rene Blau Georg Matthes
Chairman of IAH Commission Secretary Co-Chairman
for Groundwater Protection of IAH Working Group
"Groundwater Protection Areas"

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PART I

THEORETICAL BACKGROUND OF PROTECTION ZONES

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CHAPTER 1

BEHAVIOUR OF CONTAMINANTS IN GROUNDWATER

G.Matthess, A.Pekdeger and J.Schröter

1.1. Introduction

Changes of groundwater quality may be caused directly or indirectly by various activities of man. Direct influences result from natural or artificial substances which are introduced by man into the geochemical cycle of the earth, and ultimately reach the groundwater zone. Indirect influences should be considered to be those changes of quality which are brought about without the addition of substances by man's interference with hydrological, physical and biochemical processes.

Contaminated groundwater may be defined as groundwater which has been affected by man to the extent that it has higher concentrations of dissolved or suspended constituents than maximum permissible concentrations fixed by national or international standards for potable, industrial or agricultural purposes. As natural groundwater (i.e. not influenced by man) may contain constituents exceeding the standard limits, contamination should be defined in these cases as any increase in the concentration of the respective constituent above its natural variations (MATTHESS, 1982) (Tab. 1.1). In this definition the term "groundwater contamination" refers to a reduction of the possible usefulness of the groundwater.

For any minor alteration or degradation of the natural quality of groundwater, resulting from natural processes or from man's activities, ZAPOROZEC (1981) proposes the term

Table 1.1: The concentration of some soluble salts in groundwater and standard mean ocean water (SMOW) compared with drinking water standards.

WHO-International Drinking Water Standards (mg/l)		Observed concentrations in Groundwater (MATTHESS, 1982) (mg/l)		SMOW (mg/l)
Cl ⁻	200 - 250	255,000	(1)	18,173
NO ₃ ⁻	50 - 100	1,950	(2)	0,06
SO ₄ ⁻	200 - 250	302,500	(3)	2,546
F ⁻	0.6 - 1.7	806	(4)	39
Ca ²⁺	75 - 100	350,000	(5)	386
Mg ²⁺	30 - 150	436,000	(1)	1,212

(1) HEM (1970)

(4) WHITE et al. (1963)

(2) GEORGE & HASTINGS (1951)

(5) PINNEKER (1968)

(3) LANG (1941)

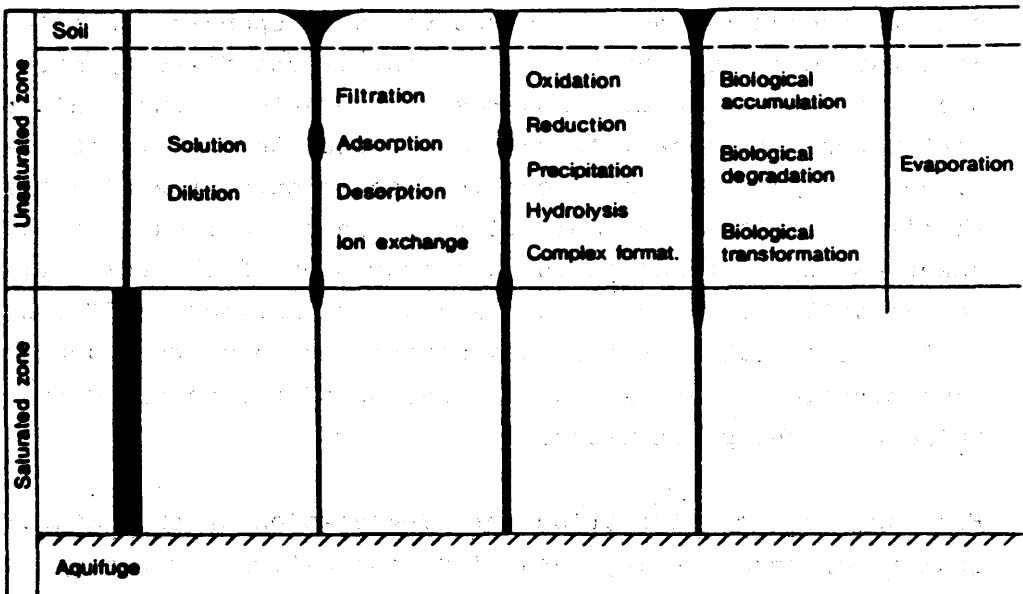


Fig. 1.1 : Schematic view of the chemical, physical and biological processes in the underground (GOLWER, 1983).

"groundwater pollution".

The type, extent and duration of anthropogenic changes of groundwater quality are controlled by man's influence, the geochemical, physical and biological processes underground and the hydrogeological conditions (Fig. 1.1; GOLWER, 1983). The geochemical, physical and biological processes which control the behaviour of contaminants in groundwater are:

Geochemical reactions: solution - precipitation
acid -base reactions
oxidation - reduction
complexation
adsorption - desorption

Physical processes : advection - dispersion
filtration
evaporation
radioactive decay
gas transport

Biological processes : decomposition of organic substances
transpiration
active movement of bacteria
bacterial adhesion

The main contaminants of groundwater are the heavy metals, organic chemicals, and others, mainly fertilizers, bacteria and viruses.

1.1.1. Heavy Metals

Heavy metals are usually defined as metals with densities larger than 5 g.cm^{-3} . This group includes 70 elements with

the atomic numbers 23-33, 40-51, 57-84 and 87-106. The heavy metals with the atomic numbers 61 and 93-106 do not occur naturally.

Heavy metals are used in various ways, as raw materials for numerous industrial products or as catalysts in chemical processes (JACKSON, 1980). Some are constituents of pesticides or fertilizers, which are distributed over large areas by industrial and agricultural activities. Principally all heavy metals may act as contaminants in gaseous, liquid or solid wastes. Appreciable amounts of some heavy metals are set free by the combustion of fossil fuels. Of special importance in groundwater are Cd, Hg, Pb, Cr, Tl and Zn (Tab. 1.2).

Indirect contamination may be due to organic pollution leading to reducing Eh-values in groundwater and thus dissolving iron and manganese. High concentrations of heavy metals in groundwater may be found in mining districts where modern leaching methods using chemical solvents or microbial oxidation of sulfide ores are used (NÖRING, 1973).

Radioactive heavy metals may occur as fission-products in connection with the processing and smelting of uranium ores, the production and reprocessing of nuclear fuel and explosives, the disposal of nuclear wastes, the escape of volatile radionuclides at nuclear power plants and with the various radionuclides used for medical or technical purposes. The use and disposal of radioactive material is generally controlled by respective laws, which may be considered as a model for environmental protection against any dangerous element or compound.

Tab. 1.2: Maximum permissible concentrations (MPC) in potable water and maximum concentrations detected in natural and anthropogenically contaminated groundwater (in mg/l) (MATTHESS 1974, 1981, 1982, SCHEFFER & SCHACHTSCHABEL 1982).

Metal	MPC (WHO)	Highest natural water	Concentrations detected contaminated by man		
			Mine water	Others	
As	50	32,300	-	56,000	
Cd	10	207	41,100	3,700	
Cr	50	400	-	14,000	
Co	n.S.	20	-	-	
Cu	50	344	45,633,000	-	
Fe	100	1,000,000	474,600	700,000	
Hg	1	0.07	-	-	
Mn	50	42,000	841,000	-	
Mo	n.S.	23 (10,000)	-	-	
Ni	n.S.	40,000	319,300	-	
Pb	100	380 (1,200)	1,300	-	
Ra (mBq/l)	n.S.	3,700	-	-	
Sb	n.S.	0.93	-	-	
Sn	n.S.	670	-	-	
U	n.S.	460 (90,000)	-	-	
V	n.S.	330	-	-	
W	n.S.	64	-	-	
Zn	5000	1,900 (177,000)	2,112,000	-	

Gaseous wastes which are apt to propagate contamination within very short time intervals over wide areas usually contain small quantities of some heavy metals, e.g. lead from traffic exhaust (GOLWER & SCHNEIDER, 1973) and the fallout radionuclide ¹⁰⁶ruthenium (AURAND et al., 1971).

Waste water, especially of industrial origin, contains heavy metals in higher or lower concentrations. Solid waste dumps and residues of mining, ore processing and smelting operations are commonly sources of higher local concentrations of heavy metals in the groundwater (MATTHESS, 1972, 1974; SCHÖTTLER, 1972).

1.1.2. Organic Chemicals

Organic chemicals are introduced by man into the geochemical cycle of the earth and can ultimately contaminate groundwater. Organic chemical contaminants may be derived from various sources. The numerous known organic chemicals, for example the pesticides, can be grouped on the basis of their physico-chemical behaviour (Tab. 1.3). Industrial and domestic flue gases and continental dusts contain organic chemicals, e.g. hydrocarbons and halogenated hydrocarbons which may reach the groundwater, dissolved in rain and seepage water (VRBA, 1981; NEUMAYR, 1981; ZOETEMAN et al., 1981).

Organic chemicals are present in municipal sewage and industrial liquid wastes. They may contaminate groundwater either by random leakages of sewers and canals or by intentional disposal into surface waters, by infiltration from septic tanks, by spreading as fertilizers or by injection into deep geological structures. Organic chemicals in solid wastes and sludges, when disposed of by con-

Tab. 1.3: Example for nonvolatile, volatile, nonionic and ionic pesticides (WEBER et al., 1972).

Nonionic-Pesticides		Ionic Pesticides
nonvolatile	volatile	
<u>Dinitroanilines</u>	<u>Dinitroanilines</u>	<u>Cationic</u>
Nitraline	Trifluraline	Diquat
Benefine		Paraquat
	<u>Halogenated Hydrocarbons</u>	Chlormequat
<u>Halogenated Hydrocarbons</u>	Aldrine	<u>Neutral</u>
	Lindane	Atrazine
DDT	Chlordane	Propazine
Endrin	Heptachlor	Simazine
Dieldrin		Ametryne
	<u>Phenylcarbamates</u>	Prometryne
<u>Phenyl-ureas</u>	Chlorpropham	Prometone
Fenuron	Propham	Amitrole
Monuron		Menazone
Fluometuron	<u>Benzonitriles</u>	
Diuron	Dichlobenil	
C-6313		
<u>Substituted Amilides and Amides</u>	<u>Organophosphates</u>	<u>Acid</u>
Propachlor	Dimethoate	Chlorambene
Propanile	Phorate	Dicamba
Alachlor	Parathion	2,4-D
Diphenamide	Disulfoton	2,4,5-T
	<u>Thiocarbamates</u>	Piclorame
	-	Dalapone
	EPTC	Dinoseb
	ODEC	DNOC
	Pebulate	Bromoxynile
	Vernolate	TIBA
	Butylate	DNBSA
		MH
	<u>Aceticmides</u>	
	CDA	

trolled tipping or by composting, are leached by rain and seepage water. Improper waste management causes frequently groundwater contamination (VAN DUIJVENBOODEN et al. 1981; JACKSON 1980).

Pesticides and herbicides used in agriculture and forestry for the control of detrimental organisms are mainly synthetic organic compounds. They are found occasionally in groundwater (NELSON et al., 1981; JACKSON, 1980) (Tab. 1.4).

The organic chemicals which come into contact with rain water, surface water, seepage water or directly with groundwater, are dissolved according to their solubility. Contaminated surface water may reach the groundwater through artificial recharge and pumping of bank-filtered river water or through used or abandoned wells, improper well construction, boreholes and excavations and other hydraulic shortcuts (Tab. 1.5).

Immiscible organic fluids, which are accidentally introduced into the ground by tanker accidents, and by leakage from pipelines or tanks, may form separate organic phases which move and behave according to their physical properties. They may come into direct contact with groundwater or be dissolved in infiltrating water (SCHWILLE, 1981). To an increasing extent, volatile chlorinated hydrocarbons as well as petroleum products (e.g. gasoline, kerosene, diesel, heating and motor oil) are considered as possible groundwater contaminants (AURAND & FISCHER, 1981; TROUW-BORST, 1981).

1.1.3. Other Chemical Contaminants

Industrial and domestic flue gases contain mainly CO_2 , SO_2 and to a lesser extent chlorine, fluorine, and other

Tab. 1.4: The observed concentrations of some pesticides in ground and surface water compared with their solubilities and drinking water standards.

	WHO-International Drinking Water Standards	EEC-Standards (Comments)	measured concen- trations (1) (mg/l)	Solubility (1)	
				in ground- water	in surface- water (20°C)
Dieldrin		0.017 (a)	0.0225	0.0002	
Lindane		0.056 (a)	0.0025	0.000622	0.01
Heptachlor	not stated	0.018 (b)		0.000115	0.001
Methoxychlor		0.035 (b)	0.45	0.0001	

(1) DABAWAS - Datenbank für wassergefährdende Stoffe, Umweltbundesamt Berlin

Tab. 1.5: Detected concentrations. and solubility of some organic compounds (ZOETEMAN et al... 1981. MATTHESS, 1982⁺).

	Solubility ⁺ (g/kg)	Highest concentration detected (µg/l)
<u>Hydrocarbons</u>		
Benzene	1.78	100
Toluene		300
Xylene		600
Ethylbenzene		300
C ₃ Benzene		300
Napthalene		30
Gasoline	0.050-0.500	
Diesel fuel	0.010-0.050	
<u>Halogenated hydrocarbons</u>		
Dichloromethane	16.29	3000
Trichloromethane	1.1	7500
1.1.1. Trichloroethane	1.32	3000
Dichlorobenzene	1.34	10
Tetrachloroethane	2.88	8150 ⁺

substances, which can be detected in atmospheric aerosols. The use of inorganic fertilizers directly increases the quantity of soluble salts in the soil containing chlorides, sulfates, nitrates and phosphates of potassium, calcium, magnesium, ammonia and sodium in varying proportions, as does the supply of organic matter and soluble salts (especially chlorides and sulfates) in manures such as dung and liquid manure (GEORGE & HASTINGS, 1951; SCHWILLE, 1962; VRBA & SVOMA, 1982).

Spray irrigation with liquid wastes and irrigation with sewage may cause an increase in dissolved salts and other substances in groundwater, especially chlorides and sulfates of alkaline earths and alkali metals. An increasingly pressing problem is the contamination of soil by the sewage of large-scale livestock farming (VASAK et al., 1981).

Domestic and industrial liquid wastes disposed to surface waters, infiltrated from septic tanks, spreaded as fertilizers or injected into deep geological structures are examples of groundwater contamination (VAN DUIJVENBOODEN et al., 1981). Furthermore, uncontrolled leakage from sewers and canals must be taken into account (VRBA, 1981). There are numerous, well-investigated cases of groundwater contaminations by disposal of municipal or industrial liquid wastes including arsenic, cyanides, nitrates and phosphates (VAN DUIJVENBOODEN et al., 1981; JACKSON, 1980; MATTHESS, 1982).

Salinity of groundwater may be increased by the application of salt for snow and ice control on highways (GOLWER & SCHNEIDER, 1973). This man-made deterioration will be more serious in the smaller recharge areas and in areas of intensive salt application. Agricultural irrigation may

bring about an increase of salt and nitrate contents of groundwater due to evapotranspiration of the irrigation water, particularly when recirculated.

The infiltration or injection of warm surface waters which have been used for cooling purposes, may change the groundwater quality because of an increased capacity to dissolve constituents in rocks. The use of heat pumps using groundwater for residential heating and cooling may also result into thermal alteration of groundwater (BLAU, 1981).

The pumping of groundwater near a coastline may cause seawater encroachment, especially when the fresh-water body is overpumped. There are similar contaminations caused by pumping of groundwater close to rocksalt deposits (VAN DUIJVENBOODEN et al., 1981).

1.1.4. Bacteria and Viruses

The contamination of subsurface water by pathogenic bacteria and viruses has caused large outbreaks of waterborne diseases. The evaluation of the case histories shows that outbreaks happened only in situations where the contaminated infiltration water could by-pass the unsaturated zone (ALTHAUS et al., 1982). The main contamination sources are septic tanks, leaky sewer lines, sanitary landfills, waste oxidation ponds and land application of wastewater.

The most important pathogenic bacteria and viruses which might possibly be transmitted in the subsurface water path are: *Salmonella* sp., *Shigella* sp., *Yersinia enterocolitica*, *Y.pseudotuberculosis*, *Leptospira* sp., *Francisella tularensis*, *Dyspepsia coli*, enterotoxigenic *E.coli* (ETEC), *Pseudomonades*, *Vibrio* sp., *Legionella* sp. and the viruses, infectious hepatitis virus, polio virus, coxsackie viruses, ade-

novirus, rotavirus, Norwalk-like virus (GERBA & KESWICK, 1981).

Using realistic data on the concentration of pathogenic bacteria and viruses in sewage, surface water and drinking water (EPA, 1978), it can be concluded that the concentration of these microorganisms has to be diminished by 7 log units in potable water with respect to the microbiological contractions in sewage water. This means that the criterion of 99.99 % elimination of pathogenic microorganisms in groundwater, broadly used as a measure of the effectiveness of the elimination processes, is not sufficient (MATTHESS & PEKDEGER, 1981; ALTHAUS et al., 1982).

1.2. Transport of contaminants in groundwater

1.2.1. Dissolution and Precipitation

The subsurface transport of contaminants are controlled mainly by their solution-precipitation behaviour, their persistence and their behaviour with respect to the physical, chemical and biological processes in the groundwater.

1.2.1.1. Solubility

The input concentration of dissolved contaminants depends on their amount and solubility. The solubility of the heavy metals is generally controlled by the most abundant anions in natural groundwater. These are hydroxide, hydrogen carbonate, carbonate, sulfate, chloride, nitrate and, in a reducing environment, sulfide. Therefore the mobility of the heavy metals depends on the solubility of their hydroxides, carbonates, sulfates, chlorides and sulfides. Some of these are listed in table 1.6, in comparison with the poorly-soluble barium sulfate.

Tab. 1.6.: Solubility products of different heavy metals and of BaSO_4 .

(25 °C)			
BaSO_4	$[\text{Ba}^{2+}]$	$[\text{SO}_4^{2-}]$	$1,08 \times 10^{-10}$
CuCO_3	$[\text{Cu}^{2+}]$	$[\text{CO}_3^{2-}]$	$1,37 \times 10^{-10}$
FeCO_3	$[\text{Fe}^{2+}]$	$[\text{CO}_3^{2-}]$	$2,11 \times 10^{-11}$
RaSO_4	$[\text{Ra}^{2+}]$	$[\text{SO}_4^{2-}]$	$4,25 \times 10^{-11}$ (20 °C)
ZnCO_3	$[\text{Zn}^{2+}]$	$[\text{CO}_3^{2-}]$	$6,0 \times 10^{-11}$
PbCO_3	$[\text{Pb}^{2+}]$	$[\text{CO}_3^{2-}]$	$1,5 \times 10^{-13}$
Ni(OH)_2	$[\text{Ni}^{2+}]$	$[\text{OH}^-]^2$	$1,6 \times 10^{-14}$
CdCO_3	$[\text{Cd}^{2+}]$	$[\text{CO}_3^{2-}]$	$2,5 \times 10^{-14}$
Fe(OH)_2	$[\text{Fe}^{2+}]$	$[\text{OH}^-]^2$	$1,65 \times 10^{-15}$
Mn(OH)_2	$[\text{Mn}^{2+}]$	$[\text{OH}^-]^2$	$7,1 \times 10^{-15}$
Hg_2CO_3	$[\text{Hg}^+]^2$	$[\text{CO}_3^{2-}]$	$9,0 \times 10^{-17}$
HgCl	$[\text{Hg}^+]$	$[\text{Cl}^-]$	$2,0 \times 10^{-18}$
NiS	$[\text{Ni}^{2+}]$	$[\text{S}^{2-}]$	$2,0 \times 10^{-21}$
$\text{ZnS, } \beta$	$[\text{Zn}^{2+}]$	$[\text{S}^{2-}]$	$1,1 \times 10^{-24}$
PbS	$[\text{Pb}^{2+}]$	$[\text{S}^{2-}]$	$3,4 \times 10^{-28}$ (18 °C)
CdS	$[\text{Cd}^{2+}]$	$[\text{S}^{2-}]$	$3,6 \times 10^{-29}$
Fe(OH)_3	$[\text{Fe}^{3+}]$	$[\text{OH}^-]^3$	$4,0 \times 10^{-38}$
Cu_2S	$[\text{Cu}^{2+}]^2$	$[\text{S}^{2-}]$	$2,0 \times 10^{-47}$ (18 °C)

Organic acids, salts and bases dissociate into ions and can form true solutions. Such electrolytes, which originate either from man's activities, from the active soil zone or from aquifer materials, include humic acids, simple fatty acids and their salts, naphthenic acids and tannic acids. The solubility may be increased by forming soluble complexes with inorganic or organic compounds (e.g. fulvic acids) or it may be decreased when insoluble complexes are formed, e.g. water-insoluble humic acids. Complexation may be dependent on the pH of the system and the concentration of the substances involved. Polar organic substances, e.g. phenols, are held in solution as molecules through hydrogen bonds. Non-polar organic chemicals, such as petroleum products and halogenated hydrocarbons, are usually poorly soluble (Tab. 1.3). They tend to form separate phases as liquids immiscible in water. The solubilities and observed concentrations of some pesticides are given in table 1.4.

The solubility of other contaminants, such as chlorides, nitrates and sulfates is generally not the limiting factor in their input concentrations.

The solubility of any contaminant can be described quantitatively by the solubility product K_{SP} , whose concept is derived from the law of mass action. In natural waters, which are non-ideal solutions, the ions present are not infinitely dilute. Thus they influence other ion species through the electric field surrounding them, so that the latter appear to be present in smaller amounts. This factor is taken into consideration by using the so-called activities instead of the measured concentrations.

The activities (a_1) of dissolved substances and of the solvent can be determined by measurement of the solubility,

the dissociation constants, the vapour pressure, the freezing point and boiling point, as well as by measurement of the electromotive force of galvanic cells. However, the activities are more easily calculated from analysis values through multiplying by the activity coefficient (f_1):

$$a_1 = f_1 \cdot m_1 \quad (1.1)$$

Since the activity is by definition dimensionless (GARRELS & CHRIST, 1965), the activity coefficient has the dimension l/mole. For dilute solutions the activity coefficients of individual ions can be obtained to sufficient accuracy from equation (1.2), which is based on the DEBYE-HÜCKEL theory of inter-ionic interaction:

$$\log f_1 = \frac{A z_i^2 \sqrt{I}}{1 + a_i B \sqrt{I}} \quad (1.2)$$

A and B are pressure- and temperature-dependent empirical constant specific to the solvent, z_1 is the valency of the ion in the solution specified, and a_1 is a term dependent on the effective diameter of the ion in the solution.

For the ionic strength (I), the empirical equation for dilute solutions holds:

$$I = \frac{1}{2} \sum C_1 \cdot z_1^2 \quad (1.3)$$

The molar ionic strength is calculated in terms of molar concentration C_1 (in mole/l (litre-molarity), in mole/kg

solvent (kilo-molarity) and valency z_1 .

The law of mass action describes the equilibrium

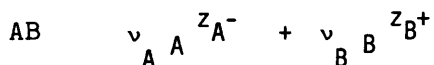


in terms of an equilibrium constant which is dependent on the given temperature and pressure:

$$K = \frac{[C]^{\nu_C} [D]^{\nu_D}}{[A]^{\nu_A} [B]^{\nu_B}} \quad (1.4)$$

In this thermodynamic version of the law of mass action the activities of the species A, B, C and D and the stoichiometric coefficients ν_A , ν_B , ν_C and ν_D are used.

For the dissociation reaction



the activity of the solid phase [AB] is unity by definition, so that eqn. (1.4) can be simplified to the solubility constant in its usual form (1.5)

$$K_{SP} = K_{AB} [AB]_{sat} = [A^{z_A^-}]^{\nu_A} \cdot [B^{z_B^+}]^{\nu_B} \quad (1.5)$$

The GIBBS free energy change for a chemical reaction under standard conditions (1 bar, 25°C) can be calculated from

the ΔG° -values of the various substances involved.

$$\Delta G_r^\circ = \sum \Delta G^\circ_{\text{reaction products}} - \sum \Delta G^\circ_{\text{reactants}} \quad (1.6)$$

Reactions in which the free energy in the reaction products is less than that in the original substances will probably proceed spontaneously unless retarded. As the solid phase surface energy is added to the GIBBS free energy, it follows that fine-grained material is less stable and thus more soluble than coarsened-grained. This is independent of the kinetic effects of smaller grain-sizes which result in faster equilibration (LANGMUIR, 1971).

The standard free energy change is related to the equilibrium constants thus:

$$\Delta G^\circ = - RT \ln K \quad (1.7)$$

where R is the gas constant $8.315 \text{ J} \cdot \text{C}^{-1} \text{ mole}^{-1}$ and T is the absolute temperature in $^\circ\text{K}$.

These equations can be used for the calculation of the equilibrium constants of the reactions. The necessary thermodynamic data have been listed, e.g. in GARRELS & CHRIST (1965). For calculation in aquatic equilibrium systems, computer programs are available e.g. WATEQF (Fig. 1.2; PLUMMER et al., 1976) and GEOCHEM (SPOSITO & MATTIGOD, 1980). They adjust the ion concentrations measured in water samples with help of iterative approaches to a chemical thermodynamic equilibrium model, taking into account the in situ temperature, pH and Eh values and the thermodynamic constants of minerals and mineral phases in the system. As result of the calculation the activities and the quantities of the species present in the solution are listed.

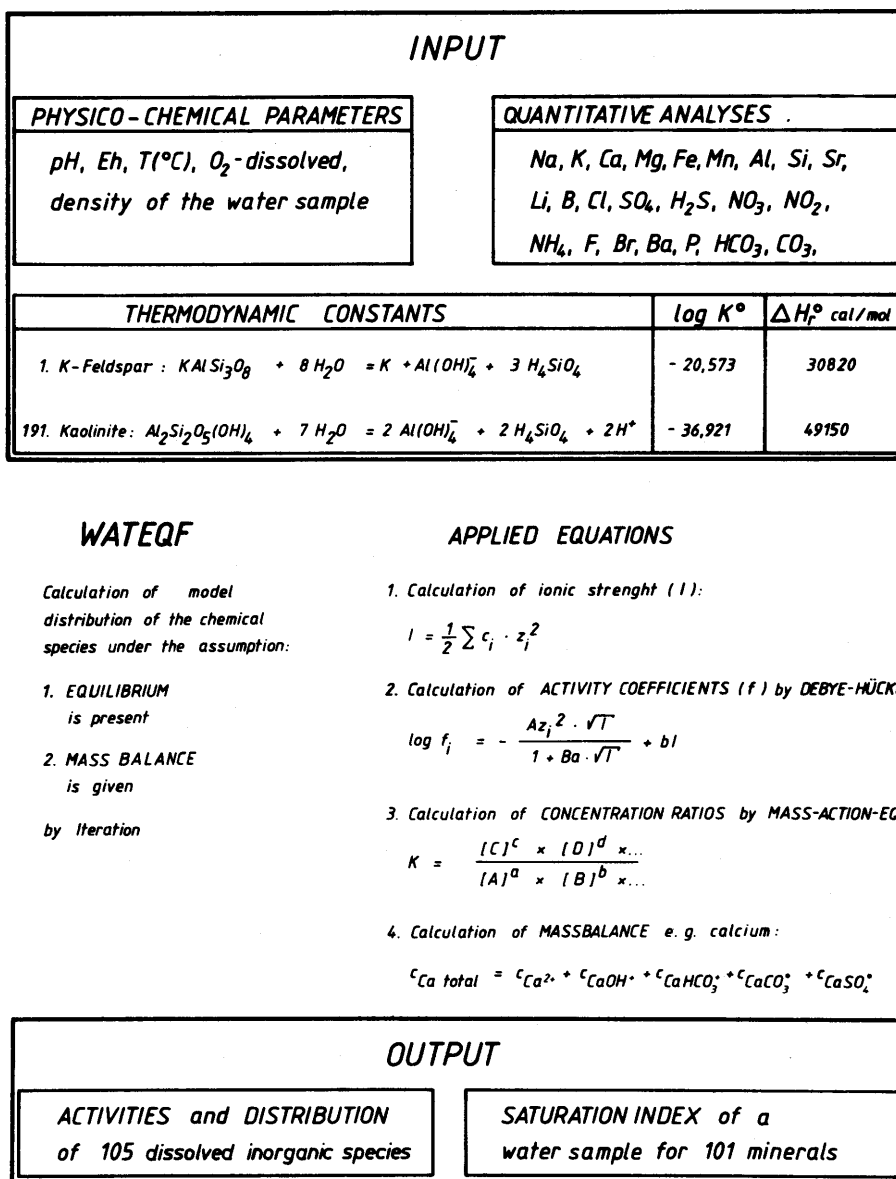


Fig. 1.2 : WATEQF - computer programm (OHSE, 1983 after PLUMMER et al., 1976).

The tendency for dissolution or precipitation can be estimated by comparing the actual ion activity product of the dissolved constituents of a solid species with its solubility product, yielding the saturation index (SI):

$$SI = \log (IAP/K_{SP}) \quad (1.8)$$

where IAP is the ion activity product $\{A\}^x \{B\}^y$ and K_{SP} is the solubility product of the solid species $[A_x B_y]$. If SI is less than 0 the groundwater is undersaturated with respect to $[A_x B_y]$ and a net dissolution of the solid should occur; at $SI = 0$ the groundwater is at equilibrium with the solid; when SI is greater than 0, the groundwater is supersaturated with the solid's constituent ions and net precipitation should occur. However, kinetic delays may prevent dissolution or precipitation (EDMUNDS, 1977; MATTHESS, 1982).

The solubility of some heavy metals, for example iron, manganese, copper and uranium is very strongly influenced by the pH and Eh of the groundwater. Important equilibrium systems include the processes dependent on the redox potential and pH value. These systems can be described with sufficient accuracy by means of equilibrium models based on the laws of chemical thermodynamics (GARRELS & CHRIST, 1965; STUMM & MORGAN, 1981).

Oxidation can be broadly defined as loss of electrons and reduction as gain of electrons. The reduction-oxidation (redox) potential serves as a measure of the relative state of oxidation or reduction in an aqueous system. In a solution which contains various oxidation states of an ele-

ment, the redox potential is measured as an electrical potential between an inert metal electrode and a standard reference electrode, both immersed in the solution.

The redox potential is usually denoted by the symbol E , or, if referred to the standard hydrogen electrode of zero volts, by E_h or E_H . The standard reduction potential E° holds for a redox pair at activities of unity and standard temperature and pressure.

The redox potential is related to the standard free energy change of a reaction, its equilibrium constants, the amounts of the reacting substances present in the equilibrium, and the standard potential, as shown in equations (1.7), (1.9) and (1.10). Equation (1.10) is generally known as the NERNST equation.

$$E^\circ = - \frac{\Delta G^\circ}{n F} \quad (1.9)$$

$$E_h = E^\circ + \frac{RT}{nF} \ln \frac{a_{ox}}{a_{red}} \quad (1.10)$$

- ΔG° = standard free energy of formation of the chemical reaction (in kJ.mole^{-1})
- R = universal gas constant ($8.315 \text{ J.}^\circ\text{C}^{-1}.\text{mole}^{-1}$)
- T = absolute temperature (K)
- n = number of electrons, represented by a multiple of e in the redox equation (difference of electrons between the oxidized and reduced substances)
- F = FARADAY's constant (unit of charge on 1 mole of electrons, 96.564 kJ.V^{-1} or $96.487 \text{ coulomb.mole}^{-1}$)
- E° = standard potential (volts) ($a_{ox} = a_{red} = 1$)
- E_h = redox potential (volts)

a_{ox} and a_{red} = activities of the oxidized or reduced substances in the chemical system under consideration.

Many hydrogeologically important oxidation-reduction reactions are defined by Eh, by Eh and pH, or by Eh, pH and the activity of the individual elements in the solution. These controls of solubility can be represented on stability field diagrams with pH plotted on the abscissa and Eh on the ordinate. The limits of the stability field of the ion species or solids at chemical equilibrium can be calculated for given concentrations of dissolved ions with the help of thermodynamic equilibrium relationships.

Eh-pH diagrams, as published by HEM (1970) and GARRELS & CHRIST (1965), are theoretical models, which contain only that information which has been used in their construction. They are always oversimplifications, because not all the components in the system are known or else cannot be taken into account. Such "mixed potentials" are mostly not amenable to quantitative interpretation (STUMM & MORGAN, 1981). However, they are useful qualitative guides to aid the understanding of relations between the solid and dissolved species, even if they cannot be used as quantitative descriptions or as a tool for the prediction of reaction rates. This approach is important for the assessment of criteria for the mobility of iron, manganese, cadmium, copper, mercury, uranium and many other heavy elements, which are affected by the Eh and pH of groundwater.

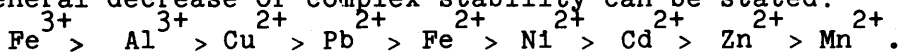
1.2.1.2. Complexation

In concentrated aqueous solutions (for ionic strengths above 0.1, but in many cases also for much lower values) complex ions appear in solution. These can be treated as

thermodynamic entities and are in dynamic equilibrium with the free ions. Complexes are combinations of cations called central atoms with molecules or anions, containing free pairs of electrons (bases), referred to as ligands. Bases containing more than one ligand atom, e.g. oxalate and citrate, are referred to as multidentate complex formers. Complex formation with multidentate ligands is called chelation, and the complexes are called chelates (STUMM & MORGAN, 1981). As a measure of the stability of a complex, a stability constant can be defined on the base of the law of mass action (11). For the association of a complex $M_d L_b$ from d moles of a metal M and b moles of a ligand L:

$$K = \frac{[M_d] [L_b]}{[M]^d [L]^b} \quad (1.11)$$

Complexation by inorganic or organic complex-forming substances may change the solubility. In pore solutions of the unsaturated zone e.g. fulvic acids, tartaric acid, citric acid and salicylic acid tend to increase solubility, whereas humic acids form chelates of low solubility. For the numerous complex-forming substances in the soil, a general decrease of complex stability can be stated:



Humic acid complexes of Cu, Pb, Cd and Zn have log K-values of 8.65, 8.35, 6.25 and 5.72, whereas fulvic acid complexes of Cu, Pb and Zn log K-values of 4.0, 4.0 and 3.6 (SCHEFFER & SCHACHTSCHABEL, 1982).

Water insoluble complexes, e.g. insoluble humic acids, may irreversibly fix cations, which will occupy two thirds of the positions of the total binding capacity of about 200-600 meq metal ion/100 g humic acid (FÖRSTNER & MÜLLER, 1974).

The solubility of an element in natural waters can be determined from the activities of all the dissolved species in equilibrium with the stable solids and from the ratios of the activities of the dissolved species to their concentrations. The difference between the calculated solubility and the measured value gives the possibility of estimating the degree of complex formation or of checking the completeness of the analyses. The formation of complexes is considered in the above-mentioned thermodynamic programmes: WATEQF (inorganic complexes) and GEOCHEM (also organic complexes).

1.2.1.3. Colloidal Solutions

Colloidal solutions contain electrically-charged particles in the size range 10 nm to μm . The charge originates through 3 principal causes. Individual molecules on the surface of a particle can dissociate; thus H^+ or OH^- ions can be separated off and the colloids are left with positive or negative charges respectively. Surface charge may be caused by lattice imperfection at the particle surface and by isomorphous replacement within the lattice. Furthermore, positive or negative ions can be adsorbed from the liquid, and these will charge the particles accordingly (STUMM & MORGAN, 1981). In natural waters (i.e. pH 5-9), colloids of ferric hydroxide are positively charged whereas colloids of MnO_2 are generally negatively charged.

Many of the humic substances are present in natural waters as colloids. Humates tend to form negatively charged colloids, which often are highly dispersed, e.g. stabilized iron III oxide. The occurrence of highly charged ions, e.g. Ca^{2+} and Mg^{2+} , generally leads to the breakdown of the

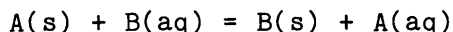
colloidal state, and causes flocculation of the colloids (STUMM & MORGAN, 1981). In the opposite sense the effect of charged substances is known as peptization.

Suspensions and emulsions contain larger, more widely dispersed, particles, which occur within the water as a few large molecules. Thus suspensions and emulsions are transitional to colloidal dispersions.

1.2.1.4. Precipitation and Coprecipitation

Processes which change the chemical properties of a pore solution, e.g. its temperature, Eh or pH or the admixture of other pore solutions with different dissolved constituents may give rise to the precipitation of some compounds. In the precipitates some foreign ions are commonly trapped in or substituted within the structure of the newly formed substances. This process, so-called coprecipitation, is very effective in removing trace elements, e.g. copper, lead, zinc, tungsten and vanadium, when ferric hydroxide and manganese hydroxide are precipitated.

The significant feature of coprecipitation processes is that the new solid phase is more stable and insoluble in the final solution than the original solid phase. Thus the propensity of an ion to be absorbed into a mineral lattice can be expressed by solubility arguments (JACKSON et al., 1980). For the solid solution of B in A, STUMM & MORGAN (1981) propose:



where (s) denotes the solid phase and (aq) the aqueous one. A coprecipitation index can be calculated from the quotient

of the solubility products of the two mineral species:

$$CI = K_{SP}(A) / K_{SP}(B) \quad (1.12)$$

Consequently in a solution of cadmium ions $B(aq)$ undersaturated with respect to CdS ($-\log K_{SP} = 28.44$) but saturated with respect to ZnS ($-\log K_{SP} = 23.96$), it is not unreasonable to expect the coprecipitation of $(Zn, Cd)S$ ($CI \sim 10^{4.5}$). The coprecipitation index merely indicates the likelihood of a reaction occurring ($CI > 1$) but does not guarantee that such will be the case.

1.2.2. Persistence

The period of persistence is frequently given as the time required for the decay of half the initial amount of a chemical compound, but it is also often expressed as the time for detectable levels of the substance to disappear entirely (ALEXANDER, 1977). The decrease of degradable contaminants with time can be approximately described by an exponential function like the radioactive decay (Tab. 1.7). Radionuclides decay with time following eqn. (1.13)

$$C_t = C_o \cdot e^{-\lambda t} \quad (1.13)$$

with C_t = concentration at time t

C_o = initial concentration

$$\lambda_t = \text{decay constant} = \frac{\ln 2}{T_{1/2}} = \frac{0.693}{T_{1/2}}$$

$T_{1/2}$ = half-life

t = time

Tab. 1.7: Radioactive heavy metals

Element	Mass number of Radio- isotope	Half-Life	Radiation
Cerium	144	285 days	β^- , γ
Chromium	51	27.8 days	γ
Cobalt	57	267 days	γ
Plutonium	238	86.4 yr	α , γ
	239	2×10^4 yr	α , γ
	240	6580 yr	α
	242	3.8×10^5 yr	α
Radium	226	1620 yr	α , γ
	228	6.7 yr	β^-
Ruthenium	103	40 days	β^- , γ
	106	385 days	β^- , γ
Uranium	235	7.1×10^8 yr	α , γ
	238	4.5×10^9 yr	α , γ
Zinc	65	245 days	β^- , γ

The halflife in the case of radioactive substances is a physical constant. However, the degradation rate and elimination rate of chemical compounds, bacteria and viruses is influenced by the chemical, physical and biological milieu.

1.2.2.1. Persistence of Organic Chemicals

Organic chemicals may disappear from soil or from groundwater by a variety of means. A reasonable number are subject to chemical reactions, often hydrolytic, to yield non-toxic products. Such non-microbial conversions, although leading to an elimination of the toxicity of the original molecule, do not result in a complete degradation or mineralization of the organic substances. In many instances, however, the decay process is attributable to microbial activity, which is verified by comparing changes in concentration with time in samples of natural soil or water and in samples that have been sterilized or treated with inhibitors to retard microbial metabolism or growth (ALEXANDER, 1977).

Due to the remarkable physiological versatility of subsurface microbial populations a multitude of organic chemicals can be metabolized. The organic chemical may serve as a source of carbon, energy, and occasionally nitrogen or sulfur, so that the population density of the active species in soils treated with pesticides or in contaminated groundwater multiplies at the expense of the chemical. Concomitant with the rise in microbial population is an increasing rate of disappearance of the respective organic compound. Another transformation process is cometabolism, which is defined as the metabolism of a compound by a micro-organism, which is unable to use it as a source of energy or as an essential nutrient (ALEXANDER, 1977).

The kind of transformation depending on a particular species are described in terms of the detoxication, degradation, conjugation, activation and changing the spectrum of toxicity. Detoxication is the conversion of a toxic molecule to a non-toxic product. Degradation or mineralization is the transformation of a complex substrate into simple products, such as CO_2 , H_2O , and sometimes NH_3 or chloride if the molecule contains nitrogen or chlorine.³ Conjugation processes involve microbial complex formation, or addition reactions, which make the substrate more complex or combine the contaminant with cell metabolites. This may be accomplished by the organism catalyzing a reaction that leads to the addition of an amino acid, organic acid, or methyl or other groups to the substrate. Activation is the conversion of a nontoxic substrate into a toxic molecule, e.g. an actual pesticide. A number of commercial chemicals, e.g. the herbicide 4, 2,4-dichlorophenoxy butyric acid (synonym: 4, 2,4-DB) and the insecticide phorate - are transformed and activated microbiologically to give metabolites that are themselves toxic to weeds and insects. The spectrum of toxicity to defined groups of organisms may be changed when they are metabolized to products which are toxic to entirely dissimilar organisms. Sometimes the metabolized products suppress the same kinds of organisms as the substrate compound. For example, the insecticide aldrin is oxidized in culture to dieldrin, which is equally harmful to insects (ALEXANDER, 1977).

Organic compounds are thereafter decomposed mainly by micro-organisms. Recent investigations have discovered surprisingly high numbers of micro-organisms in shallow unconfined aquifers (GOLWER et al., 1973, 1976; HIRSCH & RADES-ROHKOHL, 1983). Nevertheless the degradation of organic contaminants must generally be slower in groundwater compared with soil, for which most data are known.

Evidence is accumulating that these micro-organisms may, under certain circumstances, transform many of the organic contaminants that enter the sub-surface environment. These transformations can lead to total destruction of the contaminant or to the production of new organic contaminants (MATTHESS, 1982; WILSON & McNABB, 1983).

Degradation takes place in an aerobic environment faster than in an anaerobic environment. Under anaerobic conditions, the micro-organisms receive necessary oxygen by reducing oxygen-bearing compounds, particularly nitrates and sulfates. Substances not containing oxygen and with a low hydrolysis constant are more persistent in groundwater than alcohols, ethers and esters. Oxygenated compounds which do not follow this hypothesis generally are alkyl-substituted or contain halogen atoms in the molecule, which results in a much lower degradability (ZOETEMAN et al., 1981).

The diversity of substrates is matched by a diversity of species. Nevertheless, it is rarely possible to predict which species or even which genus is responsible for a particular transformation in nature (ALEXANDER, 1977).

The numerous observations that micro-organisms are important or essential for the degradation of organic chemicals in natural environments lead to two practical observations. First, it seems plausible to expect that the environmental factors governing heterotrophic populations would have a comparable influence on biochemical destruction. Second, the prolonged durability of the compounds designated as persistent is itself convincing evidence that the microbial community has little or no action on the long-lived chemical. The molecules that thus endure have been termed recalcitrant, that is, they fail to be metabolized or

mineralized at significant rates. The quantity of information on longevity of such toxicants is enormous, yet the reasons for varying durations of effectiveness are still largely uncertain (ALEXANDER, 1977).

It is not clear whether the elimination of organic compounds observed in the field gives rise to a secondary organic compound or whether a total mineralisation takes place (SCHWARZENBACH & WESTALL, 1981; ALEXANDER, 1977; fig. 1.3, tab. 1.8).

1.2.2.2. Persistence of Bacteria and Viruses

Two groups of micro-organisms can be differentiated when the subsurface survival of bacteria and viruses is considered:

i) allochthonic pathogenic micro-organisms (parasitic bacteria and enterotoxine-producing bacteria) which enter the groundwater due to contamination

ii) autochthonic groundwater micro-organisms

The autochthonic microbial groundwater micro-organisms flourish under favourable ecological conditions, developing high population densities ($\approx 100/\text{ml}$) (HIRSCH & RADES ROH-KOHL, 1983). The allochthonic bacteria are usually eliminated in the subsurface environment, generally faster than organic chemicals (Fig. 1.4). Under oligotrophic conditions bacteria and viruses may survive without a substantial decrease, or with even slight increase, in the germ number during the first few weeks. After this period, the elimination of bacteria and viruses may be approximately described by an exponential function (Eqn. 1.1; MERKLI, 1975; BERG, 1967).

Tab. 1.8: Persistence of organic substances in groundwater and soils
(ZOETEMAN et al. 1981, MATTHESS 1983).

Organic chemical	Estimated half life	
<u>Hydrocarbons</u>		
Benzene	1	yr
Toluene	0.3	yr
Xylene	0.3	yr
Ethylbenzene	0.3	yr
C ₃ Benzene	0.6	yr
Napthalene	0.6	yr
<u>Halogenated hydrocarbons</u>		
Dichloromethane	10	yr
Trichloroethane	2	yr
1.1.1. Trichloroethane	1	yr
Dichlorobenzene	1	yr
<hr/>		
Selected Data on Persistence of Pesticides in soils (Alexander, 1977)		
Chlordane	2 - 4	yr
DDT	3 - 10	yr
Dieldrin	1 - 7	yr
Heptachlor	7 - 12	yr
Toxaphene	10	yr
DDVP	17	days
Methyl demeton S	26	days
Thimet	2	days

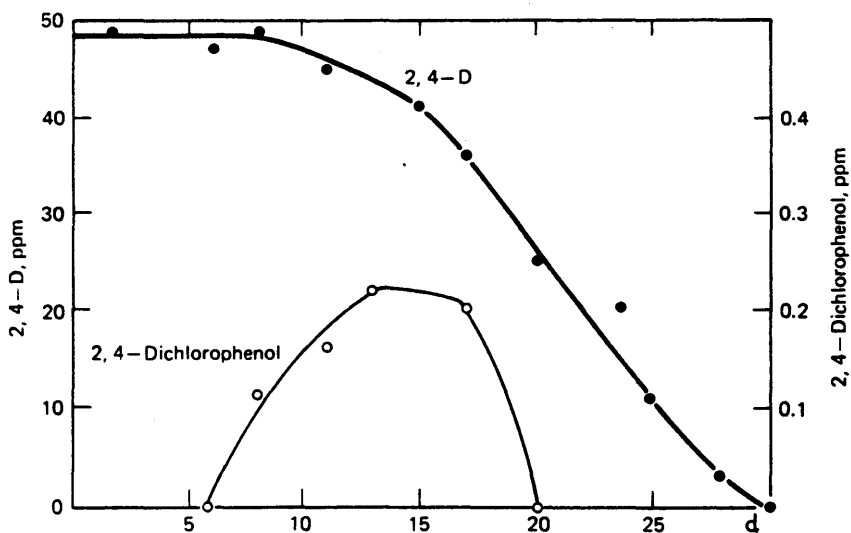


Fig. 1.3 : Metabolism of 2,4 - D (2,4 -dichlorophenoxyacetic acid) and formation of 2,4 - dichlorophenol in soil. Note that the concentration of the product is low (ALEXANDER, 1977).

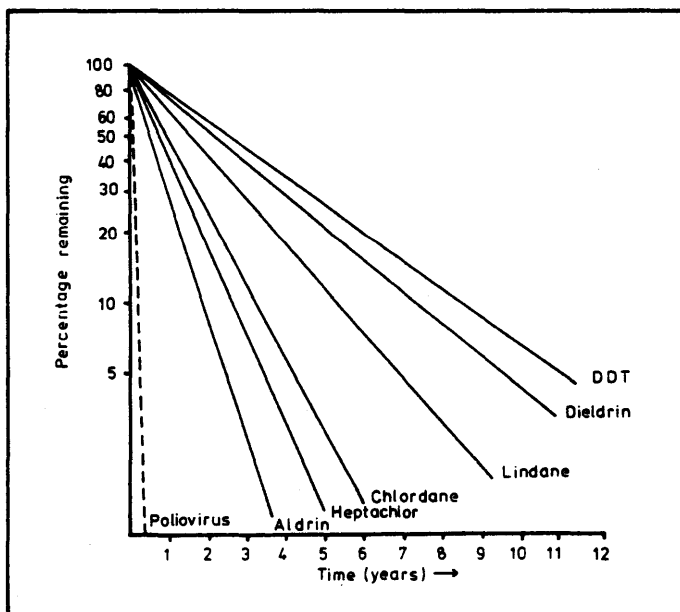


Fig. 1.4 : Degradation of some organic chemicals compared with polio virus (after EDWARDS, 1966).

$$C(t) = C_o e^{-\lambda_t(t-t_o)} \quad (1.14)$$

$t \leq t_o$ and $t_o \leq 20$ days

$C_o, C(t)$ = initial concentration and concentration at time t

λ_t = elimination constant = $\frac{\ln 2}{T_{1/2}}$

t = time

$T_{1/2}$ = half-life (mostly between 1 and 20 days)

Bacteria and viruses may survive many weeks in sewage, septic tanks and waste deposits, e.g. *Salmonella* sp., persists 3-10 weeks.

Bacteria and viruses can survive even longer (> 6 months) in the deeper parts of the unsaturated zone and in groundwater, when oligotrophic conditions are present. This means that under certain ecological conditions bacteria and viruses may survive a long period in the unsaturated zone, until they are transported into and propagated in the groundwater.

Their elimination is the combined effect of the physical (temperature), biological and chemical conditions. The biological factors are, as described, the most important factors for the survival of pathogenic bacteria and viruses. Elimination is faster at high temperature (37°C), at pH-values of about 7, at low oxygen concentrations and at high content of dissolved organic substances, conditions encountered in contaminated soils and groundwater. In this case the autochthonic bacteria are activated, and they act antagonistically towards pathogenic microorganisms. The elimination constant depends on the physical, chemical and biological parameters mentioned above and is specific for different microbial species (Fig. 1.5-1.9). It is not

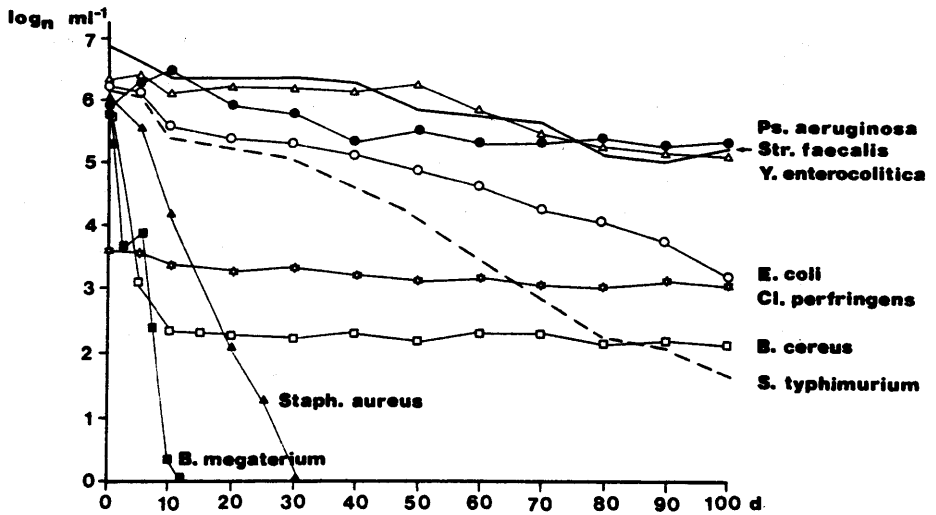


Fig. 1.5 : Persistence of bacteria in sterilised ground-water (FILIP et al., 1983).

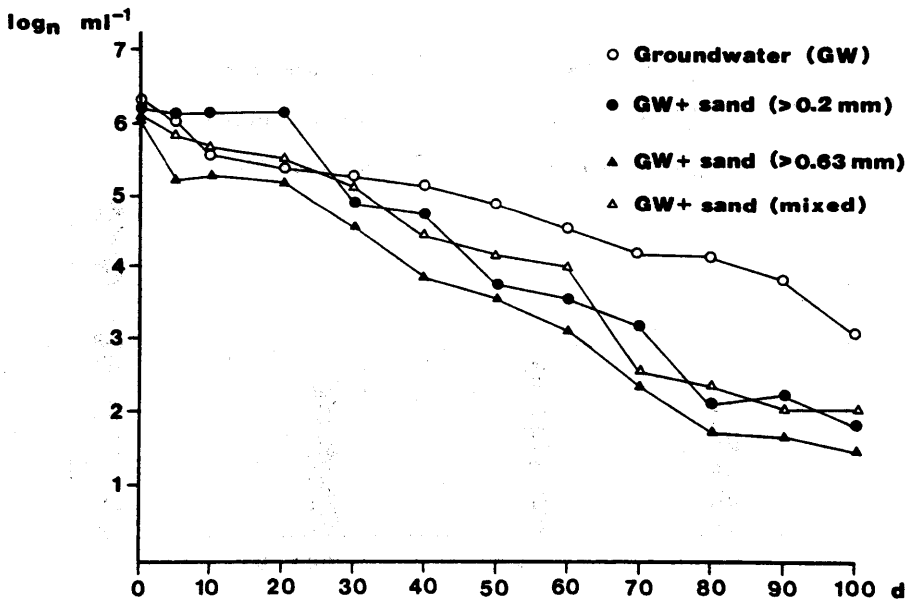


Fig. 1.6 : Persistence of *E. coli* in groundwater in the presence of solid substances (FILIP et al., 1983).

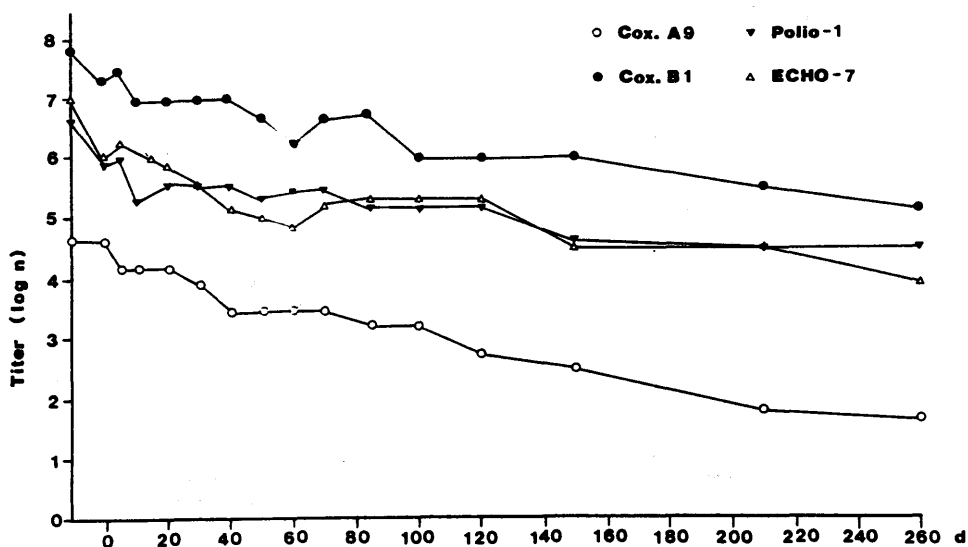


Fig. 1.7 : Persistence of viruses in sterilised groundwater (DIZER et al., 1983).

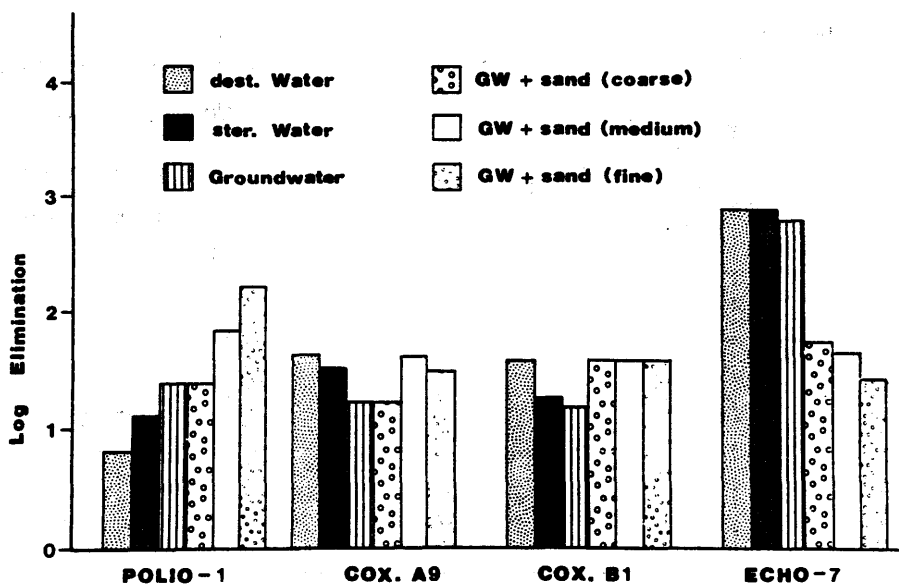


Fig. 1.8 : Persistence of viruses in the presence of solid substances (DIZER et al., 1983).

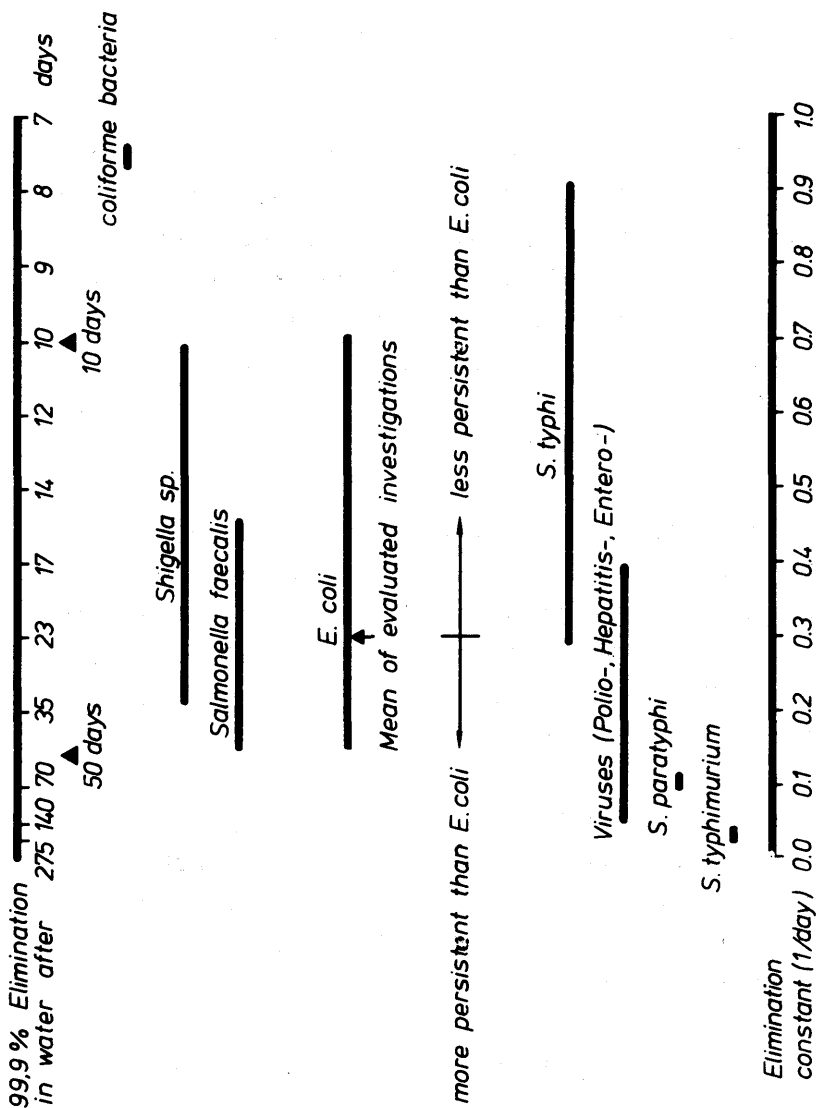


Fig. 1.9 : Elimination constant and 99.9 % elimination of some relevant bacteria and viruses in groundwater (PEKDEGER & MATTHESS, 1982).

possible to predict, with the required accuracy, the elimination constants on the basis of the controlling factors, and therefore, they must be measured for each specific species and environment. The published values are contradictory and vary over a broad range (MATTHESS & PEKDEGER, 1981; ALTHAUS et al., 1982).

The published data on the removal of degradable contaminants and bacteria and viruses in the subsurface are not consistent with the long persistence times observed in laboratories and contaminated wells. Therefore, the persistence time cannot be the only criterion characterizing the purifying effect of subsurface passage of water.

1.2.3. Adsorption and Retardation

Geological materials, both inorganic and organic, have surfaces with small unbalanced electrical charges which attract ions in groundwaters. The forces which bind these ions range from VAN DER WAALS forces to chemical adsorption by valence bonds. Valence bond ions are eventually adsorbed into the internal structures of the minerals. This adsorption mechanism is important in ion exchange, where equivalent quantities of bound ions are displaced by other ions from the solution. The most effective adsorbing substances in soils and rocks are clay minerals, zeolites, hydroxides of iron and manganese, humic substances, plant roots, microbial slimes and micro-organisms.

Dissolved and suspended contaminants are subject to adsorption. This may be described for dilute suspensions by the FREUNDLICH isotherm, which defines the equilibrium between the concentration of the dissolved and suspended (C_s) and adsorbed (C_a) species (1.15)

$$C_a = k C_s^n \quad (1.15)$$

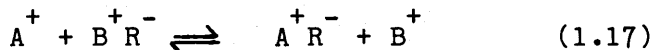
where k and n are assumed to be specific constants for the investigated rock and the contaminant. Another possible description may be the LANGMUIR' isotherm (1.16) which is possibly the better mathematical definition:

$$C_a = \frac{K \cdot b \cdot C_s}{1 + K \cdot C_s} \quad (1.16)$$

where K is a constant relating to the bonding energy and b the adsorption maximum when the adsorbent is completely saturated.

The stoichiometric adsorption-desorption process of bound and dissolved ions is called ion exchange. The direction, extent and velocity of an ion exchange process depends upon the relative concentration and the properties of the competing ions and of all other dissolved components. The exchange processes are reversible and can be described by the law of mass action. The intensity of the bond is different for the various ions and exchange substances. The intensity increases with the valence state, with the atomic number and probably with the ionic radius.

The ion exchange process with the competing ions A^+ and B^+ and the cation exchanger R^-



can be described by a selectivity quotient

$$K_B^A = \frac{(A^+R^-)(B^+)}{(A^+)(B^+R^-)} = \frac{(A^+R^-)/(A^+)}{(B^+R^-)/(B^+)} \quad (1.18)$$

where (A^+) and (B^+) are the concentrations of these ions in solution and (A^+R^-) and (B^+R^-) are their concentrations in the adsorbed phase.

The selectivity quotient may be considered as a mass action equilibrium constant if (1) the activities of all four forms may be calculated, (2) the adsorption reaction is reversible and (3) secondary reactions which are not readily reversible, such as ion fixation due to lattice collapse, do not occur and so prevent the attainment of true equilibrium. The estimation of the activities of the adsorbed phases, A^+R^- and B^+R^- , has been discussed by TRUESEDELL (1972).

The amount of exchangeable ions in meq/100 g solids at pH 7 is commonly known as the ion exchange capacity.

The selectivity quotient is related to the distribution coefficient (K_d), if the cation exchange capacity (Q) of the exchanger R^- and the total competing cation concentration in solution (C) are known and the system under consideration is at equilibrium (JACKSON et al., 1980):

$$K_B^A = K_d^A \frac{[C - (A^+)]}{[Q - (A^+R^-)]} \quad (1.19)$$

If the contaminant A^+ is present in amounts much less than

B^+ , (A^+) is much less than C and (A^+R^-) than Q , the relationship can be simplified to

$$K_d^A = \frac{K_B^A \cdot Q}{C} \quad (1.20)$$

Therefore the distribution coefficient is directly proportional to the cation exchange capacity and the selectivity quotient, and inversely proportional to the total competing cation concentration. If (A^+) and (A^+R^-) are very small, (B^+) is close to C and therefore K_d^A is proportional to $(B^+)^{-1}$. More complex cases, for example involving monovalent-divalent exchange, may be developed using similar reasoning.

The empirical distribution coefficient K_d is a useful measure for the affinity of a specific geological material for a certain contaminant. It is defined as the ratio of the adsorbed substance in meq/100 g solid to the concentration of the same substance in solution in meq/l. In dilute suspensions, K_d is equal to the coefficient of the FREUNDLICH' isotherm.

The continuous adsorption - desorption reactions cause a retardation of the contaminant with respect to the surrounding groundwater, which is described by the retardation factor R_d , the quotient of the mean groundwater velocity (v_w) to the mean transport velocity of contaminant (v_c). The retardation factor can be calculated if the distribution coefficient K_d of the contaminant is known with the

aid of the one-dimensional retardation equation:

$$R_d = \frac{v_w}{v_c} = 1 + \frac{\rho_b}{n} \cdot K_d \quad (1.21)$$

using the bulk density of the aquifer material (ρ_b) and its porosity (n). The calculation of the transport velocity of contaminants with the help of K_d - and R_d -values holds only if one reversible, kinetically-fast adsorption process is present (JACKSON, 1980). In other cases the transport characteristics of a contaminant deviate from an ideal tracer (Section 1.3.).

From this discussion it follows, that adsorbed heavy metals can be partially desorbed by other competing ions in groundwater if these ions occur in sufficiently large concentrations. The higher the ionic concentration the faster the contamination will move through successive exchanges.

Heavy metals will also be adsorbed selectively: hydroxides and oxihydrates of iron favour zinc, copper, lead, mercury, chromium, molybdenum, tungsten and vanadium whereas the hydroxides and oxihydrates of manganese prefer copper, nickel, cobalt, chromium, molybdenum and tungsten and clay minerals, zinc, copper, lead and mercury (KRAUSKOPF, 1956).

The unspecific adsorption due to the electrostatic forces of grain surfaces has a relatively small bonding force. The specific adsorption, especially by hydroxo-complexes of the heavy metals ($Me(OH)^+$, $Me(OH)_2^0$), on the surface of Fe-, Al- and Mn-oxides have stronger bonding forces. Therefore the specific adsorption on oxide surfaces increases with increasing tendency to form hydroxo-complexes and hydroxi-

des in the order $Cd < Ni < Zn \ll Cu < Pb$ (SCHEFFER & SCHACHTSCHABEL, 1982).

The hydrogeological implication of the complexing of the transition metal ions (e.g. Fe, Mn, Co, Zn, Pb, Cu, Ni, Cr) by inorganic or organic ligands is to alter their adsorption or precipitation from solution. The adsorption of radioactive cobalt and zinc is significantly decreased in the presence of dissolved organic carbon (DOC), which is found in all natural waters. This decrease is due to the formation of organo-cobalt and organo-zinc complexes whose sorption behaviour may differ from that of the hydrated metal ions and whose increased solubility in solution reduces the effectiveness of precipitation (JACKSON et al., 1980).

Chlorinated hydrocarbons and other non-polar organic compounds are increasingly adsorbed the more lipophilic they are. As a measure of lipophilicity the octanol/water partition coefficient K_{ow} can be used. The content of organic carbon C_{oc} in the solid phase and the K_d are related:

$$K_d = C_{oc} \cdot K_{sc} \quad (1.22)$$

where K_{sc} is a corrected sorption constant. The water solubility S and the corrected sorption constant K_{sc} are closely related to the octanol/water partition coefficient K_{ow} :

$$\log S = -n \log K_{ow} + c \quad (1.23)$$

$$\log K_{sc} = a \log K_{ow} + b \quad (1.24)$$

By combination of eqns. (1.23) and (1.24), equation (1.25) may be derived, which describes the adsorption behaviour of

a substance as a function of their solubility in water S (ZOETEMAN et al., 1981):

$$\log K_{sc} = - 0.782 \log S - 0.27 \quad (1.25)$$

For an average groundwater flow velocity of less than 1 m/d it is assumed that adsorption equilibrium is reached. At higher flow velocities substances may move faster than can be predicted from their distribution coefficients.

For a sedimentary aquifer in the lower Glat Valley (Switzerland) SCHWARZENBACH & WESTALL (1981) published R_d -values between 1 and 7 for volatile chlorinated hydrocarbons and an R_d of 142 for hexachlorobenzene. The sediment contains 20 wt-% of fine-grain fraction ($< 125 \mu m$) and 0.02 % organic substance; its density is 2.5 g/cm^3 and its porosity is 25 %.

The transport velocity of pathogenic bacteria and viruses may also differ from groundwater velocity (Fig. 1.10). The micro-organisms are subject to adsorption on soil and rock particles. The adsorption of bacteria and viruses takes place quite rapidly (24h and 2h respectively; ALTHAUS et al., 1982). The desorption rate is less known and needs to be measured by further investigation.

The retardation factors can be obtained by laboratory and field tests (MATTHESS & PEKDEGER, 1981). Thus, retardation factors between 1 and 2 were found for the indicator bacteria *Escherichia coli* and the tracer bacteria *Serratia marcescens* (JUNG & SCHRÖTER, 1983; HAVEMEISTER & RIEMER, 1983; HAVEMEISTER et al., 1983; SCHRÖTER, 1983), if the scale of the experiments were large enough ($> \text{few meters}$). In small scale experiments the formulae used yield an apparently higher retardation factor, due to the fact that

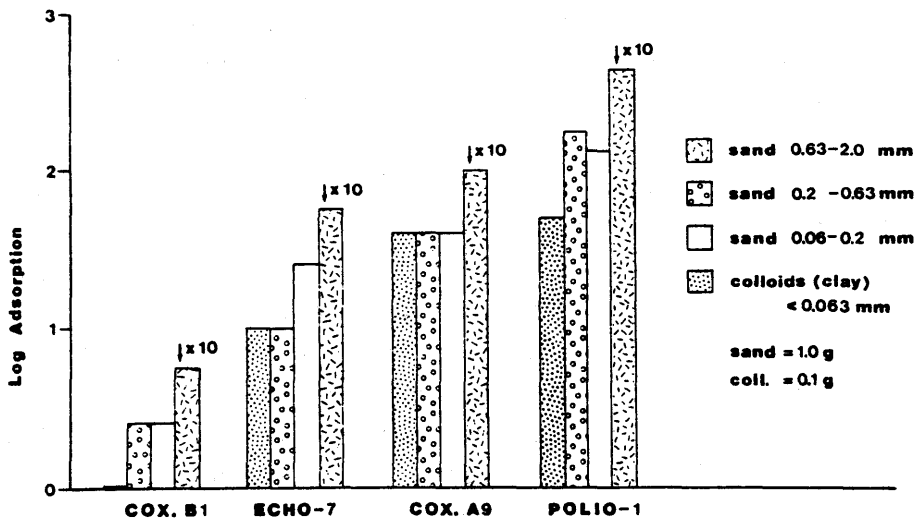


Fig. 1.10: Adsorption of viruses on underground particles (DIZER et al., 1983).

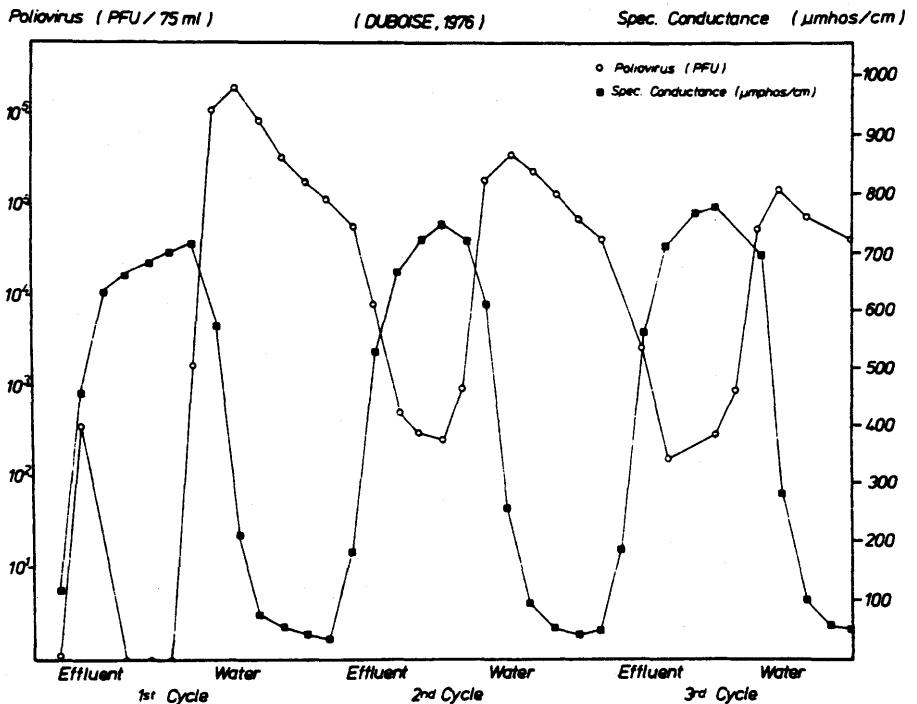


Fig. 1.11: Transport of viruses in sewage effluent and distilled water (DUBOISE et al., 1976).

a certain portion of the bacteria are held by filtration mechanisms which are reversible over short distances. Filtration processes are discussed later in this paper.

Model calculations using the data of known adsorption coefficients and elimination constants, show that underground passage can provide a very effective protection against virus contamination. Most of the pathogenic viruses, especially the polio viruses, have high retardation factors which range up to 500. However, the viruses can be desorbed, when the chemistry of water changes (Fig. 1.11; DUBOISE et al., 1976), for example when cation concentrations decrease (e.g. due to very heavy rainfall), and thus may then travel further. This may also explain why in arid climates, the land treatment of sewage is generally reported to be more effective than in humid regions.

1.2.4. Advection and Dispersion

The extension of an anthropogenically-influenced groundwater zone depends on the hydrogeological conditions. The prediction of the effects of anthropogenic interference requires a knowledge of the position of the water table, the hydraulic gradient, the distance of any wells or springs from any hazardous activities and the properties of the rocks, such as adsorption capacity and hydraulic conductivity. The subsurface movement of any contaminant is influenced by the moisture content and water balance of unsaturated zone, the hydraulic gradient and the water balance of the saturated zone. These parameters are controlled by the volume of water in the system, which depends on climate, topography and hydraulic conductivity. The nature of the rock defines the hydraulic conductivity and the porosity. Groundwater velocities in porous aquifers are

usually less than one metre/day up to a few metres/day, velocities above 10 m/day are restricted to very coarse sediments and high hydraulic gradients. In fractured aquifers, groundwater flow velocities up to 8000 m/day are quoted, and, in karstic aquifers up to 26,000 m/day. Therefore, it can be concluded that the propagation of contaminants in fractured and karstic aquifers is much faster than in non-indurated porous aquifers (MATTHESS & PEKDEGER, 1981). The greater width of interstices in the former enable the subterranean transport of suspended matter, particularly microorganisms, viruses and substances giving rise to turbidity.

Contaminated groundwater can be diluted by mixing with pure groundwater until the concentrations of the contaminants virtually reach natural background levels. The process of mixing causes a distribution of the contaminants in time and space so that the concentration in the contaminated groundwater plume decreases with time and transport distance, whereas the volume of the contaminated plume increases (Fig. 1.12). This processes may be described by the general transport equation in vectorial form (BEAR, 1972):

$$\frac{\delta C}{\delta t} = \text{div} \left(\frac{D}{R_d} \cdot \text{grad } C \right) - \frac{v_w}{R_d} \cdot \text{grad } C - \lambda_t C \quad (1.26)$$

- D = coefficient of hydrodynamic dispersion = $D' + D_d + D_e$
- D' = coefficient of hydromechanic dispersion
- D_d = diffusion coefficient
- D_e = coefficient of active mobility of bacteria
- $\text{grad } C$ = concentration gradient
- v = average groundwater velocity
- R_d = retardation factor
- λ_t = elimination constant

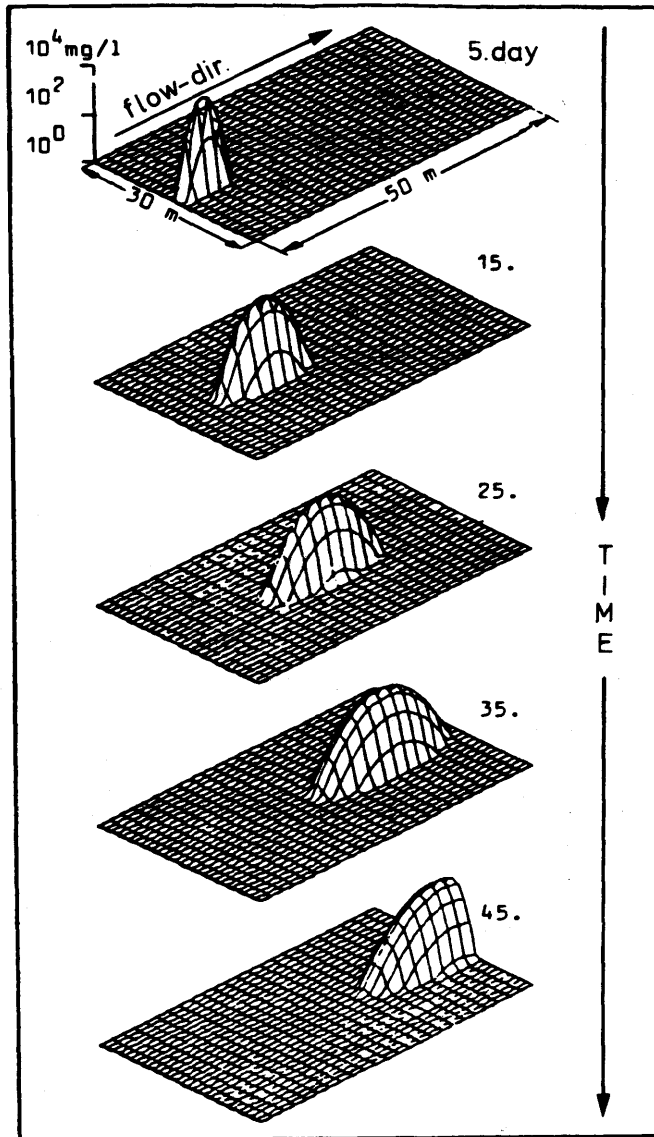


Fig. 1.12: Distribution of a contaminant in time and space in a sandy aquifer (SCHRÖTER, 1983).

The dispersion of contaminants in groundwater is described by the hydrodynamic dispersion coefficient D . It depends on both the textural and structural characteristics of the aquifer, given by the dispersivity (α) and on the groundwater velocity v_a (Fig. 1.13, eqn. 1.16):

$$D = \alpha \cdot v_a^b \quad \text{with} \quad 0,9 < b < 1,2 \quad (1.27)$$

The longitudinal dispersion coefficient and the longitudinal dispersivity, measured in flow direction, are 10 to 1000 times greater than the transverse dispersion coefficient and the transverse dispersivity.

Numerical solutions are available for practical purposes with defined boundary conditions (LENDÄ & ZUBER, 1970; HIBSCH & KREFT, 1976), which allow the treatment and prediction of transport processes (DAMRATH et al., 1979; FRIED 1975, 1981; KONIKOW, 1981; ROBERTS & VALOCCHI, 1981; SCHRÖTER, 1983).

$$c_{\max}(s, y, t) = \frac{GM \cdot s}{4 \cdot \pi \cdot n_e \cdot M \cdot v_a \cdot t \sqrt{D_e \cdot D_t}}$$

$$c(s, y, t) = c_{\max}(s, y, t) \cdot \exp - \left(\frac{(s - v_a \cdot t)^2}{4 \cdot D_e \cdot t} + \frac{y^2}{4 D_t \cdot t} \right) \quad (1.28)$$

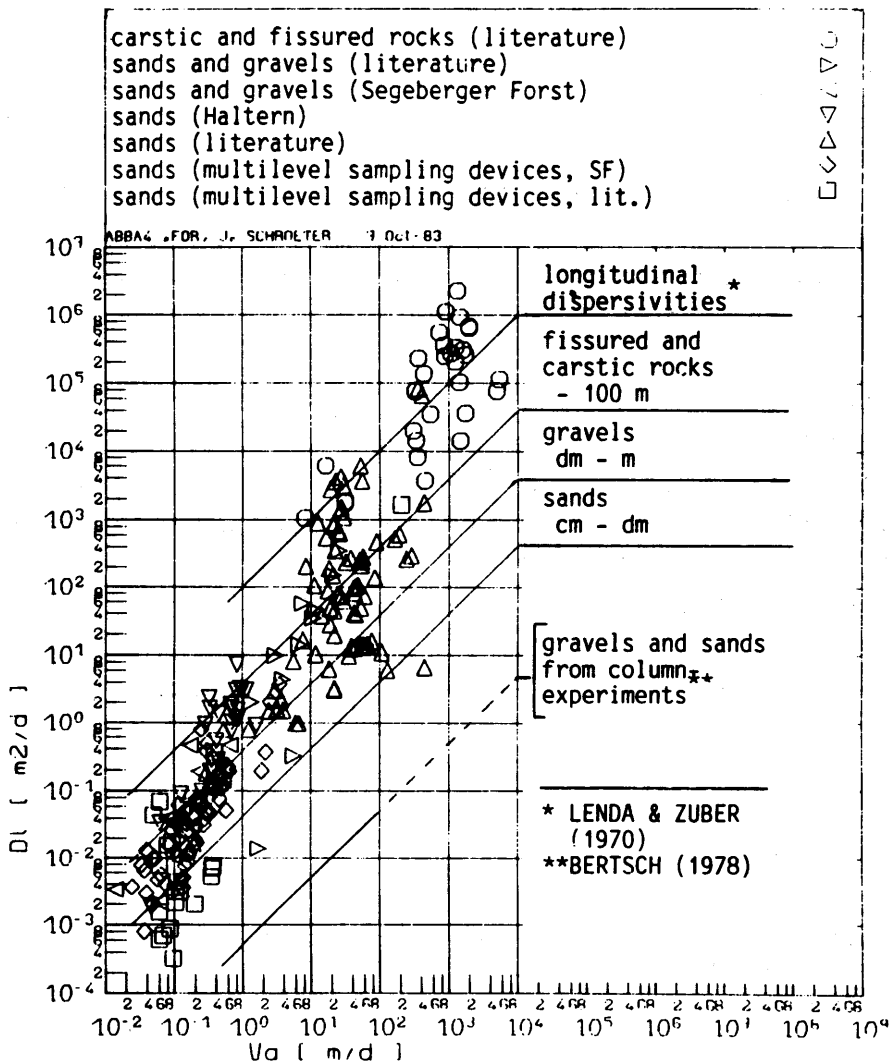


Fig. 1.13: Longitudinal dispersion coefficient in dependance of mean flow velocity (SCHRÖTER, 1983).

$$c_{\max}(s,t) = \frac{GM \cdot s}{n_e \cdot F \cdot v_a \cdot t \sqrt{4 \cdot \pi \cdot D_e \cdot t}}$$

$$c(s,t) = c_{\max}(s,t) \cdot \exp - \frac{(s - v_a \cdot t)^2}{4 \cdot D_e \cdot t} \quad (1.29)$$

GM = input mass

s = flow distance

y = perpendicular distance to flow distance

t = time

n_e = effective porosity

M_e = thickness of the two dimensional aquifer

F = cross section of the one dimensional aquifer

D_l = longitudinal dispersion coefficient

D_t = transversal dispersion coefficient

The dispersion coefficient and the dispersivity can only be measured exactly with an ideal tracer. Transport processes of contaminants influenced by adsorption/desorption- or filtration-processes will show differing transport velocities and dispersion coefficients (MATTHESS & PEKDEGER, 1981; SCHRÖTER, 1983). The multiple reactive solute transport in unidirectional groundwater flow regimes is described by the combined two-dimensional mixing cell/analytical solution presented by SCHULZ & REARDON (1983).

The coefficient of hydrodynamic dispersion and dispersivity increases with decreasing porosity, increasing grain size

and uniformity coefficient. These effects can be measured only in laboratory experiments (KLOTZ, 1973; SCHRÖTER, 1983) but, with homogeneous materials, KLOTZ (1973) found no causal relation between dispersivity and hydraulic conductivity (K). The K-value increases with decreasing unconformity, while the dispersivity decreases with decreasing unconformity.

The volume of a contaminant plume is not decreased by flowing through a porous medium with a lower dispersivity value. Thus, the effects of each nonhomogeneous layer through which a tracer passes are accumulated in higher dispersivity values (GSF, 1979, 1980; SCHRÖTER, 1983). Only few laboratory experiments have dealt with stratified nonhomogeneous layers; BERTSCH et al. (1972) has described tracer experiments in a tank filled with three layers of different sands and gravels. In each layer dispersivity values were measured, which are equal to those measured in column experiments. However, the different hydraulic conductivities of the three layers caused a spreading of the tracer and three distinct peaks reached the measuring points at different times. Hence the mass of the tracer substance was divided into three parts depending on the respective K-values. The over-all dispersivity calculated from the tracer breakthrough in the three layers was much higher than the values for the individual layers and in the same order of magnitude as measured in the field.

The dispersivity coefficients measured in column experiments are in the order of 0.1 cm - 1 m, in field experiments in the order of 0.1 - 100 m, and in the fractured and karstic rocks in the order of 10 - 1000 m (MATTHESS & PEKDEGER, 1981).

Under field conditions, each aquifer has to be considered as an nonhomogeneous layered sediment. In field studies with multilevel sampling devices (PICKENS & GRISAK, 1981; SCHRÖTER, 1983), the different groundwater velocities in the single layers and dispersivity values for each layer can be measured, which show that they are of the same order as those of column experiments. However, a well, which penetrates a stratified aquifer, shows a much broader breakthrough curve, reflecting the K-value distribution of the aquifer. This effect was theoretically described by MERCADO (1967) and GELHAR et al. (1979):

$$\alpha_1(s) = \left(\sigma K / \bar{K} \right)^2 \cdot s \quad (1.30)$$

An increase of dispersivity with increasing flow distance (s) through porous media and through karstic or fractured aquifers can be seen in a comparison of about 150 tracer field studies (Fig. 1.14). Although the results of these tracer experiments are influenced by various disturbances, e.g. by high tracer input volumes, by changes in the flow direction, which causes irregular breakthrough curves, and by adsorption processes of the different tracers used (SCHRÖTER, 1983), the comparison gives a general idea of the relation between structural and textural influences on dispersivity and fits the theoretical evaluation quite well. The changes in flow direction of the groundwater, caused by the variation of hydraulic conditions or geological nonhomogenities, give rise to an increase of measured dispersivities, which generally increase along the flow path. The general relation between dispersivity (α_1) and flow distance (s) is given by the equation (1.31):

$$\alpha_1(s) = a \cdot s^b \quad (1.31)$$

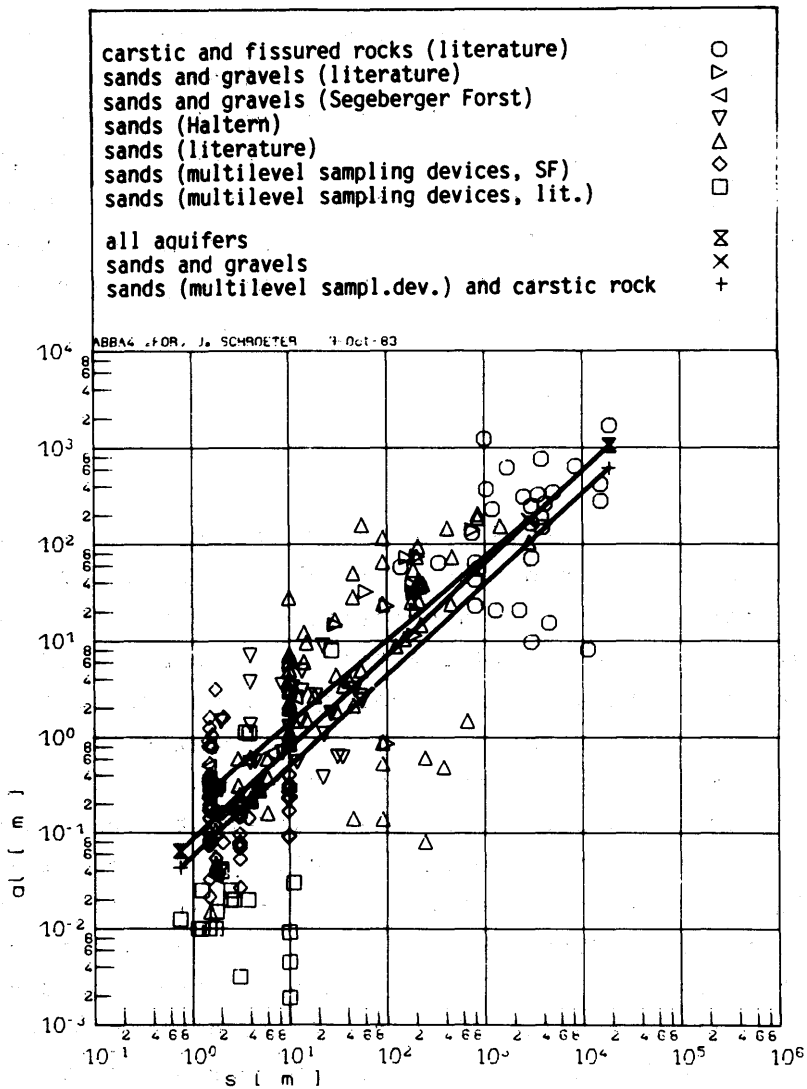


Fig. 1.14: Longitudinal dispersivity in dependance of flow distance (SCHRÖTER, 1983).

The influence of increasing porosity in comparable porous aquifers results in an increase of dispersivity. The long distance tracer tests were carried out mainly in karstic rocks with large-diameter solution channels. The short distance tests were performed in sandy aquifers, often with multi-level devices. The comparison of the results of tracer experiments with multi-level sampling devices on the one hand and karstic and fractured rocks on the other hand gives therefore a greater slope to the dispersivity/distance relationship.

Investigations, carried out in fluvioglacial gravels (BEHRENS & SEILER, 1982) and in sandy aquifers (PICKENS & GRISAK, 1981) lead to similar equations. The factors and exponents are listed in table 1.9. In these relations the exponent b depends on the horizontal inhomogeneity of the investigated aquifer, while the factor a is, as mentioned, a function of the textural characteristics and the vertical inhomogeneity of the sediment.

SCHRÖTER (1983) verified the results of field tracer experiments with a transport model simulating a multilayer groundwater flow, in which for each layer a dispersivity value, corresponding to laboratory measurements, and different measured K -values were inserted. The model also included mixing in the observation wells. The calculated results for velocities, dispersion coefficients and dispersivities were within 20 % of the results of the tracer experiment (Fig. 1.15).

The equation of MERCADO (1967) and the computer model of SCHRÖTER (1983) indicate an apparently unlimited increase of the dispersivity related to the flow distance. For example the, K -range dependent, dispersivity increase for

Tab. 1.9: Coefficients and exponents of the relation between dispersivity and flow distance following eq. (18)

general equations

	coefficient a	exponent b	n	Author
all aquifers (eq. 19a)	0.085 ± 0.016	0.96 ± 0.06	283	SCHRÖTER (1983)
porous media without multilevel sampling devices (eq. 19b)	0.199 ± 0.046	0.86 ± 0.08	131	
multilevel sampling devices and fissured and carstic rock (eq. 20)	0.057 ± 0.014	0.94 ± 0.08	152	

Special fields

	coefficient a	exponent b	
fluvioglacial gravels	0.29	0.72	BEHRENS & SEILER (1982)
sandy aquifer	0.1	1.0	GRISAK & PICKENS (1981)

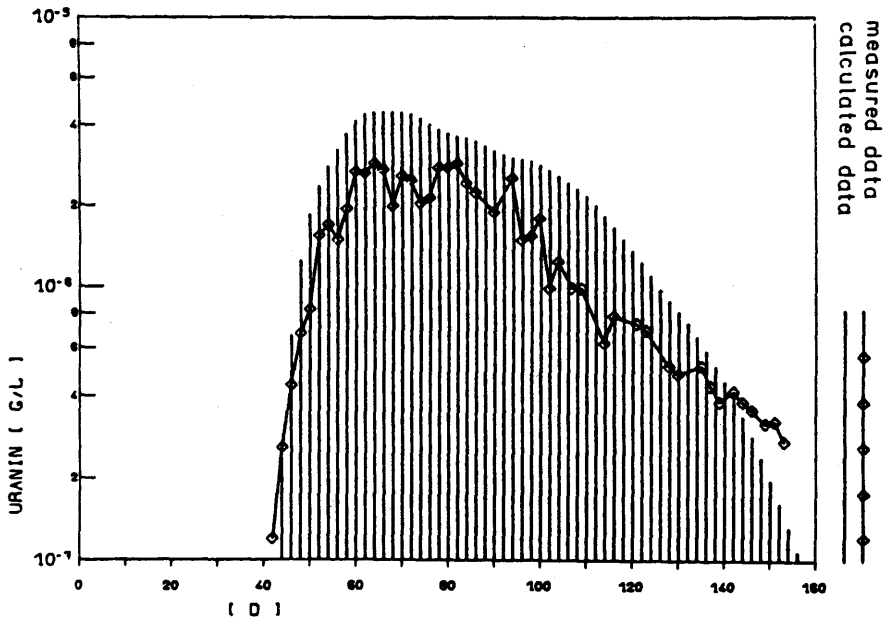


Fig. 1.15: Comparison of measured and calculated break-through curves in a flow distance of 55 m (SCHRÖTER, 1983).

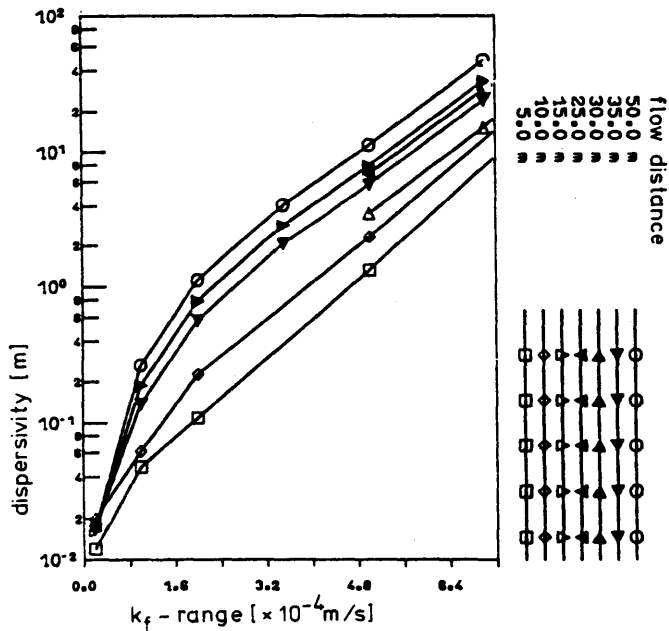


Fig. 1.16: Dependence of dispersivity in a layered model-aquifer with different K-values (mean 5×10^{-4} m/s) from K_{max} - K_{min} and from flow distance. The dispersivity of single layers = 0.01 m (SCHRÖTER, 1983).

different flow distances is shown in a tracer transport model for an ideal stratified aquifer, consisting of three layers with different K-value-ranges (mean $K = 5.0 \times 10^{-4}$ m/s) (Fig. 1.16). In reality the layers of a stratified sediment include horizontal inhomogeneities or geological interfaces, e.g. sedimentary structures like cross bedding. Within an observed range, large enough to smooth out textural and structural inhomogeneities, a statistical homogeneity or a homogeneous distribution of the anisotropies in the aquifer is reached. So the dispersivity value approaches a limit, which is considered to be characteristic for the investigated area (BEHRENS & SEILER, 1982; SCHRÖTER, 1983). Due to this boundary condition, it can be assumed that scale dependent homogeneity yields dispersivity values which characterize specific sediment types in large aquifers.

1.2.5. Gas Transport

The oxygen input and the CO_2 output between the atmosphere and the biologically active lithosphere ("soil respiration") is mainly controlled by gas diffusion (ALBERTSEN, 1979; HANNEN, 1892). The diffusion flux rates Q_1 can be calculated according to FICK's equation:

$$Q_1 = D_1^* \cdot \text{grad } C_1 \quad (1.32)$$

when the rock diffusivity D^* and the concentration gradient C_1 are measured. The rock diffusivity is affected by the porosity and the moisture content (ALBERTSEN, 1977; ALBERTSEN & MATTHESS, 1978):

$$D^* = f \cdot n_o \cdot D_o' \quad (1.33)$$

in which f is a correction factor to take account of the curvatures of the pore channels (tortuosity), n_o is the gas effective porosity and D_o' is the diffusivity of the dry rock. For sandy materials with natural water contents, ALBERTSEN (1977) indicates the empirical relation:

$$\frac{D'}{D_o'} = 0.78 \frac{\epsilon n}{\epsilon_t} - 0.27 \quad (1.34)$$

The increase in anthropogenic sources resulted in the recognition of gas exchange as a purification factor in the removal of highly volatile contaminants from the underground. Mineral oil products and halogenated hydrocarbons with high vapor pressure (Tab. 1.10) belong to the group of highly volatile environmental chemicals which often appear in cases of contamination (Fig. 1.17) (ALBERTSEN & MATTHESS, 1978; GOLWER, 1983).

Highly volatile substances, e.g. gasoline, benzene and volatile chlorinated hydrocarbons, may preferentially escape by diffusion from contaminated soil into the ground air and the free atmosphere (FRAGADIAKIS et al., 1979). In the area of Frankfurt (Main) airport, tetrachloroethene concentrations up to 16.4 mg/m^3 were registered in the ground air at a depth of 1 m, whereas the "zero value" for the ground air in this area lies in the order of 0.005 mg/m^3 (FRITSCHI et al., 1971). The increased values could be traced to tetrachloroethene contamination.

The volatility is a measure of the tendency of a substance to vaporize. It is the amount of vapor in a given air volume at saturation. However, the volatility of a substance does not appreciably interfere with its concentration in

Tab. 1.10: Vapour Pressure of chemicals (BROCK NEELY. 1980)

Chemical	Vapour Pressure (mm Hg)
Toluene	30
Tetrachloroethylene	14
p-Dichlorobenzene	1
Trichlorobenzene	0.5
Biphenyl	9.7×10^{-3}
Pentachlorobiphenyl	7.7×10^{-5}
Hexachlorobenzene	1.0×10^{-5}
DDT	10^{-7}

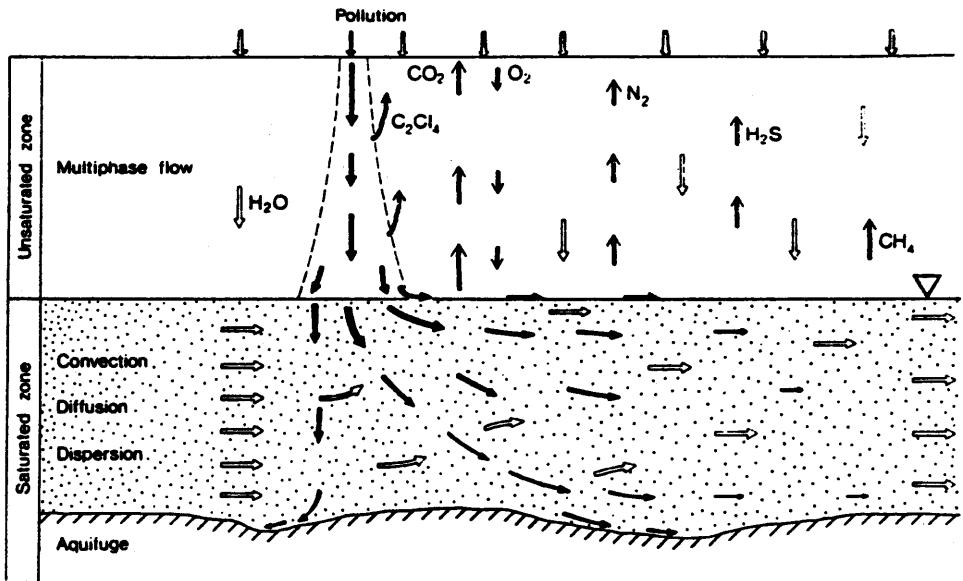


Fig. 1.17: Transport of volatile chemicals in the under-ground (GOLWER, 1983)

groundwater. Thus the elimination effect of high vapour pressure is often overestimated in groundwater (ZOETEMAN et al., 1981). Notwithstanding the slow gas emanation from groundwater into the unsaturated zone, this process may result in increased CO_2 and hydrocarbon contents in the ground air, which allow measurement of the extent of the contaminated zone (ALBERTSEN & MATTHESS, 1978).

1.2.6. Filtration

The removal of microorganisms or of other corpuscular contaminants from sub-surface water is a complex process. It includes the time dependent processes described above and filtration processes depending on the length of the transport path. The filtration efficiency, which is indicated by a decrease of the initial concentration C_o to the observed concentration C_x , can be described by a factor F (IWASAKI, 1937):

$$C_x = C_o \cdot e^{-\lambda F \cdot x} \quad (1.35)$$

Microorganism transport may be limited by the pore size of the sub-surface material and by the size of the microorganisms. However, mechanical filtration cannot be very effective in sandy and gravelly strata due to the small diameters of bacteria (0.2 - 5 μm) and viruses (0.02 - 0.25 μm) compared with the diameters of pores, which in uniform sands are generally more than 40 μm . In natural sediments with heterogeneous grain-size distribution some proportion of the pore diameters can interfere with bacteria transport (3 - 10 %).

Although the measurement of filter factors in the field is extremely difficult, because of the other aforementioned processes which also affect microbial transport capacity,

some calculated filter factors and the travelling distances are given in table 1.11. The filter factors are much higher in the unsaturated zone (> 10x) than in groundwater.

The particle accumulation on solid substance surfaces is mainly affected by sedimentation, flow processes, diffusion and interception (Fig. 1.18). YAO et al. (1971) give the following equations (1.36-1.40):

$$\ln \frac{C}{C_0} = - \frac{3}{2} \cdot (1-n) \alpha \eta \left(\frac{x}{d} \right) \quad (1.36)$$

$$\eta = \eta_D + \eta_I + \eta_G \quad (1.37)$$

$$\eta_D = 4.04 \text{ Pe}^{-2/3} = 0.9 \left(\frac{kT}{\mu d p v_f} \right)^{2/3} \quad (1.38)$$

$$\eta_I = \frac{3}{2} \left(\frac{dp}{d} \right)^2 \quad (1.39)$$

$$\eta_G = \frac{\rho_p - \rho}{18 \mu v_f} \quad (1.40)$$

with C_0 , C = influent and effluent concentration

n = porosity

α = stabilisation coefficient

η = single collector efficiency

d = grain size of filter material (m)

d_p = diameter of suspended particles (m)

L = bed depth (m)

η_D , η_I , η_G = single collector efficiencies for diffusion, interception, sedimentation

Pe = PECLET number = $v_a d / D$

k = BOLTZMANN constant ($1.38054 \cdot 10^{-23} \text{ J/}^{\circ}\text{K}$)

T = absolute temperature ($^{\circ}\text{K}$)

μ = water viscosity (N.s/m^2)

v_f = filter velocity (m/s)

ρ_p , ρ = density of suspended particles and water (t/m^3)

g = gravity constant (0.80665 m/s^2)

Tab. 1 11: The calculated filter factors (λ_F) in field studies and estimated travelling distances for 99.9 % filtration in different aquifers.

	λ_F (1/m)	S(m)
gravel	~ 0.01	~ 100
sand, coarse	~ 0.1	~ 10
sand, fine	~ 1.0	~ 1

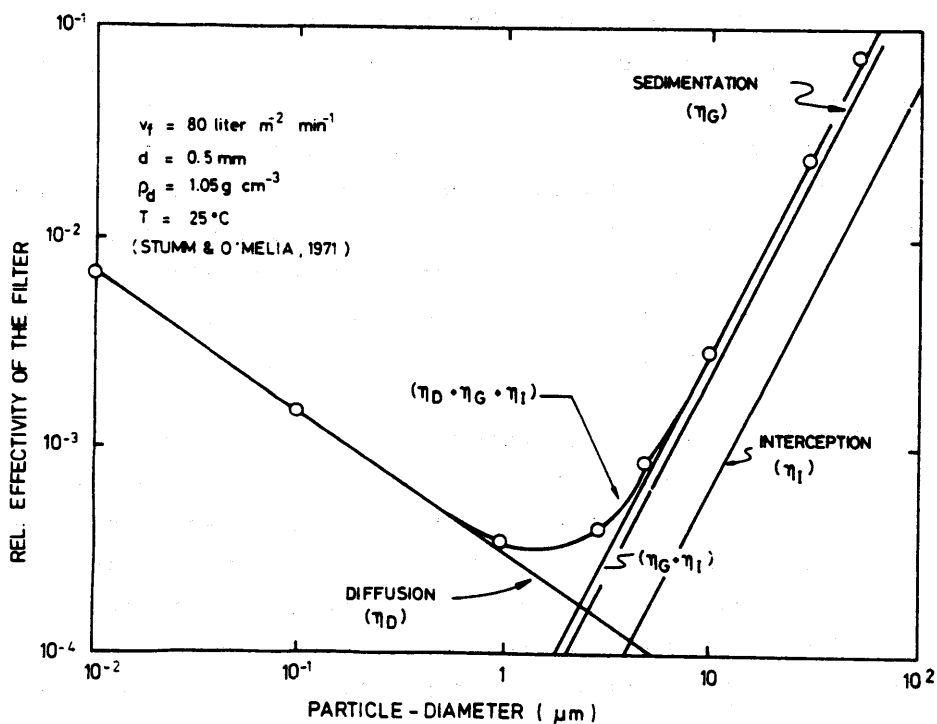


Fig. 1.18: Filtration parameters in porous media (YAO et al., 1971).

The comparison of filtration efficiency of clay minerals ($< 2 \mu\text{m}$ fraction, $\rho_p = 2.7 \text{ g/cm}^3$) with that of bacteria ($\rho_p \approx 1 \text{ g/cm}^3$) show that, the bacteria are eliminated more effectively than clay minerals. The filtration efficiency for clay minerals predicted from eqns. (1.35)-(1.39) agrees reasonably with experimental results. The reason for the deviation of bacteria removal from theory is presumably the active mobility of bacteria ($\approx 0.1 - 1 \text{ m/d}$). The effective diameter of a bacterium is also greater than $1 \mu\text{m}$ due to its irregular shape and the presence of filaments on its surface.

The intensity of bacteria and virus attachment to the underground solid materials depends on the adsorption mechanism. The generally negatively-charged bacteria and viruses are strongly adsorbed by anionic adsorbents and only slightly by cationic adsorbents. The solid substances of the underground are generally negatively charged. Exceptions are the iron and manganese hydroxides and humic substances at low pH values. The negatively charged microorganisms stay in suspension at high pH values (Fig. 1.19), as the repulsive electrostatic forces are stronger than the VAN DER WAALS forces.

The dissolved cations in water decrease the repulsive forces of the grain surfaces. Monovalent cations are adsorbed by the solid substance and decrease their charge deficiency. Bivalent cations can also cause a positive charge deficiency so that the electrostatic forces can be more efficient for bacteria and virus adsorption. Under these conditions, the mass forces are more effective and an accumulation of particles can take place. This can be demonstrated by the dependence of virus adsorption on the solute concentration of the water mentioned above (Fig. 1.11).

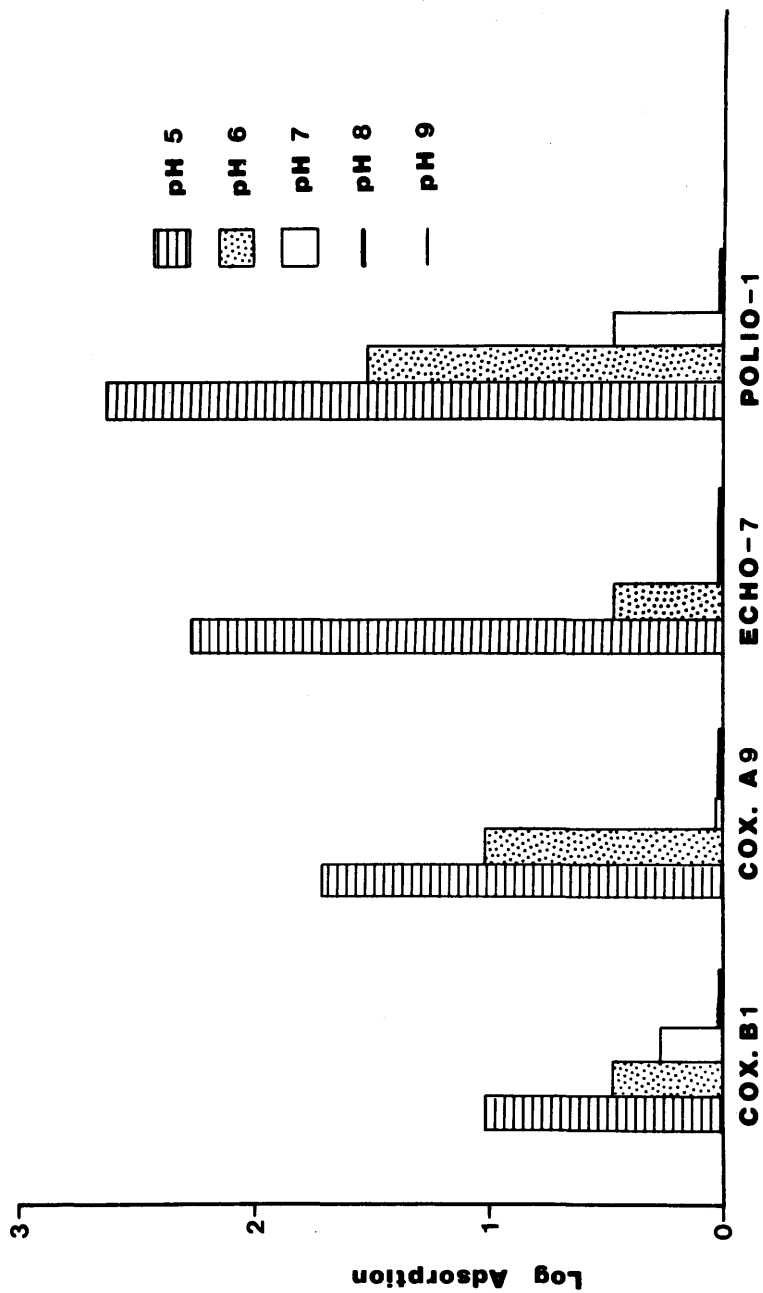


Fig. 1.19: Adsorption of viruses at different pH-values
(DIZER et al., 1983).

The duration of the contaminating process and the initial concentration of contaminants are very important in filtration efficiency. During a continuous contamination process by organic substances and microorganisms the contaminated plume becomes smaller with time because elimination and filtration mechanisms are favoured. At very high initial concentrations, flocculation and aggregation can occur at the source of contamination so that only limited transport into the aquifer can take place.

1.2.7. Movement of Immiscible Organic Fluids

Organic fluids may seep into the ground as a result of leaking storage tanks, or when accidentally spilled. If the volume of liquid is sufficiently large, the contaminants can migrate down to the water table and affect the quality of groundwater. Fluids with high densities (e.g. most chlorinated hydrocarbons) may also infiltrate from leaking sewers (NEUMAYR, 1981), from which they may escape through the finest leaks due to their hydraulic properties. The hydraulic conductivity for any fluid (K_{fl}) depends from its specific weight (γ_{fl}), its dynamic viscosity (η_{fl}) and the permeability of the aquifer (k) (JACKSON, 1980).

$$K_{fl} = \frac{\gamma_{fl}}{\eta_{fl}} k \quad (1.41)$$

From this equation it follows that substances such as gasoline, benzene and most of the volatile chlorinated hydrocarbons will be more mobile than water in a given aquifer, whereas diesel fuel and heating oil will move slower than water. This can be shown by the ratio of the permeability of the organic fluid with respect to that of water (Tab. 1.12). However, in a porous aquifer the relative permeabilities of the organic phases are usually higher than that

Tab. 1.12: Density . kinematic viscosity at 20°C and ratio K_{fl}/K_w

	g/cm^3	mPa . s	K_{fl}/K_w
Water	0.9982	1.0050	1
<u>Hydrocarbons</u>			
Benzene	0.879	0.652	1.36
Gasoline	0.725-0.785	0.65	1.54
Diesel fuel	0.82 -0.86	2.80-6.40	0.15-0.36
<u>Halogenated hydrocarbons</u>			
Dichloromethane	1.327	0.3282	3.0675
Trichloromethane	1.462	0.3765	2.6738
1.1.1. Trichloroethane	1.337	0.65	1.8577
Dichlorobenzene	1.306	0.8159	1.2340
Tetrachloroethane	1.598	1.0951	0.9194

derived from eqn. (1.11) due to the fact that they are moving in the bigger pore channels, whereas the more wettable water phase is filling the smaller pores (JACKSON, 1980).

Organic fluids which are lighter than water (Tab. 1.12) may form impregnation bodies on the groundwater surface and may spread on the groundwater surface following the hydraulic gradient (Fig. 1.20). The chlorinated hydrocarbons which are mostly heavier than water (Tab. 1.9) may sink to the base of the aquifer and form a more-or-less flat impregnation body. This body will move following pressure gradients in the body and the morphology of the base of the aquifer (SCHWILLE, 1981). When infiltration is finished, the impregnation body will spread until saturation is reached or until the fluid is concentrated in a depression. At saturation the immiscible fluid is distributed in numerous isolated droplets, which are dissolved in the percolating water with time. Thus downstream of an impregnation body a contaminated plume is formed, the initial concentration of which is controlled by the solubility of the respective fluid. The laws of the hydrodynamic dispersion described above hold for the transport of the dissolved substances (SCHWILLE, 1981).

1.3. Discussion

The transport of contaminants in the underground must be studied in laboratory and field experiments by comparing their behaviour with that of ideal nonreactive tracers.

Field studies give more realistic values, but they do not allow the quantification of controlling factors, because the physical, chemical and biological processes take place

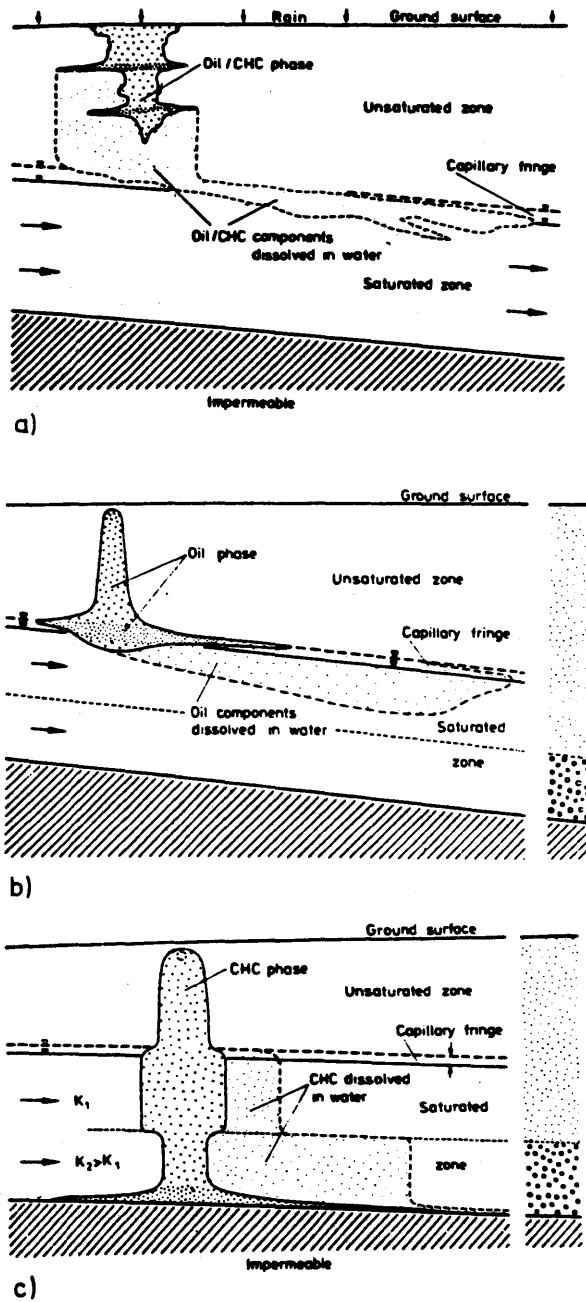


Fig. 1.20: Migration pattern of immiscible fluids (SCHWILLE, 1981).

- a Oil/Chlorohydrocarbon
- b Oil
- c Chlorohydrocarbon

simultaneously during the transport of contaminants in groundwater. Laboratory tests can be carried out under closely defined conditions to aid the understanding of the importance of individual factors, affecting the transport of contaminants. The number of field studies possible is restricted by their high financial and time demands. Laboratory studies can be used to test a large number of different geological materials and groundwater types. A combination of both methods is optimal.

As an example, the transport behaviour of ^{103}Ru is shown (Fig. 1.21) in different soil types. The processes described lead to different distribution patterns under identical conditions of precipitation, seepage velocity etc. (ALBERTSEN & MATTHESS, 1980). In this case different retardation coefficients, which are the most important factors for heavy metal transport, lead to different contaminant transport velocities.

In figure 1.22 the transport behaviour, dispersivities and retardation factors of E.coli and ^{82}Br are shown, which were calculated on the basis of data from BETZ et al. (1983).

The transport velocities of Lindane and Simazine are slower than the "ideal" tracer lithium (Fig. 1.23). The elongated breakthrough curves and the calculated dispersion parameters of these two pesticides show time-dependent irreversible adsorption processes. Lindane is more strongly adsorbed than Simazine and therefore measured in lower concentrations. In the case of Dicrotophos the high volatility of this compound leads to a very sharp breakthrough curve although the measured concentrations are low.

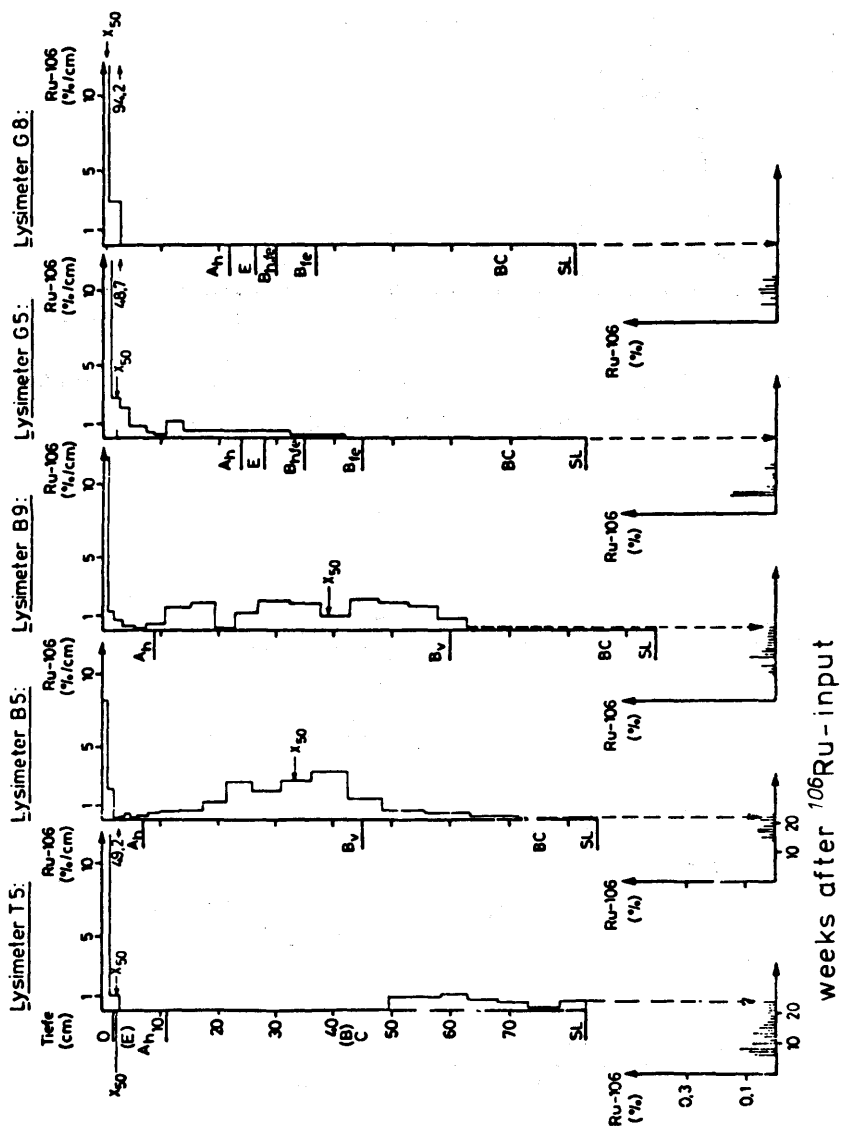


Fig. 1.21: Distribution- and transport-pattern of ^{106}Ru in different soil columns (ALBERTSEN, 1978).

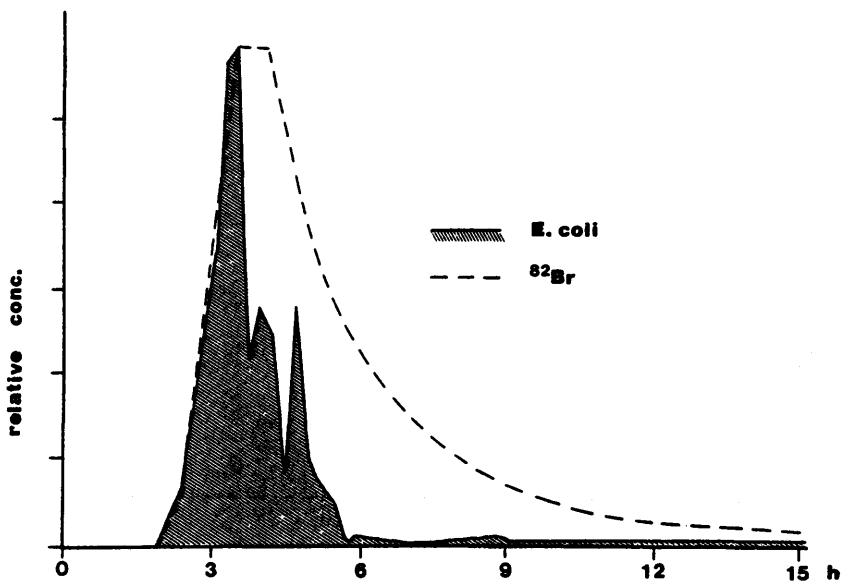


Fig. 1.22: Breakthrough curves of *E.coli* and ^{82}Br in coarse gravel in a flow distance of 20 m (ALEXANDER & SEILER, 1983).

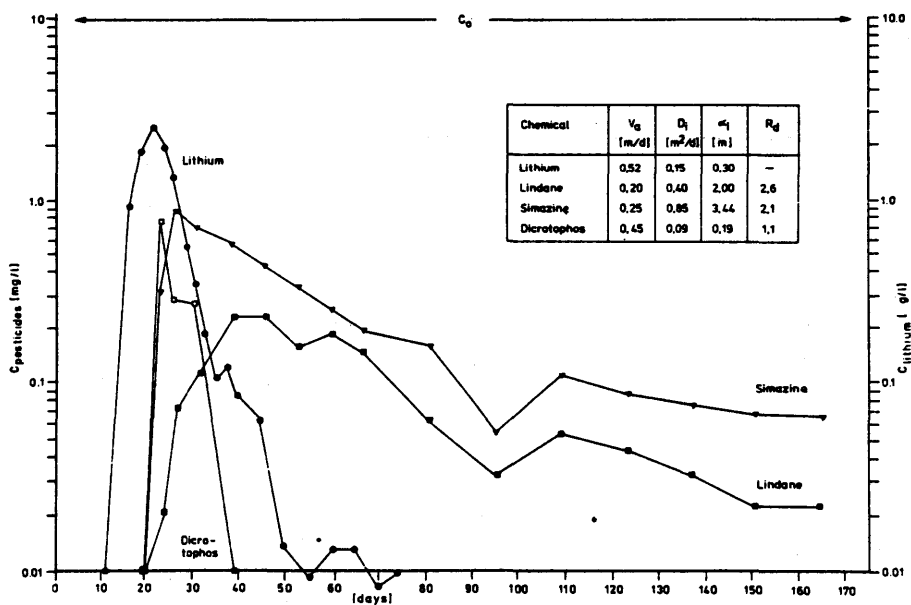


Fig. 1.23: Breakthrough curves of lithium, as an "ideal tracer", Simazin, Lindane and Dicrotophos in a sandy aquifer in a flow distance of 11.7 m (calculated after data from BETZ et al., 1983).

A similar phenomenon, in which part of the contaminant is irreversibly eliminated, can be seen in the case of bacteria transport. Due to the filtration processes the bacteria are transported mainly in the larger pores, which causes a sharp break-through curve even after long traveling distances (ALEXANDER & SEILER, 1983; HAVEMEISTER et al., 1983). However, the lateral dispersion of bacteria is greater than that of an conservative tracer. ALEXANDER & SEILER (1983) reported a broader lateral distribution of *E.coli* compared with the conservative tracers, Bromine-82 and fluoresceine, probably due to the active mobility of bacteria.

The breakthrough curves of conservative tracers used simultaneously are broader, since in their case the tracer also travels in the smaller pores with low velocities (Fig. 1.23). In layered aquifers high dispersion coefficients for the fluorescent tracer pyranine are measured depending on the different transport velocities and retardation values of each layer. Contrarily bacteria are transported mainly in the layers with higher conductivity and porosity and therefore show a very sharp breakthrough curve and lower dispersion coefficients (HAVEMEISTER et al., 1983; Fig. 1.24).

These few examples demonstrate, that knowledge of the behaviour of contaminants is necessary for the assessment of groundwater protection areas.

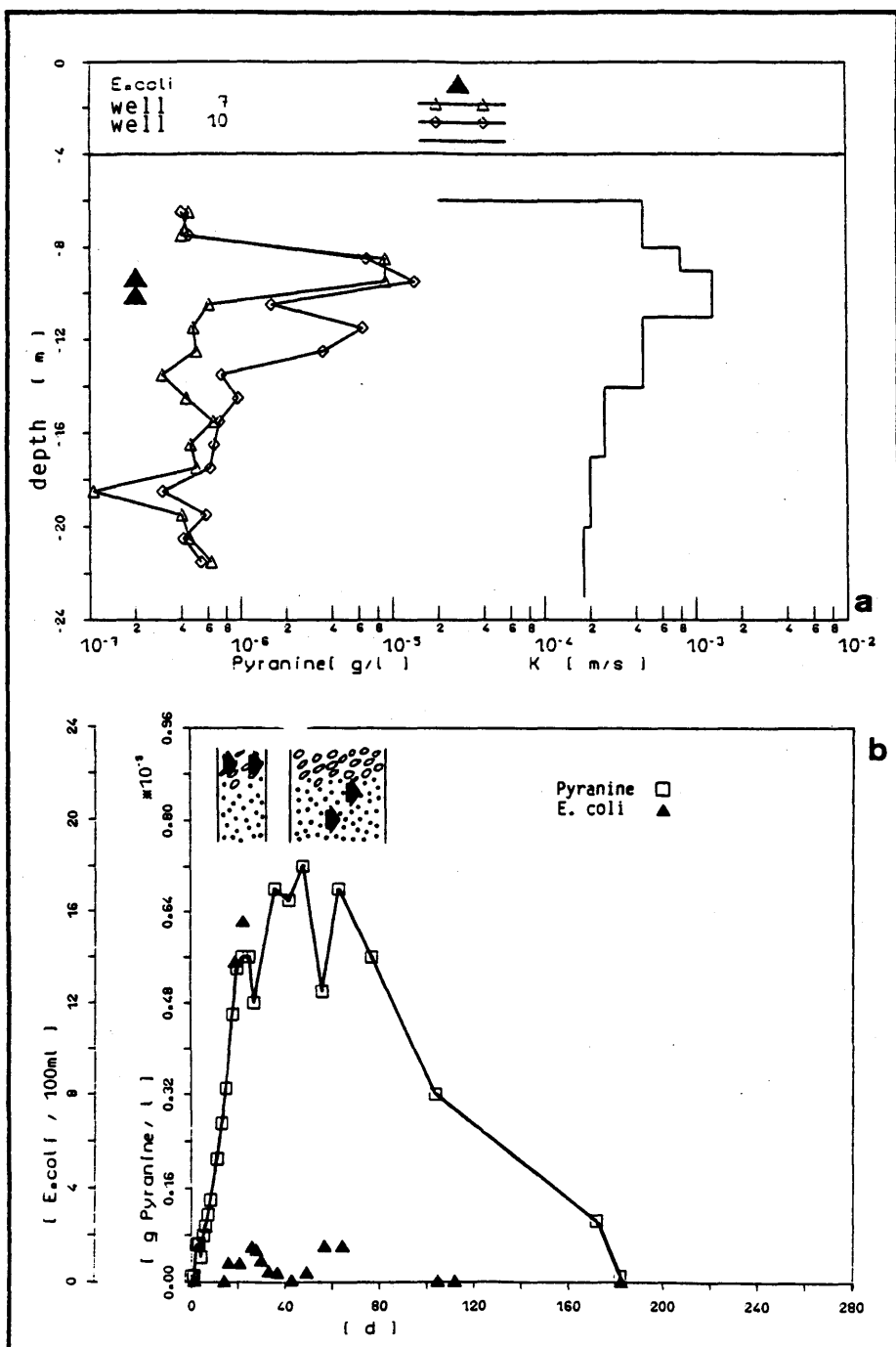


Fig. 1.24: Breakthrough curve of Pyranine compared with E.coli.

1.4. References

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CHAPTER 2

NOTIONS D'HYDRAULIQUE ET MÉCANISMES HYDRODYNAMIQUES DE PROPAGATION EN MILIEU POREUX SATURÉ.

par

L.Zilliox

2.1. Situation du Problème

La délimitation de zones de protection par le tracé de "périmètres" autour d'un captage d'eau en nappe alluviale est fondée sur l'application d'équations générales de l'hydrodynamique.

Permettant d'établir un classement des dangers à éviter en fonction de leur importance et de leur incidence, non seulement sur la qualité bactériologique mais encore sur la qualité chimique de l'eau, ces périmètres diminuent les risques de pollution mais ne les suppriment pas. Ceci est admis depuis des décennies (BUYDENS, 1962). Des travaux plus récents ont montré comment l'effet de l'hétérogénéité d'un aquifère va consister à accroître considérablement la distribution des temps de transfert d'un polluant. Alors la notion de transmissivité de l'aquifère, qui a rendu de nombreux services en hydrogéologie, paraît difficilement extrapolable au transport de matière (cf. BRISSAUD & DE MARSILY, 1977). Il en résulte que la protection des eaux souterraines nécessite une connaissance, la plus complète possible, des phénomènes de transport miscible en milieu poreux: l'intérêt portant sur la qualité de l'eau, il n'est plus suffisant de considérer l'aptitude de l'aquifère à transmettre un flux d'eau, mais il faut tenir compte de la capacité de l'aquifère à sélectionner les éléments transportés par l'eau et à les disperser.

2.2. Fonctionnement Hydraulique d'un Milieu Poreux et Echelle d'Etude

1) Pour étudier l'écoulement de l'eau à travers le système complexe formé par les interstices interconnectés d'un milieu poreux (aquifère alluvial par exemple), l'expérimentateur peut se placer à une échelle de loin supérieure à celle des pores pour considérer le réservoir perméable dans son ensemble.

Si l'objectif est de connaître les phénomènes se déroulant à l'échelle du réservoir lors de l'étude du mouvement d'une nappe d'eau souterraine libre défini par le champ de la charge hydraulique, le milieu sera considéré comme un milieu continu: on pourra appliquer alors la théorie du mouvement potentiel avec tous les moyens de résolution mathématique, numérique, analogique.

Dans le cas des mouvements à potentiel on peut appliquer le "principe de superposition": si on connaît dans le plan (x, y) le mouvement autour d'un puits (Fig. 2.1) et celui d'un courant rectiligne (Fig. 2.2) on détermine, par superposition, les caractéristiques de l'écoulement composé d'un puits placé dans le courant rectiligne (Fig. 2.3).

En appelant V, vitesse amont uniforme du courant rectiligne et Q, débit du puits, les équations des lignes de courant, $\psi = \text{const.}$ du mouvement résultant, sont obtenues par addition des fonctions de courant $\psi_c = -Vy + \text{const.}$ (courant rectiligne) et $\psi_p = \frac{Q}{2\pi} \theta + \text{const.}$ (puits), soit $\psi_c + \psi_p = \psi = -Vy + \frac{Q}{2\pi} \theta + \text{const.}$, ($\theta \equiv \text{arc. tg } \frac{y}{x}$).

La figure 2.3 est utilisée dans la définition du périmètre

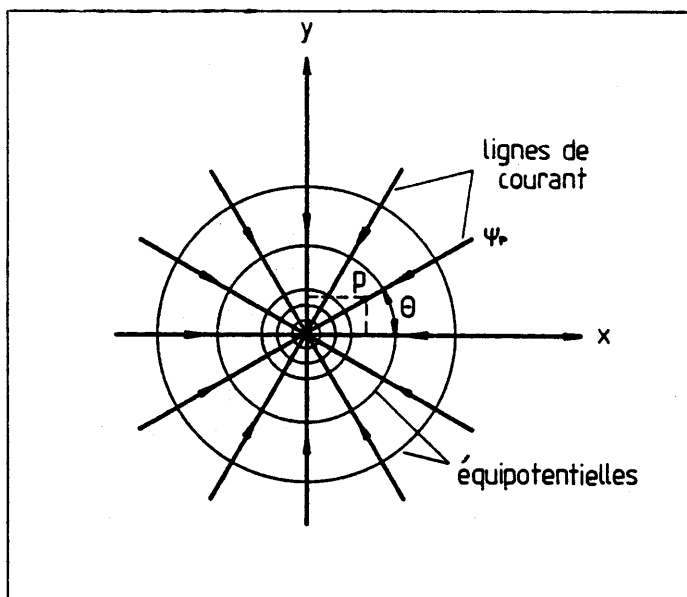


Fig. 2.1.: Ecoulement vers un puits

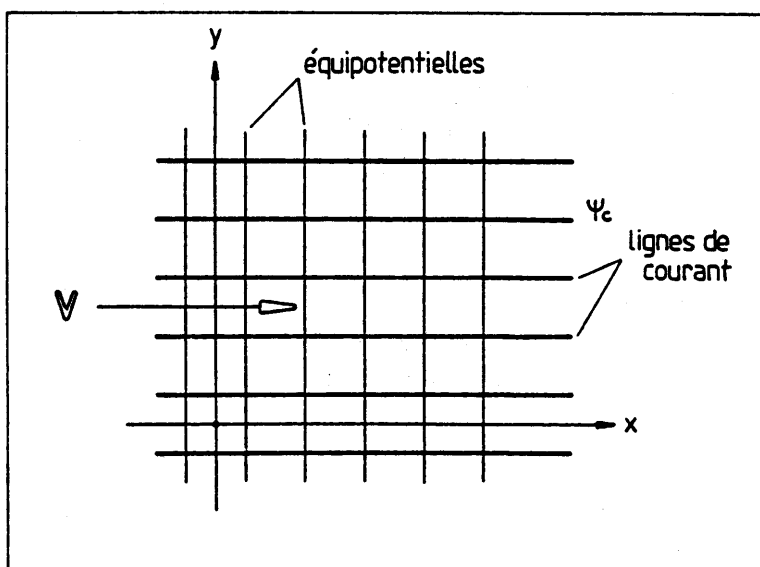


Fig. 2.2.: Ecoulement rectiligne

de protection qui prend en compte notamment la ligne de partage ψ_0 , correspondant à la limite d'influence du puits.

2) Dans de nombreux cas cependant, le comportement du ou des fluides contenus dans le réservoir - il s'agit d'eaux contaminées soit physiquement (influences thermiques), soit chimiquement (substances dissoutes), soit biologiquement (microorganismes) - résulte de mécanismes mis en jeu à l'échelle des pores et il est nécessaire d'acquérir tout d'abord une bonne connaissance de ces phénomènes qui auront une répercussion à l'échelle du réservoir entier.

Entre les échelles globale - niveau du réservoir - et ponctuelle - niveau du pore - a été introduite l'échelle locale, niveau intermédiaire du "volume élémentaire représentatif" (noté VER et désigné aussi par "bloc élémentaire de milieu poreux"). A cette échelle l'objectif est d'établir des lois dites "macroscopiques" par des prises de moyennes (au sens des probabilités) sur les lois élémentaires de l'hydrodynamique (établies en mécanique des fluides en milieu continu) connues par exemple dans un pore (pris comme canal unitaire d'écoulement). En fait, on passe ainsi d'une caractérisation géométrique détaillée et délicate (voire inutile souvent) des pores au VER, bloc suffisamment grand, pour y inclure un nombre important d'espaces vides et de pleins. de sorte qu'un effet de moyenne s'y manifeste; on pourra y raisonner comme dans un milieu continu et y définir localement des "grandeurs macroscopiques".

3) La loi de DARCY est l'équation du mouvement d'un fluide à l'échelle du VER où les singularités du niveau granulométrique (échelle des pores) ne sont plus prises en compte; dans l'hypothèse d'un milieu continu elle exprime simplement la relation entre la vitesse fictive "macroscopique" (débit unitaire q) et le gradient hydraulique J par $q = KJ$,

avec K = coefficient de DARCY (ou coefficient de perméabilité, ou conductivité hydraulique).

4) Remarque: L'échelle du VER est généralement celle de l'échantillon de milieu poreux en laboratoire. Mais toute partie d'aquifère naturel présente des hétérogénéités dont les dimensions vont de celle des espaces vides (existant aussi dans le VER) à celle de la distance par exemple entre un point d'alimentation et un point de captage, ou encore de la distance entre puits d'injection et piézomètre de repérage lors d'un essai de traçage in situ. Dans ces cas, vue l'extension du champ, il sera nécessaire de définir des paramètres caractéristiques à cette échelle supérieure. Ce ne seront plus ceux localisés d'un VER, mais ceux qui intégreront les "variations régionales" et devront être déterminés par des méthodes statistiques.

2.3. Paramètres importants caractérisant un milieu poreux (de type aquifère alluvial)

1) Dans la mise en valeur d'un aquifère, il est nécessaire de mettre en parallèle deux préoccupations: celle relative à l'étude quantitative de la ressource EAU, où l'on traitera du mouvement de masses d'eau sans distinction de composition, et celle relative à la qualité de la ressource. Dans le premier cas, on évoquera la capacité du réservoir à contenir de l'eau et son aptitude à transmettre un flux d'eau, dans le second cas, la capacité du réservoir à sélectionner des éléments transportés par l'eau et de son aptitude à disperser ces éléments.

Dans cette optique, trois paramètres du réservoir sont à mettre en avant: la porosité, la perméabilité et la dispersivité. Leur

importance se situe respectivement au niveau du c o n t e -
n u (stock d'eau), du f l u x (circulation de l'eau), de
la p r o p a g a t i o n (transport par l'eau).

Limitée à des considérations au voisinage d'un captage
d'eau, la réflexion sur une mise en parallèle des aspects
Quantité et **Qualité** de la ressource **Eau** en relation avec les
paramètres de l'aquifère a conduit à la synthèse présentée
dans le tableau 2.1.

2) Commentaires sur les paramètres

Porosité

Lors d'essais sur échantillon de milieu poreux, le volume
d'eau gravitaire recueilli rapporté au volume total de
l'échantillon indique la valeur de sa porosité efficace.
Alors que cette porosité efficace s'obtient comme un
rapport de deux volumes, la porosité cinématique s'obtient
par le rapport de deux vitesses: vitesse de DARCY/vitesse
macroscopique moyenne. Cette porosité cinématique est-elle
fonction du gradient de charge hydraulique? Il n'est guère
de mesure connue faite pour répondre à cette question.

N'est-il pas risqué de vouloir déterminer cette porosité
cinématique par traçage in situ au niveau d'une portion de
réservoir, sachant que les hétérogénéités feront assimiler
la vitesse macroscopique moyenne à la vitesse correspondant
au transit par les zones les plus perméables? L'essai de
traçage serait plus pertinent pour reconnaître l'existence
de cheminements préférentiels que pour traduire ces chemine-
ments par une porosité dite cinématique, sans commune mesure
avec la valeur de la porosité totale du milieu.

Tab. 2.1: Essai de mise en parallèle QUANTITE - QUALITE de la ressource EAU en relation avec les paramètres de l'aquifère.

Exploitation de la ressource EAU		Aspect QUANTITE	Aspect QUALITE
		Exemple d'évaluation de la ressource : <u>rendement d'un puits de captage</u> , une question de DEBIT TOTAL d'exploitation.	Exemple de protection de la ressource : <u>zones de protection du captage</u> , une question de VITESSE LOCALE de propagation.
Concepts et méthodes		On pense TRANSFERT en masse d'eau au sens de l'approche hydro-dynamique ; On se fonde sur la loi de filtration (de Darcy) ; On prend en compte la notion de transmissivité (de PERMEABILITE moyenne et de puissance de l'aquifère) ; On donne une image par un réseau global de lignes de courant et d'équipotentiellles.	On pense TRANSPORT de soluté au sens des approches hydrodispersive et hydrochimique ; On se fonde sur la loi de conservation de la masse de soluté ; On prend en compte le champ des vitesses, les effets de mélange et d'interaction (DISPERSION et échanges) ; On donne une image par la répartition spatio-temporelle des concentrations (évolution de courbes iso-concentrations).
Objectif pratique		On s'intéresse aux capacités productrices et à la réserve disponible (bilan)	On s'intéresse aux trajets et durées de parcours, aux concentrations en éléments (valeurs critiques, normes)
Paramètres significatifs concernant principalement :	Contenu	POROSITE efficace (teneur en eau drainable)	POROSITE cinématique (teneur en eau mobile)
	Flux	PERMEABILITE moyenne (obtenue à partir de la transmissivité du réservoir)	PERMEABILITES locales et leur distribution dans l'espace (la transmissivité n'a plus de sens ici ; l'hétérogénéité de perméabilité est à prendre en compte)
	Propagation	(aucun paramètre car la propagation n'est pas dissociée du flux)	DISPERSIVITE, paramètre intrinsèque d'aptitude du milieu à déterminer la dispersion de fluides de composition non uniforme
	NB : Le paramètre de SURFACE SPECIFIQUE, défini comme rapport de la surface totale des vides intersticiels au volume total du milieu, intervient au niveau des échanges entre fluides et solides, agit sur les vitesses de propagation, "dépasse" les critères hydrauliques, pose des difficultés dans son appréhension quantitative à l'échelle d'un aquifère (il est spécifique à l'échelle d'un matériau donné).		

Perméabilité

Rappelons que le coefficient de perméabilité K (de dimension LT^{-1}) dépend non seulement des caractéristiques sédimentologiques du réservoir mais encore des caractéristiques physiques du fluide en circulation. On introduit ainsi la perméabilité k (grandeur spécifique au milieu poreux, de dimension L^2 , encore appelée perméabilité intrinsèque) liée à K par la relation $K = k \rho g / \mu$, où g est l'accélération de la pesanteur, ρ et μ étant respectivement la masse volumique et la viscosité dynamique du fluide. Dans les milieux alluviaux (empilement de sédiments) on note toujours des valeurs de la perméabilité horizontale bien supérieures à celles de la perméabilité verticale (en fait dans un tel milieu la perméabilité est à considérer en général comme une propriété tensorielle).

La connaissance des valeurs de la perméabilité en différents lieux et à différentes cotes d'un réservoir permet d'en apprécier l'hétérogénéité. Il n'est cependant pas sûr que l'on puisse toujours tirer parti de cette connaissance pour prévoir le comportement global du réservoir.

La composition des perméabilités locales pour déterminer une perméabilité régionale qui traduit "en moyenne" la distribution des hétérogénéités d'un aquifère, ne s'opère rigoureusement que dans les cas simples du réservoir stratifié à succession de couches parallèles ou perpendiculaires à l'écoulement d'eau.

Cette situation est certainement à l'origine de la mise en oeuvre croissante de démarches stochastiques à côté d'approches déterministes; courante en hydrologie, cette méthodologie se développe en hydrogéologie pour prendre en compte la variabilité spatiale aléatoire de paramètres ca-

ractéristiques. La même analyse sur l'hétérogénéité d'un réservoir que celle effectuée à partir de perméabilités locales et régionales, se retrouve dans la caractérisation des propriétés dispersives du réservoir.

Dispersivité

Dans un réservoir aquifère de type alluvial, le transport d'un traceur (substance miscible à l'eau sans en modifier les caractéristiques physiques et non interactive avec les alluvions) d'un puits d'émission à un puits de repérage permet d'accéder à un coefficient de dispersion à partir de la courbe de restitution donnant la répartition des concentrations en traceur dans le temps au puits de mesure (Les méthodes pratiquées se trouvent dans une littérature abondante sur les sujet).

Pris selon la direction de l'écoulement moyen de l'eau souterraine, ce coefficient, noté D_L et appelé coefficient de dispersion longitudinal (de dimension $L^2 T^{-1}$), s'exprime en général par la relation $D_L = \alpha_L u^m$, où u est la vitesse macroscopique moyenne de l'écoulement (rapport du débit unitaire à la porosité cinématique du milieu), où m est une constante prise égale à 1 dans les conditions de circulation de l'eau dans un aquifère alluvial et où α_L est un coefficient de dispersion longitudinal intrinsèque appelé dispersivité longitudinale.

R e m a r q u e : On introduit de la même façon D_T et α_T , paramètres similaires correspondant aux effets dispersifs, transversaux (dans un plan perpendiculaire à la direction moyenne de l'écoulement dans le réservoir).

Tout comme le paramètre perméabilité, la dispersivité est représentative de l'échelle de mesure: elle correspond, au

niveau local, à la variabilité des vitesses dans l'espace poreux et, au niveau régional, aux hétérogénéités du champ des vitesses macroscopiques représentatif de la nature sédimentologique du réservoir. C'est le paramètre dont la détermination sur le terrain est la plus délicate.

3) La connaissance globale de l'aquifère, la détermination de paramètres sur certains sites, ne lèvent pas les incertitudes quant à la variabilité des mesures dans l'espace. Cette variabilité se manifeste sur la figure 2.4 où sont reproduits les résultats de mesures des vitesses locales sur un site de l'aquifère rhénan par mise en oeuvre de la méthode de dilution ponctuelle. Ces résultats sont le reflet de la nature et du degré d'hétérogénéité de l'aquifère.

Il faut d'ailleurs élargir dans ce contexte la notion d'hétérogénéité au-delà des effets dus à la nature sédimentologique du réservoir (créatrice de cheminements préférentiels comme de zones à diffusion retardée par exemple); il s'agit de prendre en considération l'influence de zones à saturations différentielles (effets capillaires en écoulement polyphasique, piégeage d'eau en aquifère aéré notamment) ou encore l'incidence de contrastes de densité (à l'origine du développement d'instabilités perturbatrices dans les déplacements miscibles par exemple).

2.4. Connaissance de Mécanismes Hydrodynamiques de Propagation en Milieu Poreux Saturé

1) Le transport d'un soluté (contaminant de l'eau) en milieu poreux saturé, homogène et isotrope, est régi par une équation de conservation de la masse qui exprime que le flux de soluté est affecté

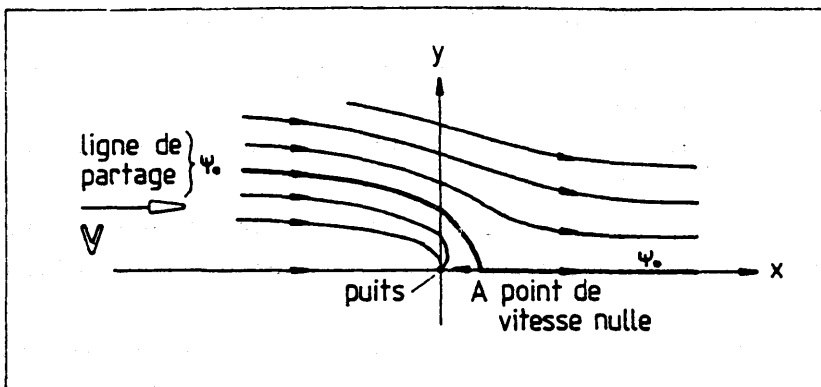


Fig. 2.3.: Puits dans un écoulement rectiligne (symétrie/Ox)

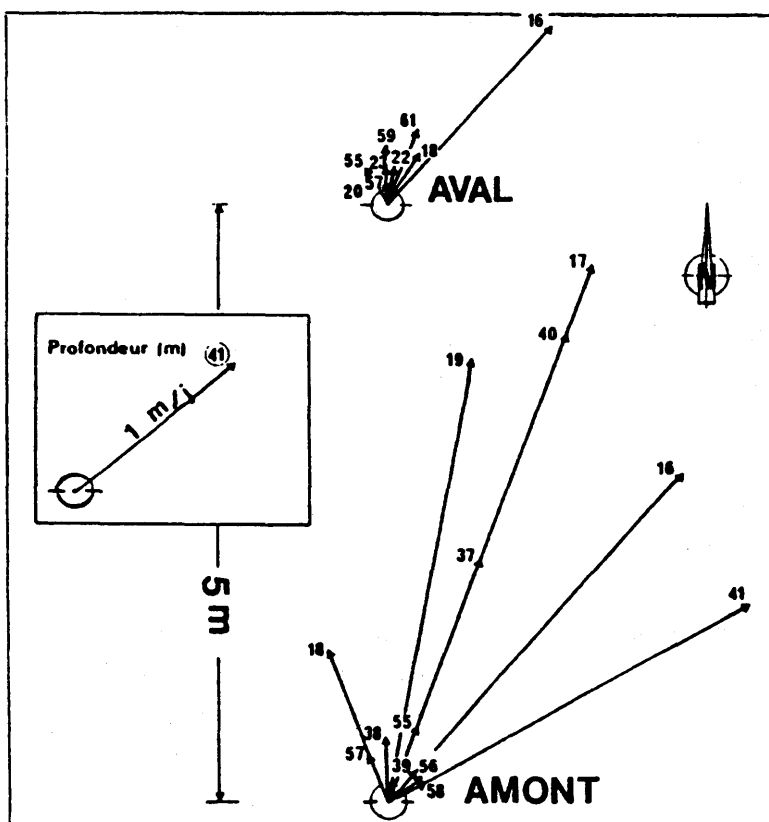


Fig. 2.4.: Distribution verticale des vitesses dans la nappe phréatique rhénane. Périmètre de Munwiller - Alsace (d'après IMF - Strasbourg, 1979)

- 1) par le mouvement convectif (déplacement moyen)
- 2) par la dispersion apparente (diffusion moléculaire et dispersion mécanique)
- 3) par les réactions géochimiques (échanges d'ions, physi- et chimie-sorption...)
- 4) par les processus biochimiques, radiologiques (dégradation biologique, décroissance radioactive éventuelle).

Cette équation s'écrit dans le cas monodimensionnel, compte tenu des mécanismes 1), 2), 3) et 4), sous la forme ci-apres

$$\underbrace{\frac{\rho}{n} \frac{\delta S}{\delta t}}_{(3)} + \frac{\delta C}{\delta t} = D_a \underbrace{\frac{\delta^2 C}{\delta x^2}}_{(2)} - \underbrace{\frac{q}{n} \frac{\delta C}{\delta x}}_{(1)} - \underbrace{\lambda C - \frac{\lambda \rho S}{n}}_{(4)} \quad (2.1)$$

où

C = concentration en soluté $[ML^{-3}]$

t = temps $[T]$

x = coordonnée selon la direction de l'écoulement $[L]$

S = quantité retenue sur le milieu poreux $[-]$

ρ = masse volumique apparente du milieu poreux $[ML^{-3}]$

λ = constante de dégradation $[T^{-1}]$

n = porosité cinématique $[-]$

D_a = coefficient de dispersion apparente $[L^2 T^{-1}]$

q = débit unitaire $[L T^{-1}]$

Les mécanismes présentés dans la suite ne concernent que le mouvement convectif et la dispersion apparente. Les processus de réaction, tels les échanges entre phases (liquide et solide) ou encore la biodégradation, peuvent conduire à des modifications au niveau des répartitions de concentrations mais sont directement liés à la nature particulière du type de contaminant envisagé.

2) Représentation simplifiée du mécanisme d'entraînement et de mélange: cas du déplacement miscible unidirectionnel en milieu poreux homogène.

Du point de vue physique, une eau marquée par une substance (traceur coloré par exemple) à la concentration C_0 est introduite à l'instant $t = 0$ dans une colonne de milieu poreux saturée en eau pure; ceci correspond à un échelon de concentration, à l'entrée de la colonne, tel qu'il est représenté sur la figure 2.5. Sous l'effet du déplacement de l'eau pure par l'eau marquée à travers le milieu poreux, la courbe de répartition des concentrations dans la zone de mélange entre les deux liquides miscibles évolue en forme de S.

Ce processus de mélange peut être décrit à l'échelle des pores par la prise en considération de mécanismes élémentaires d'écoulement, analogues dans leurs effets à l'agitation moléculaire (mouvement brownien) s'observant à l'échelle microscopique.

Les recherches expérimentales concernant le déplacement miscible, entreprises par H.O. PFANNKUCH (1963), ont clairement montré l'influence réciproque de la diffusion moléculaire et de la dispersion mécanique.

Au déplacement unidirectionnel, selon l'axe Ox , correspond un modèle mathématique qui se présente sous la forme de l'équation aux dérivées partielles:

$$\frac{\partial C}{\partial t} = D_L \frac{\partial^2 C}{\partial x^2} - u \frac{\partial C}{\partial x} \quad (2.2)$$

qui, avec $u = q/n$ et $D_L = D_a$, est identique à l'équation du § 2.4.1., où les termes (3) et (4) sont négligés.

Avec les conditions aux limites adéquates (cf. fig. 2.5), la solution de cette équation (évolution des concentrations selon une courbe en S) correspond à une intégrale de Gauss. Le coefficient D_L est déterminé à partir de l'écart type σ_L de la courbe de répartition, par la relation $\sigma_L^2 = 2 D_L T$ ou $T = L/u$, correspond à la durée du transport convectif à la vitesse réelle moyenne u sur le trajet $x = L$; ainsi on note que $C(x = L, t = T) = C_0/2$.

Pour un aquifère alluvial où $D_L = \alpha_L \cdot u$ (cf. § 2.3.2.3) on aboutit à la variance $\sigma_L^2 = \frac{L^2}{2} \alpha_L \cdot L$, relation qui exprime l'indépendance entre la largeur de la zone de mélange (elle est mesurée par $2 \sigma_L$, correspondant à la différence des abscisses relatives à $C/C_0 = 0,16$ et $C/C_0 = 0,84$) et la vitesse u .

On remarquera qu'avec l'augmentation du trajet $x = L$, la valeur de σ_L augmente mais moins rapidement que L : la zone de mélange (effet dispersif) perd de sa signification relative vis-à-vis du déplacement convectif au fur et à mesure que la distance L croît.

3) Représentation schématique tridimensionnelle de l'évolution d'une zone de mélange en milieu poreux homogène.

Partant d'une source de contamination ponctuelle dans des conditions de stationnarité et considérant l'évolution des concentrations dans l'espace à trois dimensions, la détermination des surfaces d'égale concentration est obtenue à partir d'une équation aux dérivées partielles. Dans l'hypothèse d'une isotropie des perméabilités et des dispersivités du milieu, d'un débit de source constant dans un courant

rectiligne, on aboutit au schéma de la figure 2.6.

Si u représente la vitesse réelle moyenne (direction de l'écoulement Ox), C la concentration, D_T le coefficient de dispersion dans le plan y, z , m le débit de la source (en masse de contaminant par unité de temps), la variation spatiale de la concentration est donnée par

$$C(x, y, z) = \frac{m}{4\pi D_T x} \exp\left(-u \frac{y^2 + z^2}{4 D_T x}\right) \quad (2.3)$$

solution de l'équation simplifiée

$$u = \frac{\partial C}{\partial x} = D_T \left(\frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right) \quad (2.4)$$

Si une norme définit pour la substance polluante concernée, une valeur limite C_L , alors, selon l'indication de la Figure 2.6, on détermine l'enveloppe Σ délimitant la zone de contamination proprement dite.

Relative à l'axe Ox , avec $D_T = \alpha_T u$ (cf. § 2.3.2.3), où α_T est la dispersivité du milieu dans le plan y, z , la concentration s'exprime par

$$C(x, 0, 0) = \frac{m}{4\pi \alpha_T u} \cdot \frac{1}{x} \quad (2.5)$$

A condition de connaître les valeurs de m et de α_T , on accède à la portée $x = L$ de la contamination (cf. fig. 2.6) par

$$L = \frac{m}{4\pi \alpha_T u} \cdot \frac{1}{C_L} \quad (2.6)$$

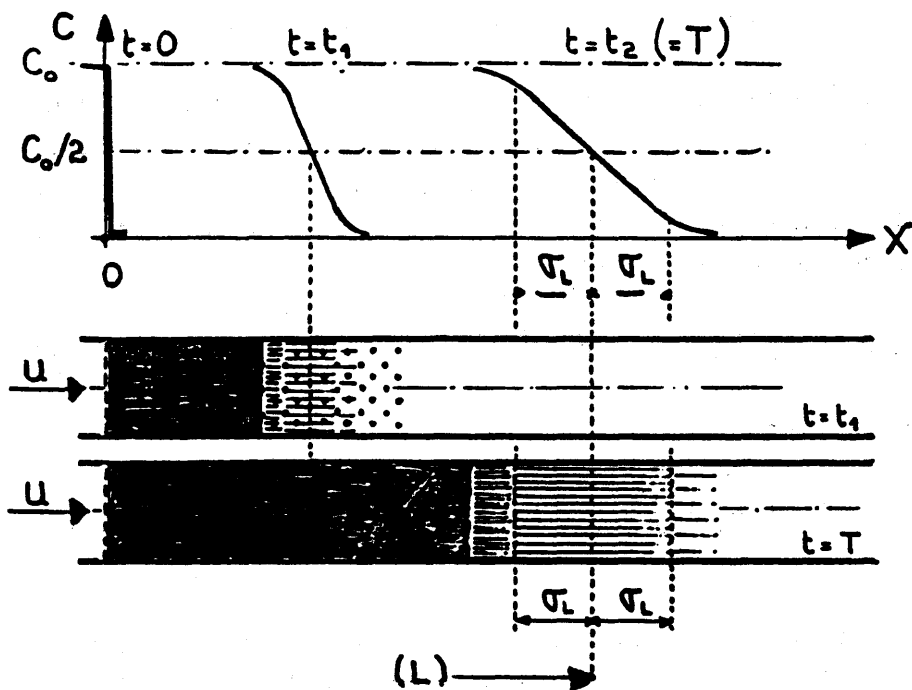


Fig. 2.5.: Evolution de la zone de mélange dans le déplacement miscible unidirectionnel

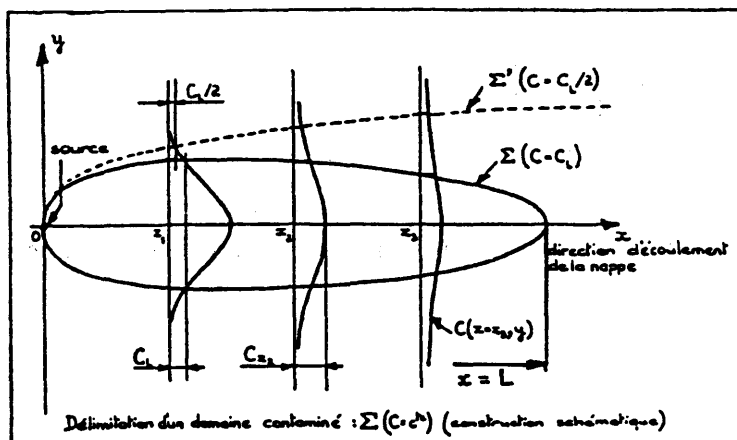


Fig. 2.6.: Vue schématique d'une zone contaminée. Cas tridimensionnel (coupe $z = 0$)

4) Influence de structures hétérogènes du milieu sur les mécanismes hydrodispersifs (Schémas bidimensionnels).

a) Déplacement miscible en milieu stratifié parallèlement à la direction de l'écoulement.

Les expériences effectuées sur modèle bidimensionnel vertical au laboratoire ont permis d'étudier les mécanismes de dispersion dans le cas du déplacement horizontal d'une eau douce par une eau légèrement salée et colorée à la fluorescéine la matrice poreuse est formée de deux milieux poreux disposés en couches alternées d'égale épaisseur (RENAULT et al., 1975).

L'échange transversal en régime transitoire a été mis en évidence par visualisation pour une alternance de couches de 5 cm d'épaisseur de deux milieux dont le rapport des conductivités hydrauliques est de 10 (cf. figure 2.7). L'analyse des concentrations, effectuée à la sortie du modèle aménagée de telle façon qu'elles soient représentatives de la concentration moyenne à la limite aval de la matrice poreuse à tout instant, confirme ces observations: sur la figure 2.7, la forme en dents de scie de la partie de la courbe correspondant aux échanges entre couches ($t \geq t_2$) est due à la façon irrégulière dont se font ces échanges (on comparera les résultats expérimentaux au tracé théorique en pointillé correspondant à l'absence de dispersion longitudinale dans chacune des couches et d'échanges transversaux entre les couches).

Le régime asymptotique a pu être atteint dans les limites du modèle pour deux milieux plus proches au point de vue conductivité hydraulique, avec une faible épaisseur de couches (1 cm).

Grâce à une technique de micro-prélèvements il a été possible de suivre l'évolution des vitesses locales moyennes dans

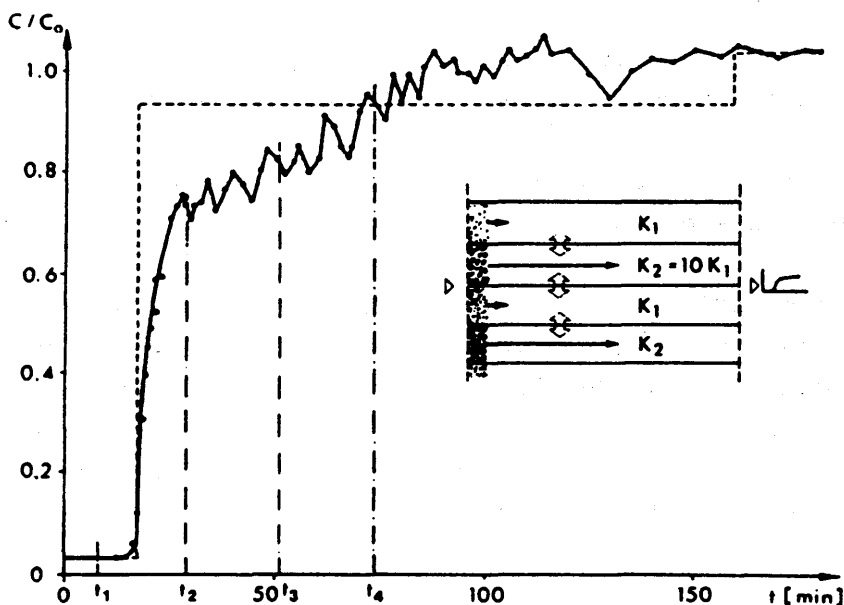


Fig. 2.7.: Courbe réponse d'un déplacement eau douce - eau salée en milieu stratifié

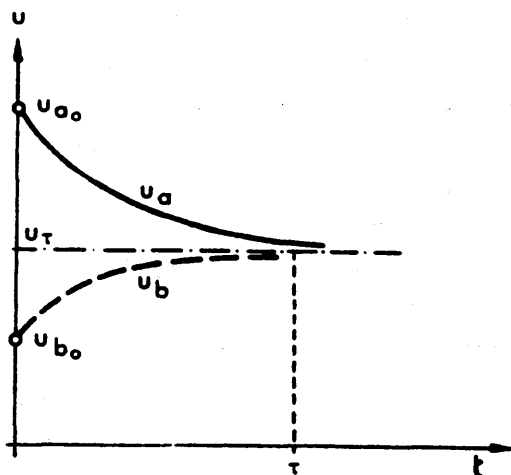


Fig. 2.8.: Evolution en régime transitoire des vitesses de déplacement d'un front miscible (ligne des $C/C_0 = 0,5$) en milieu stratifié parallèle à l'écoulement.

u = vitesse locale dans les couches perméables,
 u_a = vitesse locale dans les couches peu perméables,
 u_b = vitesse locale dans les couches perméables, (u_{ao} , u_{bo} = valeurs initiales "hydrodynamiques" à l'entrée de la matrice)

u = vitesse commune atteinte après le temps asymptotique

les deux types de milieu poreux et de constater leur convergence vers une valeur moyenne commune atteinte après un parcours correspondant au "temps asymptotique" τ (cf. graphe de la figure 2.8).

Ce mécanisme conjugué d'accélération et de décélération de la propagation dispersive par rapport aux flux hydrodynamiques dans les strates respectives, illustre la dépendance du paramètre "dispersivité" de la distance de parcours tant que les échanges transversaux sur l'ensemble de l'épaisseur de la matrice poreuse n'ont pas contribué à atteindre la valeur asymptotique de ce paramètre.

Dans ce contexte PICKENS & GRISAK (1981) indiquent que pour des systèmes hydrogéologiques où une dispersivité (asymptotique) constante peut être obtenue, l'incidence d'une dispersion dépendant initialement d'effets d'échelle reste minimale dans la prévision à long terme du transport de soluté. Pour le comportement en phase transitoire, ces auteurs ont présenté des fonctions de dispersivité α_L variant linéairement avec la distance moyenne de parcours L : à l'échelle d'un aquifère, la simulation du déplacement relative à la totalité de sa puissance est réalisée à partir de $\alpha_L = 0,1 L$.

b) Incidence d'une rupture verticale de perméabilité sur une zone de mélange stable issue d'une source ponctuelle d'eau salée placée dans un écoulement uniforme horizontal d'eau douce.

Dans le cas du milieu homogène-isotrope et de conditions d'écoulements stationnaires, le processus de mélange eau douce - eau salée a été décrit par ZILLIOX & MUNTZER (1971).

La figure 2.9 montre comment une zone de mélange stable est modifiée par la présence d'une hétérogénéité formée par une tranche verticale de milieu poreux dont la perméabilité est supérieure à celle du milieu environnant: on peut en particulier passer dans cette tranche à un régime d'instabilités conduisant à augmenter la dispersion dans le plan vertical.

A l'aval d'une telle perturbation on constate une extension en profondeur de la zone de mélange qui peut varier dans le temps; on remarquera par contre (et ceci est intéressant au voisinage immédiat d'un puits de captage d'eau) que la concentration moyenne dans la zone de mélange (zone de contamination) diminue dans ces conditions.

L'importance de ce phénomène hydrodynamique dépend évidemment du contraste de perméabilité, de la dimension de la tranche à forte perméabilité, des conditions initiales de l'écoulement et des contrastes de densité entre liquides.

2.5. Conclusions et Perspectives

Après l'analyse synthétique des éléments présentés dans les paragraphes précédents une conclusion s'impose: si un simple calcul de dilution permet dans bien des cas d'évaluer le taux de contamination possible d'un captage d'eau, il ne peut être fait abstraction, au plan de la prévision, de l'ensemble des conditions "d'écoulements hétérogènes" pouvant se présenter dans un aquifère poreux.

Le transfert des connaissances de base (depuis l'analyse des mécanismes élémentaires jusqu'à l'intégration nécessaire des divers processus) à l'action pratique de mise en place de périmètres de protection en situation réelle, nécessite encore une démarche concertée pluridisciplinaire.

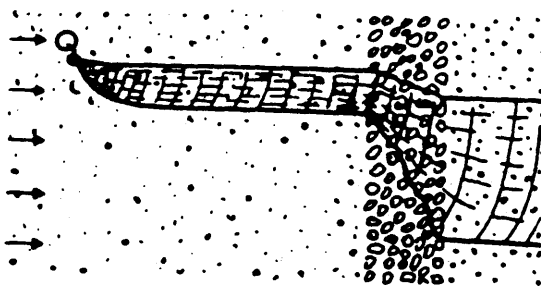


Fig. 2.9.: Influence d'une rupture de perméabilité transversale sur la dispersion verticale (avec contraste de densité)

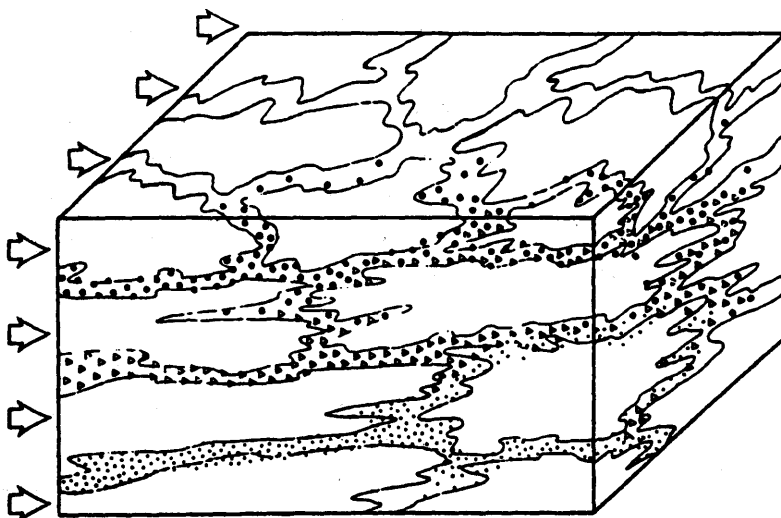


Fig. 2.10. Schéma de l'organisation des écoulements, à l'échelle décamétrique, avec formation de la dispersion (d'après BRISSAUD, 1982)

Dans l'examen des mécanismes hydrodispersifs et la mise au point d'outils de simulation, il est utile de répondre aux questions telles:

- le modèle théorique correspond-il vraiment aux lois physiques?
- le modèle numérique, s'il fonctionne bien, peut-il garantir l'utilisation des résultats obtenus dès lors que les valeurs attribuées aux paramètres fondamentaux sont peu sûres ou encore introduites par tâtonnement?

L'analyse de résultats de terrain a conduit F.BRISSAUD (1982) à entrevoir une "conceptualisation de la structure d'écoulement à l'échelle d'un système aquifère" et à proposer "un schéma de l'organisation des écoulements" (Figure 2.10). L'auteur y voit un ensemble de chenaux, liés à la distribution des perméabilités, à travers lesquels transite l'essentiel du débit d'eau. "Ces chenaux, possèdent une importante tortuosité et s'interpénètrent". Ainsi se trouve mise en évidence "une organisation anastomosée de l'écoulement analogue à celle décrite depuis longtemps, à l'échelle microscopique, par la théorie de la dispersion. Par analogie avec les résultats obtenus dans les milieux homogènes, on peut penser que la dispersivité est liée à la dimension et à la morphologie des volumes poreux peu concernés par l'écoulement enserrés entre les chenaux".

Sur la base de cette représentation schématique on peut comprendre qu'à l'échelle d'une parcelle expérimentale l'équation de dispersion puisse être mise en échec alors qu'à l'échelle de plusieurs kilomètres (les distances parcourues étant dans ce cas beaucoup plus grande que les dimensions des éléments qui structurent les cheminements fluides), cette équation reste valable pour représenter (comme dans un milieu homogène) les distributions de concentration observées.

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PART II

GROUNDWATER PROTECTION IN DIFFERENT AQUIFER TYPES

CHAPTER 3

PROTECTION OF THE GROUNDWATER QUALITY IN POROUS PERMEABLE ROCKS

by

H.G. van Waegeningh

3.1. Introduction

This paper deals with a system for the protection of the groundwater quality, which is based on the delay times of groundwater in aquifers that are composed of essentially unconsolidated, porous permeable sediments. As the flow velocity in such aquifers is generally not very high, the resulting protection areas will have realistic dimensions. In the case of high flow velocities, the protection areas could be impractically large, and realisable areas can be attained by changing the required delay time, but thus introducing more risks to water supply.

The advantage of a delay time basis for protection areas around groundwater supply sources is that it can be related not only to economical-technical factors, but also to the behaviour of contaminants underground.

A delay time system has been developed and is applied in The Netherlands, and will be explained and illustrated.

3.2. Situation in The Netherlands

The hydrogeological situation of The Netherlands can be summarised as follows. Since the Tertiary the greater part of the country has been a continuously-sinking basin, in

which sedimentation of marine and fluvial sands and clays has taken place. There was little tectonic activity (faulting). During the Pleistocene ice ages some sediments have been thrust vertically by ice but most formations are horizontally layered. The topography is flat and only locally hilly. Only in the extreme south and east of the country are consolidated rocks (sandstone and limestone) found. In comparison with many other countries one is dealing mainly with rather uniform, horizontally layered aquifers of unconsolidated sands and clays. In these aquifers there is laminar flow of groundwater with velocities of 10-100 m per year. Another important feature in The Netherlands is the high groundwater level, which over large areas varies only between 0-2 m below surface.

This picture is oversimplified; the situation is much more complicated and some details are important when discussing protection problems.

Another important difference from many other countries is that the production of drinking water is concentrated; there are about 100 water-supply companies with 240 wellfields, which yield over 700 millions m^3/a . Private wells for drinking water-supply are rare, and nearly all houses have piped supplies.

Because of the great value of groundwater in drinking water-supply, the accent is always on the protection of drinking water-supply areas in discussions on soil and groundwater protection; thus on the protection of production wells. This does not mean that the protection of other areas can be neglected.

In principle the entire recharge area around wellfields should be protected against any change in the properties of groundwater, so that the natural quality is preserved. This would be the most reliable and simple procedure to protect groundwater for drinking water purposes. However, protection

of the entire recharge area against all influences is not a very realistic option, especially in densely-populated or highly-industrialised countries. Moreover, that strict criterion is not absolutely necessary, because of the acceptability of some small changes in quality or because of attenuation processes in the underground.

3.3. Some general considerations

To achieve the second-best solution, certain fundamentals have to be defined.

- a) Some risk to water-supply has to be accepted when choosing a second-best solution; this is a realistic view, since in practice conflicts between the security of water-supply and other interests can not always be settled in favour of water-supply. Technical guidelines for protection could help in solving the conflicts.
- b) Protection of groundwater quality must be of a preventive nature from the public health view point; moreover rehabilitation afterwards is very complicated in technical, financial and administrative respects.
- c) Because of the properties, the quantity and the place and the risk of emission of a pollutant, a system of zoning of the recharge area or the protection area, is desirable; this zoning could be dependent on soil properties or on properties of the possible contaminant or on short and longterm protection.
- d) Until now the relevant sciences can not give satisfactorily precise predictions on the behaviour of contaminants in the underground; one still has to extrapolate rather general conclusions from much research. Also one can not rely on the elimination of pollutants by

attenuation and degradation underground. In making a prognosis in a protection problem, as in all public health issues, one has to allow a factor of safety.

- e) A protection system should be based as far as possible on scientific results. On the other hand the socio-economic circumstances strongly influence the necessary restrictions and prohibitions in the different protection zones. There is a direct relationship between the zoning criteria and the manageable restrictions, together with differences in degradability of contaminants.

3.4. Contaminant hydrogeology

Contaminant hydrogeology is the science which studies the transport and behaviour of contaminants in the underground; the most elements are hydrodynamics and hydrogeochemistry. Both have already been reviewed, what remains is to point out some special properties of porous permeable rocks.

In hydrogeochemical respects, the most important differences between porous permeable rocks and fissured or karstic rocks are the more intensive contact of the contaminant with the rock matrix and the relatively longer delay time. Within the first type the physico-chemical and microbiological reactions of degradation or attenuation will have better effects comparative to fissured and karstic rocks. When considering processes in the unsaturated zone with vertical transport, it can be stated that, in principle, there is no difference between the different types of aquifers. The physico-chemical and microbiological attenuation processes in the unsaturated -and especially the soil - zone are much more effective than in the saturated zone. When enough insight of and specific data on these processes exist, the unsaturated zone should be taken into account when solving a protection

problem. The same holds for semi-permeable covering layers; only when their presence is proven and their properties known can they be reckoned upon. Up to present most attention has been paid to the transport and behaviour of pollutants within the aquifer and thus mainly to horizontal transport. There is not enough knowledge to take into account the influence of the soil and associated vertical transport; the influence has to be evaluated for each case separately. Moreover some hazards to groundwater quality can take place directly within the aquifer or bypass the unsaturated zone. The development of hydrodynamic computer models has resulted in a sound insight of groundwater flow and pollutant pathways. In principle the possibility for calculation of solute transport was previously available, but the techniques were mainly applied to resource problems; for example the technique of equal squares of equipotential and flow lines. With modern methods calculations can be made rapidly; the availability of computer models coincided with interest in solute transport and pollution problems. Some simple examples of results are given in figures 3.1 and 3.2.

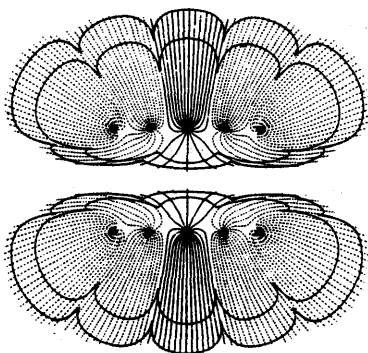


Fig. 3.1: Flow lines and lines of equal delay time around a well.

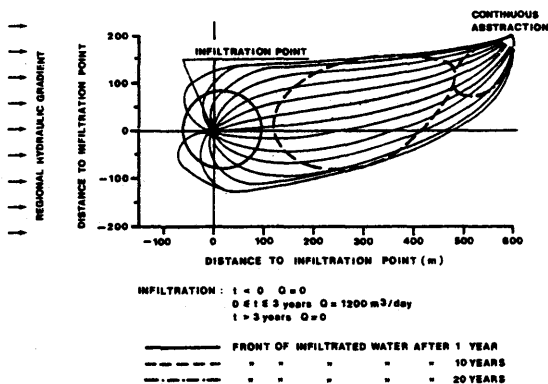


Fig. 3.2: Moving fronts of polluted groundwater

When data are available, it is possible in principle to have most intricate patterns of groundwater flow computed and drawn automatically, including delay times for groundwater. Together with the knowledge of attenuation processes and retardation factors, it is possible to construct computer models of the movement and behaviour of pollutants in the underground. It is even expected that models of the saturated and unsaturated zones can soon be combined, so that it will be possible to predict where, when and at what concentration a pollutant can be found in the underground following emission. However, although techniques are available, lack of exact data often prohibits the use of such intricate models. Besides research in the field of contaminant hydrogeology, other research is also necessary. In relation to protection it has already been mentioned that prevention is the important factor. However, in practice, protection also must be curative, in the sense of rehabilitation of polluted groundwater. So research on preventive and curative measures has to be done; for example, vulnerability atlases and monitoring networks, which are important tools in regional planning and in control of the groundwater quality. In the curative sense attention should be paid to rehabilitation methods. It is not appropriate to digress on these topics here.

3.5. Delay Times for Protection Zones; the System in The Netherlands

Until about eight years ago the system of protection in The Netherlands was almost a copy of the system in Western Germany. The zoning system was based on a delay time of fifty days and on a distance of two kilometers (see Chapter 6). As the distance of two kilometers for the boundary of a zone III A has no scientifically-based background, much resistance was met with the establishment of protection zones in

The Netherlands. When implementing protection areas one has to argue as strong and convincing as possible. So in The Netherlands another zoning system has been introduced based on technic-economic conditions, translated into delay times of groundwater. As not to deviate too much from the historical position the aim was to keep as far as possible the same restrictions and prohibitions as previous.

Historically the first protection exerted was against pathogens (bacteria and viruses) and rapidly degradable chemicals. The criterium of fifty days of delay in the aquifer, required for the decay of germs, is being studied nowadays with modern techniques.

Pending the results of research, in The Netherlands delay time - zones of at least fifty days are recommended; where possible zones of one hundred days or one year should be established. This catchment area should be protected very strictly, since inadequate time for rehabilitation is available. In Dutch conditions the zone will reach 30-150 m from the wells.

In the case of severe pollution of a persistent compound within the recharge area, an attempt must be made to repair the damage. In many cases this repair will take a long time and often will be imperfect, so the wells may be affected. The situation should be monitored in order to decide on rehabilitation measures, which may include the development of new purification processes or even the abandonment of the entire catchment, since the Water-Supply Act obliges the Water Works to provide water of good quality in sufficient quantity. For the sake of the continuity of water-supply and to exclude public health risks, a delay of groundwater in the aquifer of at least 10 years is needed, which results in the 10 - year protection zone.

Because in many cases even 10 years will not be sufficient to guarantee the continuity of safe water-supply, and

because of associated economic reasons a 25 year protection zone is also necessary. The protection zones of 10 and 25 years together are called the protection area; in the Dutch situation the different zones extend to about 800 and 1200 m respectively from the wells. The system is schematically represented in fig. 3.3.

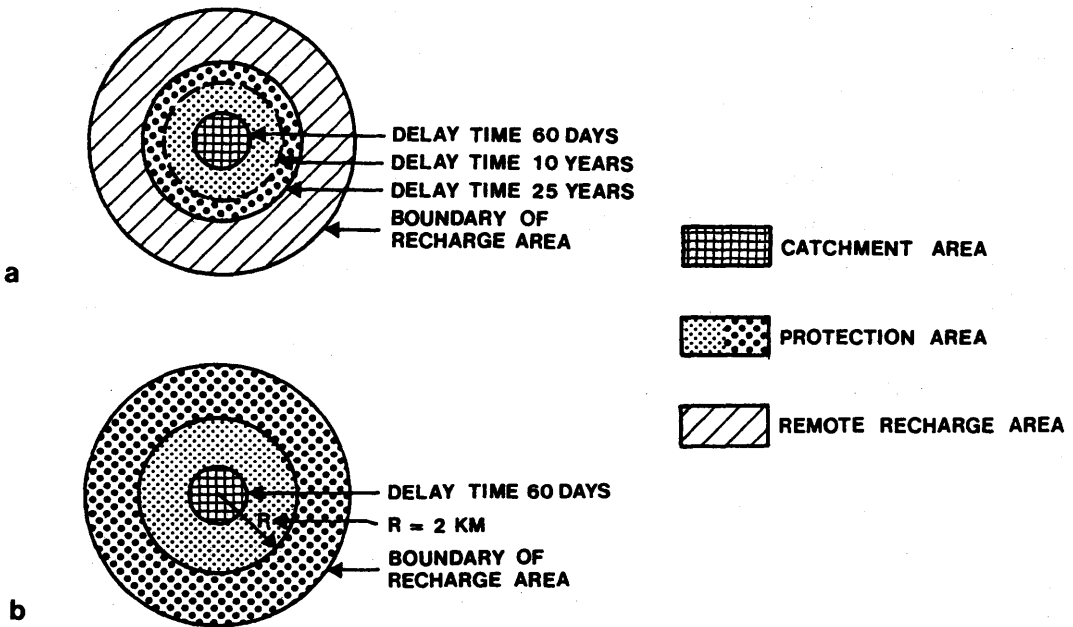


Fig. 3.3: a) Porous-permeable aquifer b) fissured karstic aquifers

It is clear that the principle of minimum delay times for protection zones in the case of high groundwater flow velocities will lead to such large protection areas that they are unmanageable in practice. It may then be necessary to reduce the required delay time. It should be recalled that in Western Germany the two kilometer boundary for zone IIIA has proven to be workable and for high velocity aquifers

this experience could be followed, as is indicated in tab.3.1.

Tab. 3.1: Restrictions in protection zones.

CATCHMENT AREA	PROTECTION AREA	REMAINING RECHARGE AREA
60 DAYS AND ≥ 30 m	10- AND 25 YEARS DELAY-TIME OR 2 KILOMETER	
PROTECTION AGAINST PATHOGENIC BACTERIA AND VIRUSES, AND AGAINST CHEMICAL POLLUTION SOURCES	PROTECTION AGAINST HARDLY-DEGRADABLE CHEMICALS	RULES OF ACT ON SOIL AND GROUNDWATER PROTECTION
ONLY ACTIVITIES IN RELATION TO WATER- SUPPLY ARE ADMISSIBLE	AS A RULE ARE NOT ADMISSABLE E.G.: - TRANSPORT AND STORAGE OF DANGEROUS GOODS - INDUSTRIES - WASTE-SITES - BUILDING - MILITARY ACTIVITIES - INTENSIVE AGRICULTURE AND CATTLE- BREEDING - GROUND-, SAND- OR LIMESTONEPITS - WASTE WATER	

It is clear that in choosing this solution, a third best solution, still more risks to water-supply are introduced, not only because of the shorter residence times, but also because of the generally more vulnerable character of the aquifer. In these cases it may be appropriate to achieve an optimum hydrologically based catchment area in view of pollution sources or to install a more advanced water purification plant, or in case of emergency to be able to abandon the catchment immediately.

Returning to delay times it still has to be stated that in principle these should be calculated within the aquifer, in horizontal direction; only when one can be very sure of the purifying or attenuating processes or of the impermeability of the covering layers, can one include the vertical delay time of the infiltrating polluted water.

Furthermore, it is stressed that outside the defined protection areas, the more remote part of the recharge area should not be forgotten, especially in view of any severe pollution of persistent character. A legislation on soil and groundwater protection should have a regulatory influence, in which the interests of water-supply must play a role.

3.6. Restrictions in protection zones

Although the accent is laid on hydrogeological factors of groundwater protection, it should not be forgotten that within certain areas restrictions on land use and on activities of people should be imposed. The character of these restrictions and the criteria for dimensioning are interrelated. Historically most attention has been paid to pathogens; nowadays the tendency is to concentrate more on pollution by chemical compounds.

The types of pollutants are different in origin (point- or diffuse sources) and in underground behaviour; and these differences not only form criteria for the dimension of the protection zones, but also for the type of restrictions. Indications of restrictions are given in the table 3.1.; it is clear that these become more severe as one approaches the wellfield.

3.7. Conclusions

Up to the present the protection system applied in The Netherlands is based on delay times, and on technical and economical arguments. However, it should be possible to change these arguments for the delay time basis into more scientifically based arguments.

Until now little is known on the physical, chemical and microbiological processes in the saturated and unsaturated

zone; moreover the available knowledge was rather scattered. Some years ago large research programmes on degradation and transport of pollutants have been started, and cooperation of different disciplines grew; recently much progress is made.

Most degradation and transport processes are time dependent, or can be translated in time dependency; and that could be the method of translating the attenuation processes into the minimum delay times required for protection zones.

A system based on scientific arguments has the advantage of making the state of the art objective.

When the required data are available, all hydrogeological and hydrogeochemical features, horizontal and vertical movement, and all time dependent physical, chemical and biological processes can be taken into account. The risk of pollution to water-supply can be assessed and introduced into the process of weighing the balance between water-supply and other interests. In this process and in legislation, the socio-economic circumstances of a country, or a region are introduced, and decisions on protection problems can be made.

In indicating systems for protection of water-supply areas, practicable solutions have to be found. It is clear that with the defining of required minimum delay times attention has to be paid to the practicability of the resulting protection zones with restrictions to land use. As differences between countries exist in technical aspects and in the socio-economical and legislative situation, it will not be possible to reach an exactly uniform system of water-supply protection in all countries. However, the basis of minimum required delay times could form a sound technical criterion for groundwater protection, the length of the delay times being defined on other factors.

CHAPTER 4

GROUNDWATER PROTECTION IN FISSURED ROCKS

by

A.Ch. Skinner

4.1. Introduction

This paper is in two parts. The first part deals in general terms with the characteristics of fissured rocks which are relevant from the point of view of groundwater protection. The difficulties of reliable quantitative estimation of pollutions movement in this type of strata are emphasised. The second part of the paper assesses the implications of this for a groundwater protection strategy for fissured rocks.

4.2. Characteristics of Fissured Rocks

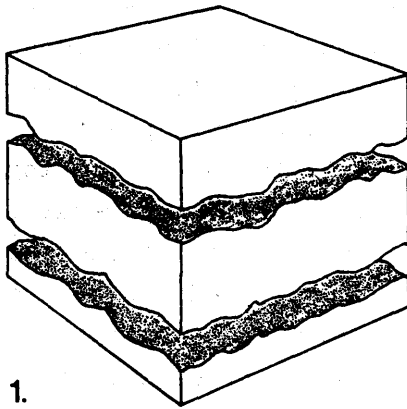
Fissuring is a significant factor in the description of the hydraulic flow regime of a wide range of aquifer types. Virtually all consolidated aquifers exhibit fissuring to some degree, and for many it is the only feature permitting significant fluid flow. The scale of fissuring varies significantly in both aperture and frequency of occurrence. The fissures may be formed by a variety of geological processes and may have their geometry modified by secondary processes of solution and precipitation. The hydraulic characteristics of the blocks of aquifer between the fissures also varies widely, from metamorphic and indurated sedimentary

rocks with negligible porosity to less consolidated sedimentary strata, which retain a significant intergranular porosity.

The spatial distribution of the fissures has a strong influence on the hydraulic characteristics of the aquifer. Fig. 4.1. demonstrates, in diagrammatic form, typical fissure configurations. Example 1 shows a situation where the fissuring is primarily sub-horizontal, as may occur where fissure development is along bedding plane discontinuities. In this case horizontal hydraulic conductivity will be dominant over that in the vertical direction. Example 2 shows a situation where the aquifer is split into fairly uniform blocks by jointing. The relative hydraulic conductivity in any direction will depend upon the relative development of the individual joint directions. Where one of the sub-vertical joint components is strongly preferred then an areal "permeability fabric" will be imposed on the aquifer, with a high permeability parallel to the prevailing joint direction. A similar situation will arise when the fissuring is developed along metamorphic shear zones in basement aquifers where a strong sub-vertical lineation is quite common (Example 3).

The frequency of fissure occurrence within the rock mass can vary over several orders of magnitude. The Chalk, the principal aquifer in the United Kingdom, is an example of an aquifer with a fine fissure structure, typically 2 mm or less in width and occur at spacings of 1 metre or less. In contrast fissure spacing in jointed basement rocks can be one hundred times greater with a similar proportionate increase in fissure aperture.

These differences in fissure type, geometry and scale pose considerable problems for the design of investigation programmes for fissured groundwater systems and the interpreta-



RELATIVE HYDRAULIC
CONDUCTIVITY

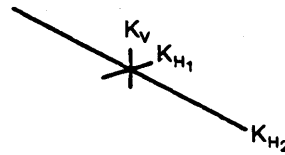
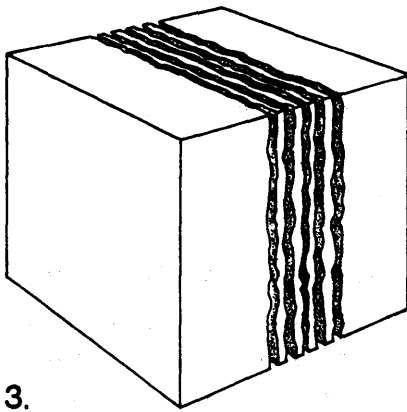
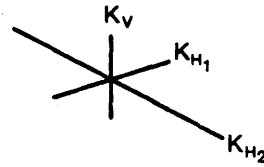
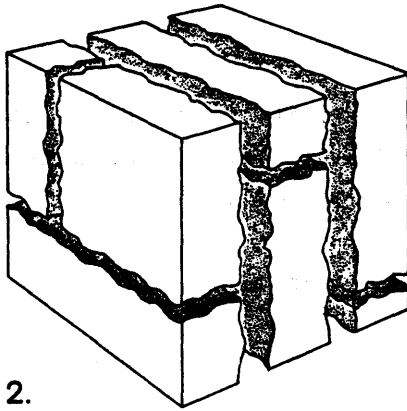
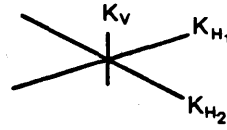


Fig. 4.1.: Effect of fissure geometry on hydraulic conductivity.

tion of the data derived from them. Experience has shown that study of a fissured aquifer, after a pollution incident has occurred, with the object of defining the location and distribution of any contaminant is a difficult practical problem. It often requires extensive drilling of monitoring boreholes which may give ambiguous or conflicting results because of the heterogeneity of the aquifer (BIRK and VORREYER, 1978). Groundwater protection procedures depend upon the prediction of what might happen to pollutants if they were introduced. This is a much more difficult problem than the study of a pollution incident, because it must be tackled with much less data and without the benefit of the tracer which is provided by the pollutant.

The understanding of groundwater protection in fissured aquifers depends upon a knowledge of many factors of which the following are considered to be the most important:

- I) magnitude and direction of groundwater flow,
- II) degree of inter-reaction between water in the fissures and in the intervening rock matrix,
- III) influence of the unsaturated zone,
- IV) degree of inter-reaction with surface water systems,
- V) rock/water reactions within the aquifer.

This list is not exhaustive, nor does it contain factors which are only relevant to fissured rocks. It is presented to identify the key areas of uncertainty which affect decisions on groundwater pollution problems in these aquifers.

4.3. Natural Groundwater Flow

Groundwater pollution problems in an idealised isotropic aquifer with no naturally imposed flux are relatively simple. Residence times in groundwater abstraction areas depend only on effective aquifer thickness, porosity and rate of abstraction. Of these the porosity is the most likely to be uncertain but the range of variation is not large and it may be measured to an acceptable level of accuracy by a number of simple methods. The experienced hydrogeologist will be able to determine a reasonable value by inspection of a sample or an outcrop.

The presence of a natural flux considerably complicates the problem and increases the number of parameters to be determined. To apply DARCY's law it is necessary to know the permeability and the groundwater potential distribution. Both of these are difficult to determine in fissured aquifers.

4.3.1. Measurement of Permeability

In fissured aquifers permeability is recognised to be a scale dependent parameter, such that the value obtained in any test will depend upon the volume of aquifer investigated. Measurements of rock samples by laboratory methods will generally only measure the permeability of the rock matrix and thus seriously underestimate the bulk permeability. More reliable estimates will be obtained by field measurements, although the value of the test may depend on the type of test and its duration. Thus a short duration single borehole drawdown or injection test may only effectively measure the permeability of a few cubic metres of rock, which may be unrepresentative of the aquifer volume as a whole. A long duration pumping test with a wide distribution of observation boreholes will give better information of a much greater aquifer volume but at much increased cost.

The concept of the representative elementary volume is commonly used to describe a minimum volume of aquifer which can be regarded as homogenous for the determination of a particular parameter. This assumes, as in Fig. 4.2. (a) that, although there may be fluctuations in the parameter value at small scales, as the volume of inspection increases, the parameter will tend to a constant value. The point at which this value is first reached corresponds to the representative elementary volume. However it is possible to envisage, in a fissured medium with families of fractures developed in different scales, that a single limiting value will not be reached but that the relationship would have a stepped form, Fig. 4.2. (b), changing as each increasing fissure set is encountered. Unless the hydrology of the system is very well understood the appropriate value of permeability to be taken for any particular problem will not be known. In these situations the measurement of permeability becomes a stochastic rather than a deterministic problem. This is well demonstrated by KOVACS (1983) in her estimation of the representative elementary volume for porosity in carbonate aquifers in Hungary. A stochastic approach requires a substantial data set for reliable use. This is hard to achieve for permeability measurements in consolidated rocks.

These problems will be compounded if the development of fissures imposes a strong directional anisotropy, where determination of permeability in aquifer tests depends not only on the distance but the direction of observation boreholes. The practical effect of this is to increase the number of observation points needed to make a useful assessment of permeability.

4.3.2. Measurement of Piezometric Head

The distribution of piezometric levels must be known to determine the natural groundwater gradient. This information

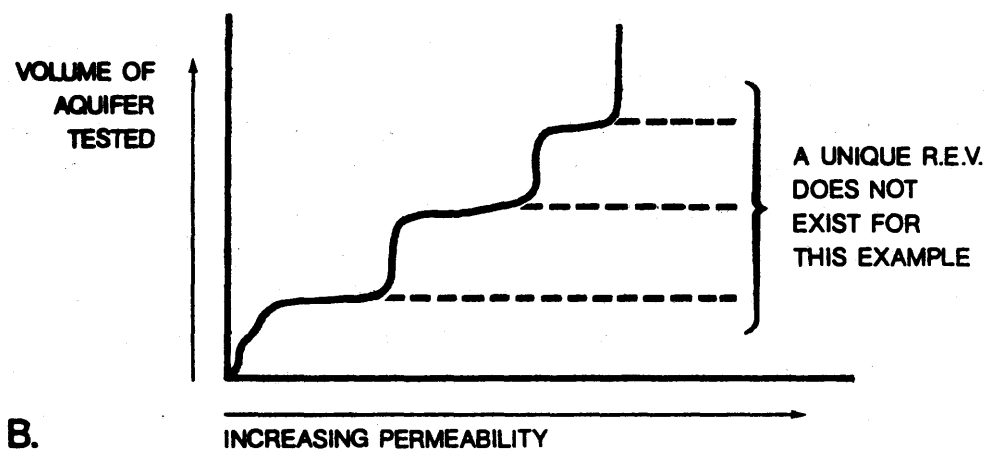
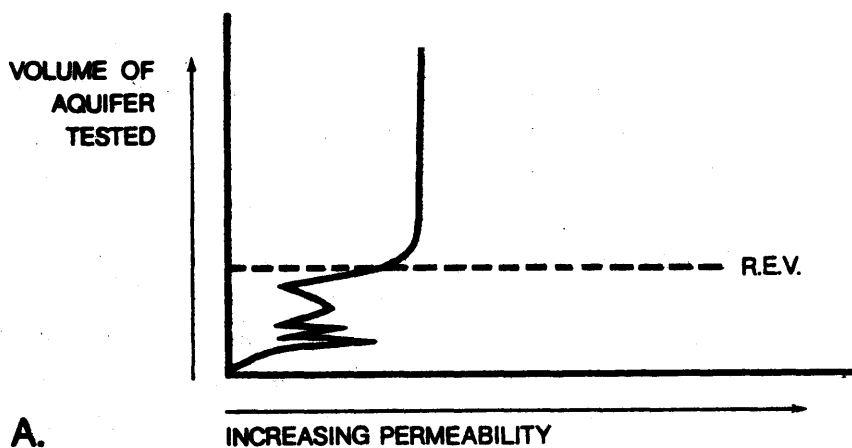


Fig. 4.2.: The concept of the representative elementary volume (R.E.V.) applied to permeability measurements in fissured strata.

is necessary even if groundwater flux to a borehole are being calculated from tracer experiments, otherwise it will not be possible to separately identify that part of the transport due to the natural gradient and that part caused by radial flow to the pumping well. Unless there is a good network of observation points it is often difficult to construct a reliable piezometric map and determine the magnitude and direction of the groundwater gradient since piezometric level will be influenced strongly by observation borehole depth and location in relation to the fissure system. It must also be remembered that in a strongly anisotropic aquifer flow nets become non-orthogonal and thus flow directions are less easy to determine.

4.3.3. Estimation of Groundwater Flow

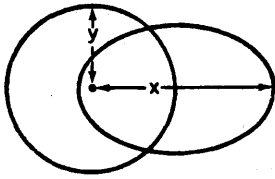
These considerations lead to the conclusion that there is bound to be uncertainty both as to the magnitude and direction of groundwater flux in a fractured aquifer unless there are adequate data. These data can only be obtained by a network of observation boreholes sited with a knowledge of fracture size, density and orientation. Tracer tests are also an essential component and can help to bring realism into the otherwise conservative estimates of flux which are likely to result from estimates based on rock permeability measurements. It is essential, as in all tracer tests, that estimates are based on positive breakthrough, and not inferred from negative evidence.

Groundwater flux in fissured aquifers is usually high. In extreme cases, even in non-kastic aquifers, rates of more than 1 km/d have been recorded. At these velocities flow conditions are turbulent and DARCY's Law is no longer valid. Rates of between 1 and 10 m/d are commonly recorded. At such fluxes protection zones based on minimum residence times become very large and strongly elliptical. As an example

Fig. 4.3. shows the relationship, based on the equations of SPITZ et al. (1980), of the ratio between the radius of the circular zone which exists in a situation of where there is no regional flux (y) to the long axis of the ellipse formed in an isotropic aquifer with a uniform regional gradient. It can be seen that for fluxes of less than 1 m/d the eccentricity of the protection zone is hardly significant, except for very large pumping rates. At fluxes of 1 m/d or more the zones become more elliptical with x/y ratios of up to 4. This effect will be more extreme if the permeability of the aquifer is greater in the direction of greatest flux. Whereas it is possible to define such zones mathematically, there are many practical problems in defining them with confidence on the ground.

4.4. Movement of Solutes in "Double Porosity" Aquifers

Many aquifers which exhibit fissure flow also have a significant porosity in the interstitial aquifer blocks. These rocks are sometimes referred to as "double porosity" or "fissured porous" rocks. This situation is worthy of special attention because it occurs in many aquifers of national significance (both of the major aquifers of the United Kingdom, the Chalk and Triassic sandstones are of this type) and it can have a significant impact on the movement of solutes. Water contained in the fissures, which is more mobile, need not necessarily be in equilibrium with water in the pores of the aquifer matrix. The extent of the equilibrium and the extent to which any pollutant is partitioned between fissure and pore water depends among other things upon the rate of flow through the fissure. In very large fissures the rates of flow will be so great that loss of pollutant to the matrix will not be significant. In smaller fissures the degree of diffusion could depend critically on flux. Thus an injection of pollutant into a fissure system under surcharge could lead to rapid transport without diffusion, whereas



$$A = \frac{2\pi mt}{Q\eta}$$

Where m is effective aquifer thickness (m)

η is aquifer porosity

Q is Volume pumped (m^3/d)

t is number of days residence time (t)

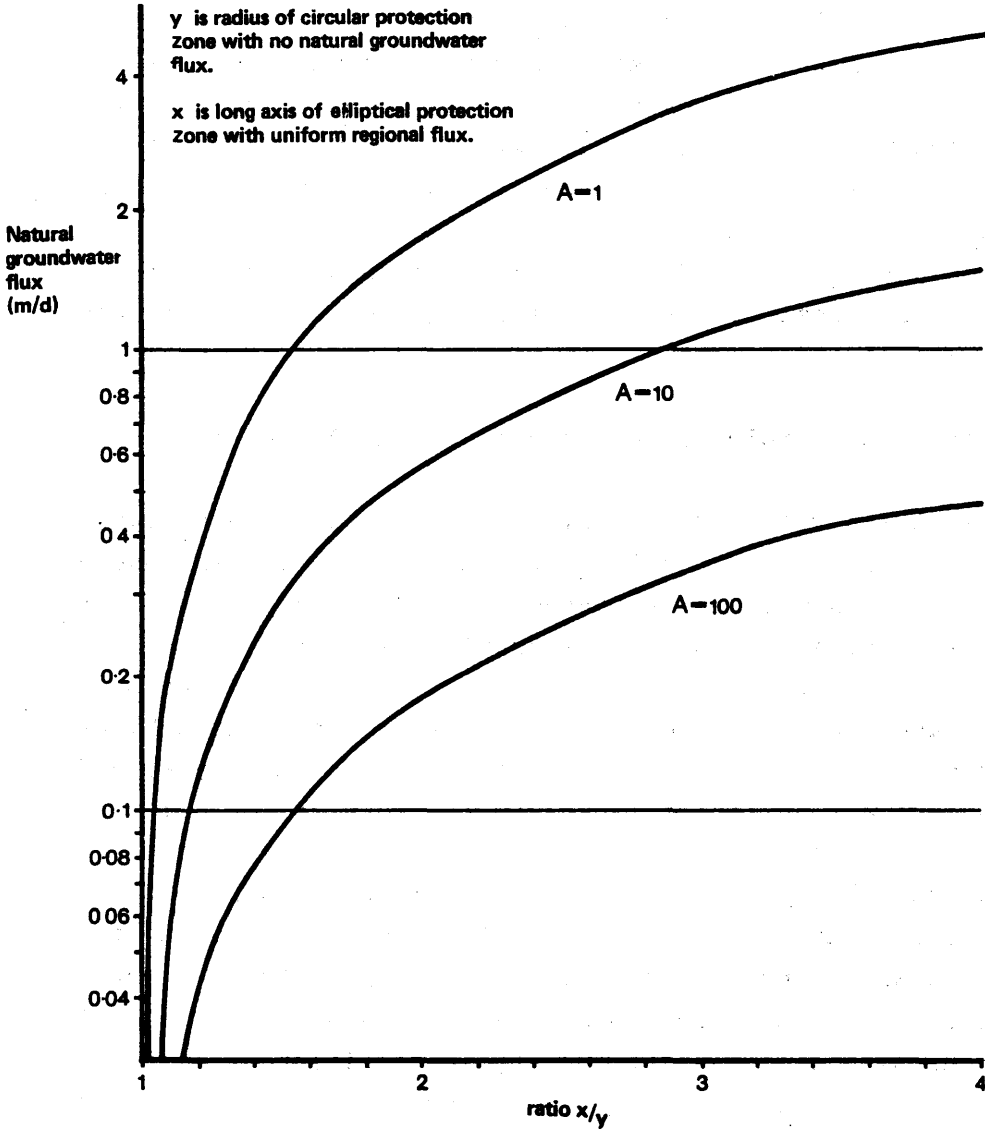


Fig. 4.3.: Eccentricity of protection areas as a function of groundwater flux homogeneous isotropic aquifers.

pollutants at lower fluxes would have adequate time to participate in a diffusion process with the matrix. These considerations are important in the design of tracer tests to assess residence times. These phenomena have been extensively discussed with respect to the Chalk, where, because of the high porosity and small pore size of the aquifer matrix, the phenomenon is especially well developed (REEVES, 1979; BARKER & FOSTER, 1981).

4.5. Role of the Unsaturated Zone

Fissured rocks, being consolidated and resistant to erosion, are often found in areas of significant topographical relief. This leads to a variable and often significant thickness of unsaturated strata above the water table. The typical low porosity of fissured rocks also means that the natural fluctuation of the water table in response to rainfall will be greater than for higher porosity rocks, with a corresponding fluctuation in the thickness of the unsaturated zone throughout the year. This unsaturated zone can provide an effective reduction in the rate of subsurface water flow and thus increase the residence times of pollutants moving from the surface towards a borehole. The effectiveness of this barrier will depend upon the nature of flow in the unsaturated zone and the intensity of the infiltration. Unless the rock matrix is very impermeable, the majority of the recharge at low infiltration rates will flow through the rock matrix and not through any fissures which are present. This will ensure a long travel time. At higher infiltration rates, such as may occur during periods of high rainfall, or if the system is locally surcharged by surface spillage, then a significant proportion of the flow will be through any fissures present. Diffusion will take place between the fissure water and the pore water, but in large fissures this will not take place at rates sufficient to significantly dilute a migrating pollutant. The access of surface pollutants

to the fissure system will also be controlled by the moisture content of the soil zone, where, at low moisture contents, the soil zone will act as a buffer regulating the rate of inflow to the underlying aquifer.

The general picture is that in many situations the unsaturated zone might be expected to provide a significant barrier to flow and thus considerably increase the protection to the aquifer. However in fissured aquifers, especially where there is a substantial vertical fissure component, this protection cannot be relied upon and it is possible for very rapid transfer of the pollutants to the water table to take place under certain recharge conditions. Unless the detailed geology and hydrogeology of the area are understood, it is difficult to place much reliance on the protective properties of the unsaturated zone. A good example of the need for detailed knowledge to assess the degree of protection is in igneous aquifers found in many tropical regions where, although the rock in its primary state is extensively fissured and thus very vulnerable, much of the groundwater development takes place in the upper extensively weathered zone, where the fissuring is either lost or no longer dominates the flow regime, and where greater protection from the unsaturated zone might be expected.

4.6. Inter-reaction with Surface Water Systems

It is a widespread practice of long standing to establish water supply boreholes in close proximity to rivers. This is done for many practical reasons, such as facilitating disposal of water during testing, but also has the advantage of exploiting induced recharge. This latter aspect is particularly relevant in fissured, indurated strata, where the storage properties of the aquifer are low and induced recharge can ensure a sustained yield at times of low recharge. The success of this practice, and the low incidence of pollution

problems arising in this situation, demonstrates the effective purifying properties of river bed substrates. It is however difficult, except with the experience of operation at an individual site, to be certain that natural quality protection will be provided. The nature of the protection may vary depending on rate of abstraction and river stage. If the protection of the river bed is breached, transport within the drawdown cone of the pumping borehole is rapid. If the stream is naturally of high quality an illusion of security may still exist, even if river water is reaching the borehole unfiltered. The problem will only be revealed when a major polluting discharge to the stream takes place. Groundwater protection policies for fissured aquifer strata should recognise this possibility (BLAU, 1981).

4.7. Rock/Solute Reactions

The wide variation in residence times within the fissured aquifer both in the saturated and unsaturated zone make prediction of pollution attenuation by inter-reaction between the solute and the rock matrix very uncertain. Where the aquifer is of the porous fractured type then rock/solute reactions will increase the partitioning effect above that which would occur by diffusion alone. This partitioning effect is dependent on the rate of flow through the fissure system. The partitioning will also vary depending on the physical and chemical characteristics of the rock matrix. The uncertainties as to the extent of rock/solute reactions in any given aquifer under potentially variable flow regimes mean that reliance on attenuating processes to protect groundwater quality is unwise.

4.8. Implications for Groundwater Protection Strategy

The proceeding discussion has illustrated a number of particular features of fissured strata in relation to pollutant migration and groundwater protection. These are:

- 1) high flow velocities
- 2) anisotropy of the aquifer
- 3) limited possibilities for attenuation and purification
- 4) difficulty in quantifying all of the above reliably without detailed investigation

Experience in the definition of protection zones based on minimum aquifer residence times by tracer tests or using non-dispersive velocity models or a combination of both is available from a number of European countries. The approach has proved effective both from a technical and administrative point of view. The approach has been developed and most extensively applied to the porous-permeable aquifer case. A similar approach applied to fissured strata faces many difficulties. Protection areas will be large and irregular in shape. Since they will be often based on poor and possibly conflicting data the natural reaction in such situations is to define the areas conservatively. It may be possible to reduce the protection area if more data are available, but this will incur additional costs. The hydrogeologist has the difficult task of deciding the level of investigative effort which is appropriate.

Very large protection areas will not meet with approval from other social and economic interests since it will be considered that the interest of water protection is being unreasonably favoured at the expense of other interests. This issue is already well recognised, and, in practice,

protection areas in fissured aquifers are often defined by a convenient "rule of thumb". VAN WAEGENINGH (1983) reports that in Germany a 2 km radius is used for the protection area in this type of aquifer. This approach, although apparently effective in this application cannot be recommended for adoption on an international scale without more rational justification. It depends heavily on the type of hydrogeological environment in which it has been proven and the existence of a legal code to provide enforcement.

It is important that these problems be recognised and that a system of groundwater protection is defined which is realistically achievable. On a worldwide scale fissured aquifers probably predominate over non-fissured and it is necessary that a rational system for approaching this problem be devised. This leads to the question of whether protection areas by themselves are adequate in heterogeneous environments and whether they should not be supplemented by other features.

4.9. Some Problems with the Protection Area Approach

The situation of the poorly described, heterogenous aquifer illustrates some of the limitations of a groundwater protection policy based on a protection areas alone:

- 1) It is inflexible and hard to change if, for example new technical information becomes available, which will modify the definition of the zone boundary.
- 2) It is s o u r c e orientated - that is the dimensions of the zone are based upon a source at a given location abstracting at a given rate. It does not protect the r e - s o u r c e as a whole and thus reserves for future exploitation may not be adequately protected.

- 3) It is based on prohibition in favour of water supply interests. There are other social and economic interests in the community and water supply cannot expect to take priority uncritically. In porous-permeable aquifers, in which the zones are relatively small, this issue does not arise to any great extent, because the balance between the various interests is regarded as acceptable. This is not necessarily the case in fissured aquifers where the zones may be much larger and more irregular and where they may be conservatively drawn to allow for data uncertainties. Other cultures, where the protection of water quality is not given a high priority, may not accept large protection zones.
- 4) Technical effort is directed towards defining multiple zones around sources regardless of whether there is any actual or potential quality threat. This could lead to a misdirection of skilled resources particularly in those countries where those resources are scarce.
- 5) The cost of any investigation falls upon the water supply agency. In situations where the problem is complex or the results ambiguous, it is difficult for the hydrogeologist to decide how far to pursue the problem and what budget to set for the study.

4.10. A Modified Approach

A system of groundwater protection is sought which

- I) is based on the concept that protection of groundwater is provided by aquifer residence time.
- II) uses the protection zone concept where appropriate.
- III) is applicable to heterogeneous fissured rock types.

- IV) provides protection for the total groundwater resources and not merely existing sources.
- V) makes the most efficient use of often scarce hydrogeological experts.
- VI) incorporates issues of stream/aquifer interactions.

To meet these objectives a three fold system is proposed.

4.11. Inner Protection Zone

The high priority to be given to the protection of existing potable water sources requires that an inner protection zone based on residence time be established. This would include residence times of up to 50-60 days and may be sub-divided depending on the national legislation.

4.12. Regional Protection Policy

The remainder of the area, including the zones of confined aquifers is divided into vulnerability classes which are defined to provide protection to the groundwater resource outside the inner protection zone. These vulnerability classes are based on various hydrogeological, soil and geological criteria and may be compiled from a combination of vulnerability maps of each of the variables in a manner which is now being developed in many different countries.

The characteristics which are generally recognised as affecting aquifer vulnerability are hydrogeological regime (water-table or confined), nature of the unsaturated zone, nature of the confining layer, thickness of unsaturated zone or depth to water table. From this information somewhat arbitrary distinctions must be drawn to establish a set of

vulnerability classes. The rate of transport through the unsaturated zone is likely to be the dominant factor. This can be characterised by the use of chemical tracers or tritium. Recent results on transport processes obtained from unsaturated zone studies, for instance into nitrate pollution, should be harnessed to improve the characterisation of aquifer vulnerability.

The value of these regional maps is that they characterise the total groundwater resource and they provide information for policy and planning purposes. If properly used they can encourage the development of potentially polluting practices in those areas where it will present least concern. They thus contribute practically to the search for a balance of interests, in that they allow water protection issues to be considered alongside other social and commercial factors influencing decision making. The regulation of diffuse pollution can be readily related to aquifer vulnerability zones. For instance if controls or restrictions on agricultural practice are to be instituted it is not reasonable to limit these only to source protection areas, otherwise potential future resources may be polluted before they are exploited. They can also include areas close to rivers which run adjacent to groundwater sources exploiting induced recharge.

4.13. Individual Investigation

The aquifer vulnerability map shows the regional picture but is not in sufficient detail or based on sufficient local factual information to demonstrate whether or not an individual potential pollution source is a threat to water resource interests. It provides a hierarchy of levels of concern but does not, of itself, provide protection to the aquifer. The protection comes through requiring scrutiny of all pro-

posals for potentially polluting practices on land and where necessary the threat to groundwater resources assessed by individual investigation. This investigation should ideally be at the financial cost of the person or organisation wishing to establish the potential pollution threat. The scope of the investigation will be determined not only by the degree of vulnerability and the complexity of the hydrogeology of the area but also by the nature of the threat. The need for investigation at the polluters expense provides a more realistic method of judging the cost/benefit aspects. This means that the most intensive investigations will be directed where the threat is greatest or where other social or economic pressures come closest into conflict with water resource interests. The assessment of the risk and the authorisation or refusal to proceed will depend both upon the findings of the hydrogeological study and the extent to which preventative measures can be built into the design to mitigate the pollution threat. The concept of residence time as a means of providing protection will be paramount in this assessment. The advantages of such an approach are:

- 1) Decisions are taken on the currently best available information and are not restricted by previously defined dimensions of protection areas which may, in the light of new knowledge, be too large or too small.
- 2) Technical resources are devoted to assessment of real cases of concern, and are thus more effectively used.
- 3) Decisions on the scope of the investigation and its cost are regulated by the potential value of the results to the investigation agency, which will normally be the organisation wishing to establish the potential hazard.
- 4) The approach offers particular advantages in respect of heterogeneous environments such as fissured strata. The

definitions of protection zones traditionally relies significantly on the use of tracers, which can be injected into the groundwater system at selected locations radial to a borehole and travel times measured. This approach is difficult to apply in highly heterogeneous aquifers since flow paths are unpredictable and positive breakthrough of tracer is necessary for a reliable confirmation of travel time. It may take extensive work to establish the shape of the protection area and even then there will be uncertainty. However if the investigation is related to one specific pollution threat at a known location then it is much easier to do realistic tracer work to determine limiting travel times to water supply sources, springs or rivers.

4.14. Conclusion

The traditional protection area strategy, although proved by practical experience to be effective in relatively homogeneous porous permeable aquifers, has limitations in other situations. The example of fissured aquifers, where the practical application of the approach is in doubt, demonstrates the need for a more comprehensive regional policy. It is suggested that this is built from existing practices of protection area definition and vulnerability mapping to provide a regional policy framework coupled with a greater emphasis on the study of individual pollution threats. This coupled approach, looking at the regional and site-specific aspects as part of one integrated exercise, attempts to achieve a realistic balance of community interests and deals with a wide range of hydrogeological situations. It should be emphasised that this proposal, like existing procedures, needs to be supported by administrative law and a system of enforcement. It relies upon the availability of hydrogeological skills and the continuing involvement of the groundwater specialist in these socially important topics. These factors need to be borne in mind when considering the application to developing countries.

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CHAPTER 5

PROBLEMES DE PROTECTION DE LA QUALITE DES EAUX SOUTERRAINES EN TERRAINS CALCAIRES

par

H. Paloc

5.1. Introduction

L'accroissement des besoins en eau en différents pays du Monde - accroissement particulièrement accentué ces dernières années tant dans les pays industrialisés que dans ceux où l'amélioration des conditions de vie est sous l'étroite dépendance de leurs possibilités d'approvisionnement en eau - conduit à devoir simultanément solliciter de nouvelles ressources de bonne qualité en augmentant le nombre et l'importance des points de captage tout en cherchant à réduire les nuisances découlant d'une occupation par l'homme, sans cesse accentuée, de l'espace naturel.

Les régions calcaires n'échappent pas à cette évolution: parce qu'elles recèlent parfois des sites privilégiés de captage (de très remarquables sources notamment) et, en même temps, parce qu'elles offrent souvent des caractéristiques géographiques et morphologiques spécifiques spécialement appréciées comme cadre de vie, elles sont de plus en plus fréquemment utilisées.

Toutefois, on peut se demander si ces utilisations diverses sont toujours compatibles et, notamment, si la vulnérabilité à la pollution que l'on a généralement coutume de souligner pour certains types de formations calcaires autorise à la fois la sauvegarde de la qualité des eaux captées et l'occu-

pation, à des fins diverses, des surfaces offertes par ces régions.

Il convient donc d'examiner d'une part si les conditions hydrogéologiques que l'on rencontre dans les terrains calcaires ne conduisent pas à nuancer quelque peu, ou pour le moins à préciser, la notion de risques de pollution de leurs eaux; et d'autre part, de quelles manières il est permis de concevoir selon ces conditions la protection d'un captage en cherchant à assurer dans le plus grand nombre de cas la permanence de la qualité des eaux livrées à la consommation.

5.2. Degrés Divers de Vulnérabilité à la Pollution des Réservoirs Aquifères Calcaires

Les conditions hydrogéologiques des roches calcaires peuvent être très variées:


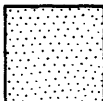

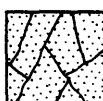


- En ce qui concerne la roche elle-même, il convient d'abord de faire la distinction entre les différentes catégories de faciès de roches calcaires qui ne se trouvent en fait regroupées au sein d'une même famille qu'en raison de leur forte teneur en carbonates (calcaires purs, marneux, gréseux, craies, tufs, marbres, dolomies, etc....). La roche peut être massive ou très stratifiée. Elle peut être poreuse (porosité matricielle) ou parfaitement étanche. Elle peut avoir été soumise à des contraintes tectoniques extrêmement diverses et son degré de fissuration est également très variable: les caractéristiques des fissures ainsi formées peuvent elles-mêmes être très différentes. Enfin, selon l'importance des actions d'altération auxquelles cette roche a pu être soumise, son degré de karstification est, lui aussi, très variable.

Ces conditions variées peuvent être regroupées en six classes qui se trouvent caractérisées par des paramètres hydrodynamiques, et donc par des degrés de vulnérabilité sensiblement différents (cf. figure 5.1). Leur différenciation sur le terrain, bien que parfois délicate en raison d'imbrications fréquentes entre ces classes qui ne se rencontrent pas toujours dans la nature avec des caractères aussi tranchés, doit constituer un premier objectif de connaissance et, spécialement, pour l'élaboration de cartes de vulnérabilité.

Au total, l'extrême hétérogénéité des paramètres aquifères de la roche ne peut avoir pour conséquence qu'une très grande diversité de comportement des réservoirs calcaires vis-à-vis de la pollution.

- En ce qui concerne le réservoir, il faut, là encore, constater que l'hétérogénéité peut être très grande. En effet, la localisation des couches aquifères peut être diverse au sein du système aquifère et sous la dépendance de conditions liées à la genèse de la roche (conditions sédimentologiques), à son âge (conditions stratigraphiques), aux contraintes qu'elle a subi (conditions structurales), aux rapports qu'elle a eu avec son environnement ancien (conditions paléogéographiques), au régime des eaux qu'elle recèle (conditions hydrologiques), à l'existence ou non et à la nature de produits de remplissage etc...

- En ce qui concerne l'environnement actuel du réservoir, l'environnement actuel du réservoir, il peut être constitué de formations géologiques d'extension, de puissance et de composition très diverses. En surface, le calcaire peut être nu ou plus ou moins masqué par des formations superficielles dont la nature et la répartition tantôt favorisent tantôt contrarient le processus de karstification de la roche sous-jacente.

		COEFFICIENT D'EMMAGASINEMENT (S°)		PERMEABILITE (K°)		DEGRE DE VULNERABILITE
		Mini	Maxi	Mini	Maxi	
MILIEU HOMOGENE		m	$< 10^{-6}$	$< 10^{-7}$ cm/s		milieu non aquifère, non vulnérable
		f	nul	nulle		
		c	nul	nulle		
		m	10^{-5} à 10^{-1}	10^{-6} à 10^{-4} cm/s		risques généralisés d'introduction de pollution avec effets de rétention, de fixation et de dispersion: faible probabilité de contamination d'un captage, ou, le cas échéant, à faible concentration mais de longue durée
		f	nul	nulle		
		c	nul	nulle		
MILIEU HETEROGENE		m	nul	nulle		risques localisés d'introduction de pollution avec propagation sans rétention notable: risques de contamination d'un captage peu éloigné
		f	10^{-4} à 10^{-2}	10^{-4} à 10^{-1} cm/s		
		c	nul	nulle		
		m	10^{-5} à 10^{-1}	10^{-6} à 10^{-4} cm/s		risques généralisés d'introduction de pollution avec effets de rétention: possibilité de contamination d'un captage peuvent être de longue durée et de concentration variable, en général peu élevés
		f	10^{-4} à 10^{-2}	10^{-4} à 10^{-1} cm/s		
		c	nul	nulle		
MILIEU SOUS RECOUVREMENT		m	nul	nulle		risques localisés, souvent ponctuels (orifices karstiques), d'introduction de pollution avec propagation très rapide et faible effet de rétention: risques importants de contamination d'un captage, même éloigné, à pic de concentration élevé mais à faible étalement
		f	10^{-4} à 10^{-2}	10^{-4} à 10^{-1} cm/s		
		c	10^{-5} à 10^{-3}	10^{-1} à 50 cm/s		
		m	10^{-5} à 10^{-1}	10^{-6} à 10^{-4} cm/s		risques généralisés d'introduction de pollution, aggravés ponctuellement (orifices karstiques) avec propagation très rapide et effets possibles de rétention: risques importants de contamination d'un captage même éloigné, à pic de concentration élevé et durée notable de restitution
		f	10^{-4} à 10^{-2}	10^{-4} à 10^{-1} cm/s		
		c	10^{-5} à 10^{-3}	10^{-1} à 50 cm/s		
Vulnérabilité indirecte, par soutirage, fonction des caractéristiques du recouvrement (r)						
<ul style="list-style-type: none">— si "r" perméable à très perméable, risques variables selon les paramètres du calcaire (cf. les différents cas ci-dessus)— si "r" imperméable à peu perméable, risques plus ou moins atténués, en général pas élevés, sauf en présence d'un karst sous-jacent à un recouvrement de faible épaisseur, ou en limite de recouvrement						

(°) S et K { m : de matrice
 f : de fissures et plans de discontinuité
 c : de cavités et conduits karstiques

Fig. 5.1.: Schematisation des différents degrés de vulnérabilité d'une roche calcaire en fonction de ses paramètres hydrauliques (coefficient d'emménagement-perméabilité)

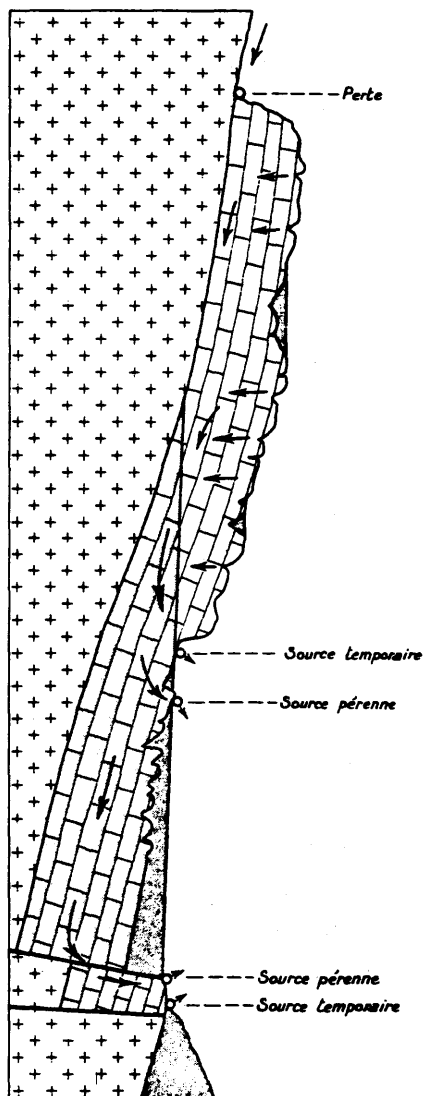
Les diverses conditions hydrogéologiques qui se rencontrent le plus souvent dans les régions calcaires se trouvent schématisées dans la figure 5.2.

En résumé, un système aquifère calcaire peut comporter selon les points de très grands contrastes dans la distribution de ses vides, dans la localisation de ses eaux et dans leurs conditions d'infiltration, d'écoulement et de restitution. Ainsi, les eaux évacuées par une source ou sollicitées par un captage ne sont, le plus souvent, que le produit d'un mélange, en proportions variables selon le régime, de volumes d'eau d'origines différentes dans le temps et dans l'espace: l'on doit s'attendre en conséquence à une notable variabilité dans la composition chimique et bactériologique des exutoires et des captages et dans les processus d'introduction, de propagation et d'élimination de toute substance véhiculée par l'eau.

5.3. Potabilité des Eaux des Réservoirs Aquifères Calcaires

Une notable difficulté apparaît dès lors qu'il s'agit de définir ou d'admettre la "potabilité" d'une eau, en raison d'une part de l'extrême diversité des substances qui peuvent s'y rencontrer, des combinaisons plus ou moins complexes de ses composants chimiques, des difficultés plus ou moins grandes de pouvoir identifier et doser certains d'entr'eux; et d'autre part des conditions souvent délicates d'appréciation des seuils admissibles et notamment pour des éléments susceptibles de se trouver dans les eaux à de très faibles concentrations (métaux lourds), conditions qu'il importe pourtant de bien connaître puisqu'il en découle le choix d'un traitement correctif approprié.

Ainsi, tout point de captage d'alimentation en eau potable devrait faire l'objet d'un suivi de la composition chimique



Caractères structuraux	Calcaire affleurant		Calcaire sous couverture
	en situation perchée	en situation noyée	
Eau	en condition de nappe libre (avec possibilité d'écoulements en charge)		en condition de nappe captive (avec possibilité de sécléurs à nappe libre)
Karstification du calcaire	sur toute son épaisseur	sur une partie de son épaisseur (parfois jusqu'à son mur)	lait karstifié lait non karstifié possible au sein de la roche
Remplissages ou terrains de couverture	discontinus		continus
Conditions hydrogéologiques			

Fig. 5.2.: Schematisation des conditions hydrogéologiques les plus fréquentes d'un réservoir aquifère calcaire en relation avec ses caractères structuraux

et bactériologique de l'eau captée dont le programme d'analyses et leur fréquence devraient être établis en fonction des modalités de captage, de son niveau d'utilisation, de la plus ou moins grande vulnérabilité du réservoir aquifère qu'il sollicite et des risques de contamination de ses eaux selon la nature et la localisation des foyers de pollutions dans le bassin hydrogéologique du captage.

Nous avons dit combien pouvaient être variables, dans le temps et dans l'espace, les caractéristiques hydrodynamiques, physico-chimiques et bactériologiques des écoulements karstiques; il en va de même en ce qui concerne les pollutions qui peuvent s'y manifester.

- Dans des conditions naturelles, c'est-à-dire en dehors de pollutions susceptibles d'être apportées par les activités humaines, certains éléments peuvent présenter des concentrations supérieures aux teneurs admissibles pour les eaux de consommation, soit de façon permanente, soit temporairement: ainsi, du point de vue chimique, bien que le type "carbonaté" prédomine le plus souvent dans les eaux des réservoirs calcaires, on constate que certaines d'entr'elles présentent des teneurs élevées en chlorures (notamment dans le cas des aquifères situés en bordure de mer et pour la plupart des sources sous-marines), en sulfates (traduisant souvent la présence de formations à évaporites dans le réservoir lui-même ou au contact du réservoir), en nitrates (liés par exemple à la présence de lignites ou de tourbières), etc... La "dureté" elle-même peut parfois se révéler excessive qui constitue alors une contrainte à l'exploitation.

On sait, d'autre part, que beaucoup de sources karstiques manifestent un trouble de l'eau plus ou moins intense, plus ou moins épisodique et plus ou moins durable, qui découle du transport de matières en suspension (argiles, limons, sables calcaires ou siliceux).

Il s'agit bien, dans tous ces cas, de véritables pollutions "naturelles" qui peuvent conduire à la nécessité de mise en oeuvre de traitements correctifs pour permettre l'utilisation de l'eau.

- En dehors des conditions naturelles, la potabilité d'une eau souterraine peut se trouver affectée à des degrés divers par l'introduction dans le réservoir aquifère de toutes sortes de substances en conséquence plus ou moins directe -et plus ou moins subie- des activités exercées par l'homme dans une région donnée. Certaines de ces substances, en raison de leur nature et de leur concentration, présentent des risques sérieux de pollution encore qu'elles puissent ne se manifester aux points de captage qu'avec des concentrations inférieures aux normes admissibles. Il convient donc de faire la distinction entre le risque potentiel que représente telle ou telle activité humaine vis-à-vis de la pollution, et le risque réel qui peut en découler au lieu de prélèvement; entre "causes" et "effets" interviennent en fait les conditions, très variables nous l'avons vu, d'écoulement des eaux dans le réservoir qui conduisent à des possibilités de dilution très différentes.

Les hydrogéologues le savent bien qui ont eu maintes et maintes fois l'occasion de constater la diversité des résultats d'expériences de traçage, tant en ce qui concerne la durée des temps de séjour que les modalités de restitution et de dilution d'un traceur selon le type de réservoir, selon la nature du point d'injection, selon la période hydrologique, selon le nombre et les caractéristiques des points de réapparition, ... mais aussi en fonction du traceur choisi.

5.4. Protection des Réservoirs Aquifères Calcaires et de leurs Sites de Captage

1.) Au plan réglementaire, les problèmes posés par la protection des eaux destinées à l'alimentation en eau potable ont depuis longtemps - mais spécialement depuis quelques décennies - préoccupés à juste titre les services officiels de la plupart des états afin d'assurer, préalablement à tout dommage, la sauvegarde de la santé publique suivant en cela la maxime: "mieux vaut prévenir que guérir".

Ces préoccupations ont conduit la plupart d'entr'eux à élaborer une réglementation sanitaire sans cesse améliorée au fur et à mesure qu'apparaissaient des problèmes nouveaux.

Le principe de cette protection "a priori" des points de captage repose sur l'établissement de périmètres dont les extensions et limites respectives sont fonction de la plus ou moins grande vitesse de propagation d'une éventuelle pollution. En France, on a retenu une différenciation en trois périmètres:

- un périmètre immédiat, établi autour du point de captage pour éviter les risques de contamination directe: au sein de ce périmètre qui doit être acquis en pleine propriété par la collectivité, toute activité se trouve interdite.
- un périmètre rapproché, à l'intérieur duquel peuvent être interdits ou réglementés toutes activités et tous dépôts ou installations de nature à nuire directement ou indirectement à la qualité des eaux.
- un périmètre éloigné, établi le cas échéant, et à l'intérieur duquel peuvent être réglementés les acti-

vités, installations et dépôts ci-dessus visés (il est bon de remarquer que les captages d'eaux superficielles ne donnent pas lieu à l'établissement de ce périmètre).

C'est à un hydrogéologue, agissant dans le cadre d'une mission officielle (en France, pour le compte du Ministère de la Santé) qu'il appartient de proposer ces périmètres en prescrivant des mesures de protection préventives qui se traduiront pour chacun d'eux par des degrés différents de servitudes dont les plus rigoureuses pourront ouvrir droit à indemnisation.

2.) Au plan pratique, c'est en tenant compte d'une part des différents degrés de vulnérabilité de la roche calcaire elle-même et de ses formations éventuelles de recouvrement, et d'autre part des relations, vitesses de transit et temps de séjour révélés par traçage, que l'hydrogéologue délimite, au sein du système aquifère auquel se rattache le captage, des zones de sensibilité différentes, et propose, pour chacune d'elles, les mesures de protection qui lui paraissent les plus appropriées.

En de nombreux cas, l'étendue des systèmes aquifères calcaires, le nombre parfois élevé de points de prélèvement de tous types qui les affectent, la situation variable des foyers de contamination et la diversité de leur caractère polluant, doivent inciter à procéder de façon globale pour assurer la protection des eaux captées. Intervenir coup par coup constitue en effet une difficulté technique compte tenu des délais requis pour l'exécution rationnelle d'un programme de traçages et pour déterminer, en tenant compte de ses résultats, les limites et l'importance respectives des bassins drainés par chaque captage: problème toujours délicat dans les aquifères karstiques en raison des possibilités de diffluence avec des conditions de liaisons variant en

fonction de l'état du remplissage du réservoir. Par ailleurs, une intervention globale conduit presque toujours à réaliser une économie très sensible par rapport aux moyens financiers qui devraient être consacrés, en procédant cas par cas, par les collectivités.

Un intérêt supplémentaire de l'approche globale est de permettre de proposer ou d'éliminer des sites possibles de captage selon leur plus ou moins grande dépendance avec des secteurs dont les degrés de vulnérabilité auront été jugés plus ou moins compatibles avec les risques de pollution auxquels ils se trouvent exposés:

- dans les secteurs plus ou moins vulnérables mais les moins exposés à des risques de pollution, et dont les captages ne révèlent pas d'altération sensible de leur qualité, on s'efforcera de faire appliquer des mesures de sauvegarde pour qu'il n'y ait pas aggravation de ces risques ou, si ce devait être le cas, pour que celle-ci soit compatible avec le maintien de la qualité de l'eau du site de captage.
- dans les secteurs plus ou moins vulnérables mais déjà contaminés et dont les captages révèlent une altération sensible, permanente ou épisodique, de leur qualité, on s'efforcera:
- soit de ramener, dans toute la mesure du possible si c'est techniquement et économiquement envisageable, la pollution à un taux compatible avec l'amélioration et le maintien de la qualité de l'eau du site de captage,
- soit d'orienter vers les secteurs les plus fortement compromis le développement de certaines activités avec, pour corollaire, un risque d'accroissement de pollutions ce qui pourrait alors exiger, dans le meilleur cas, la

mise en oeuvre d'un traitement de l'eau du site de captage, et, dans le plus mauvais, son abandon. Toutefois, en certains sites, une modification des modalités de captage ou des conditions de prélèvements pourra entraîner une amélioration de la qualité: approfondissement de l'ouvrage pour atteindre des niveaux moins contaminés, diminution des rabattements pour limiter la zone d'appel du captage, relèvement de seuil ou serrement pour réduire la part d'eau contaminée issue de niveaux plus profonds, etc....

Enfin, les mesures préconisées devront être très étroitement adaptées aux caractères spécifiques de toute substance polluante et à la connaissance de son comportement dans le terrain, soit dans le calcaire lui-même, soit dans ses éventuelles formations de recouvrement. En effet, si la pollution bactériologique est apparue longtemps comme étant la plus fréquente et la plus dangereuse dans les terrains karstiques par rapport aux autres aquifères en raison d'absence de filtration, on constate aujourd'hui que des substances chimiques, de composition et de solubilité très diverses, sont de plus en plus introduites dans le milieu naturel, substances qui exigent pour être éliminées des procédés de traitement plus ou moins complexes et de coût élevé: on se préoccupera donc de n'imposer que celles des contraintes qui, tout en étant compatibles avec le souci que soient maintenues sinon développées certaines activités, seront indispensables à la sauvegarde de la qualité des eaux captées.

Dans un autre ordre d'idée, on ne saurait perdre de vue la difficulté de faire appliquer efficacement des mesures de protection en certaines régions karstiques: des moyens suffisants en matériel et en personnel sont indispensables pour assurer le contrôle minimal sans lequel les populations pourraient ne pas accorder à l'opportunité de ces mesures un crédit suffisant.

5.5. Conclusion

Les caractères hétérogènes à bien des égards des réservoirs aquifères calcaires et les incertitudes qui en résultent souvent quant à une connaissance rigoureuse des processus de transfert des substances polluantes et, conjointement, les conséquences économiques qui pourraient découler d'une imposition trop excessive de servitudes sur de très grandes surfaces font que la réglementation de protection doit être adaptée aux conditions de vulnérabilité et de risques de contamination de chaque site de captage: en effet, pour chacun d'eux apparaît un certain nombre de particularités dont la mise en évidence doit être favorisée par une étude préalable de l'ensemble du système aquifère, étude à l'issue de laquelle des solutions différentes peuvent être proposées.

Toutefois, malgré les améliorations qui pourront être apportées à la connaissance d'un réservoir aquifère calcaire, il y aura toujours conflit d'idées quant à l'importance à accorder à la protection: "aussi grande que possible pour accroître son efficacité et aussi faible que possible pour réduire les servitudes", ainsi que le soulignait le Professeur H. SCHOELLER (Colloque d'Orléans - 1977).

Il s'agit donc de tenter de concilier les besoins de consommation de l'homme avec les besoins de protection de son environnement et de trouver un équilibre entre impératifs économiques et nécessités écologiques: à cet égard, il ne faut pas sous-estimer les difficultés qui demeureront ou qui s'accroîtront dans certaines régions calcaires et notamment dans les zones où s'intensifieront l'occupation et l'utilisation des sols à quelque fin que ce soit: le choix sera alors nécessaire, à moins de compromettre l'aménagement de

de l'espace et faute de pouvoir préserver intégralement la qualité de l'eau, entre la mise en oeuvre d'installations de traitement adaptées à la spécificité des eaux du captage et des substances susceptibles de transiter dans le réservoir aquifère, et toute autre solution de rechange s'il en existe: ce sont alors les critères de meilleure qualité et de moindre coût qui devraient déterminer la décision.

PART III

PRACTICE OF GROUNDWATER PROTECTION ZONES

CHAPTER 6

OVERVIEW OF THE PROTECTION OF GROUNDWATER QUALITY

by

H.G. van Waegeningh

The protection of the quality of groundwater is a very complex matter, as it relates not only to groundwater but also to soil and surface water; not only to quality but also to quantity available; not only to hydrogeology but also to a number of other technically-oriented sciences. Moreover, it sets a series of restrictions on society in general and on individuals; so socio-economic circumstances are relevant.

This list could be continued in detail. It will be clear, taking into account the important role of groundwater in drinking water supply, the extensive hazards to groundwater and the case histories of severe pollution, that a solution for the protection of groundwater has to be found as soon as possible. This remark relates as much to industrialized countries, which already have problems, as to developing countries which need to prevent groundwater pollution. In view of the complexity it seems desirable to restrict the subject to the protection of groundwater quality destined for use as drinking water, only one facet of the management of groundwater.

Protection comprises the application of several technical sciences into the framework of existing legislation and administration, and so doing, taking into account the socio-economical circumstances. Systems for the protection of the groundwater quality have to offer practical and realistic solutions. In practice hydrogeology plays a leading role.

The most important related sciences are chemistry and microbiology. The practising hydrogeologist is in a good position to solve the conflict between pollution hazards and groundwater quality, if he keeps contacts with other interests and disciplines, e.g. agriculturists, chemists, water supply engineers, physical planners and administrators.

Soil and groundwater protection set a number of restrictions to land use and human activity; several interests have to be reconciled. On the one hand the protection areas have to be as large as necessary for the safety of water supply, on the other hand they have to be as small as possible. Taking into account the balancing of interests, and the behaviour of contaminants, protection areas with special restrictions can be limited to only a part of the entire aquifer recharge area around a wellfield; a zoning within that area seems sensible too. A system of protection of the whole recharge area with ad-hoc decisions introduces the risk of inconsistency, and so of insufficient safety of water-supply. Bearing in mind that protection should be preventive, it could be said, as a variation on a well-know saying.

Protection is the Solution for Pollution

Using a coordinated multidisciplinary and multi-interest tackling of the groundwater protection problem, an approach already exists in many countries in Europe. In some cases this is incorporated into legislation. Because of the different socio-economical circumstances and legislative arrangements in different countries, it is difficult to compare their protection systems. However, to start discussion a tentative table (Table 6.1.) of protection systems is given, with an indication of the restrictions applying in different zones, and an indication for their size.

Tab. 6.1: Tentative comparison of protection areas.

Prohibitions	Fed. Rep. Germany	Austria	Belgium	Finland	The Netherlands	France	Switzerland	Czechoslovakia	Hungary	Sweden	Great Britain
Only Water supply activities allowed	Zone I Well field 10 m	Immediate Protection area	Immediate Protection zone	Intake area 7 7 7 7	Well field	Immediate protection (10 & 20 m)	zone I 10 — 20 m	First sanitary production zone 10 — 50 m	Protection zone	Well area	Immediate non-statutory protection area (normally 10-50 m)
Prohibition on building x Agricultural restrictions	Zone II	Protection area	100 m 24 hrs. Inner protection area	Inner protection zone	Catchment area	Inner protection area	zone II	Internal second sanitary protection zone		Inner protection area	no fixed zones but groundwater protection achieved through consultation under statutory development planning and environmental control
	50 days	50 days	(300-1000 m) 50 days	60 days	(> 30 m) 50 & 60 days)	7 7 7 7	10 days > 100 m	7 7 7 7	50 days 7 7 7	> 60 days > 100 m	procedures with priority to aquifer recharge areas (Vulnerability classifications may be used as tools in this process)
Restrictions on certain industries, storage and transport of certain chemicals and oil	Zone III A	Partial protection area	Remote protection area	Outer protection zone	Protection area 10 years delay	Remote protection area	zone III > 200 m	External second sanitary protection zone	hydro geological protection area	Outer protection area	
	2 km				Protection area 25 years delay		zone A				
	Zone III B				Far recharge area		zone B		25-100 year delay regional protection		
OUTER BOUNDARY OF RECHARGE AREA											

A system that has been adopted in some cases is that of the Federal Republic of Germany. Various important criteria are used for dimensioning protection zones. From experience it was concluded that pathogens are eliminated after an aquifer residence of, on average, fifty days. Thus zone II is bounded by a line of equal delay time of fifty days. The restrictions to be maintained relate especially to the protection against activities spreading pathogens and rapidly degradable chemicals.

Z o n e I I I A, which surrounds zone II, reaches to a distance of two kilometers at maximum. This distance originates from past experience in the industry, that no pollution effects had been found originating at larger distances. Within zone IIIA restrictions on the storage and handling of hardly degradable chemical compounds are in force. For the outer parts of the recharge area (zone IIIB) the same general restrictions are in force, but they are less rigorously maintained.

Because of the great importance of these criteria, two large multidisciplinary research programmes have been started in the Federal Republic of Germany; one on the survival of bacteria and viruses in aquifers and the other on the behaviour of chemical compounds in the underground. It is expected that the conclusions and recommendations of these studies will be of importance in the solution of the protection problems. Other important research programmes, expected to have interesting results for protection, are also going on. As important tools in protection monitoring networks and vulnerability atlases have to be mentioned.

It is remarkable that, as protection systems have been developed, little distinction has been made between different hydrogeological systems. From the standpoint of equal treatment of population and common legislation this may be

attractive, but the differences in hydrogeological properties of aquifers may be very large. Basically one can distinguish three hydrogeological systems:

- Porous permeable aquifers, which are normally unconsolidated. Their flow is intergranular, and this type of aquifer is ideal for mathematical estimation of flow regimes. There is an intensive contact between rock matrix and (polluted) groundwater, so that physico-chemical reactions can take place.
- Fissured, consolidated rocks, where groundwater flow mainly takes place through fissures, and may be turbulent. Because of lack of contact with the rock matrix, attenuation processes will be relatively limited and there are less possibilities for mathematical approach.
- Karstic aquifers where the flow takes place through the solution channels, which are irregular in dimension and direction. Here also there is little contact between rock matrix and (polluted) groundwater. In general there are no possibilities for mathematical approaches.

Depending on the various properties of the aquifer, e.g. hydraulic gradient, transmissivity, and porosity, the two last mentioned types of aquifers may have very high groundwater flow velocities. Whereas, in porous permeable rocks velocities will be in the order of 10 - 200 m per year, in fissured and karstic rocks velocities up to 27000 m per day have been measured. Flow velocity and the possibility for attenuation processes are important factors for the sizing of protection zones; other important features are the influence of the unsaturated zone and the effects of semipermeable layers.

A remark still has to be made on the sources of pollution hazard. A distinction can be made between point and diffuse sources of pollution; this distinction is not only important for the estimation of the possible effects, but also in viewing possibilities for regulation.

Point sources cause plumes of polluted groundwater downstream, which are of limited dimension. The detection of these plumes, and the control of associated hazards is rather difficult; if unpolluted water is sampled from a monitoring well, it does not prove that no pollution has taken place; a plume of polluted water may be passing just a metre below the wellscreen.

With diffuse sources of pollution contamination occurs more or less homogeneously over large areas; it can be caused by a single source, for example the misuse of manure and fertilizers in agriculture, or by a large number of different point sources together, as for example may occur in a city. Another is acid rain or dry deposition from air pollution.

Both types of pollution have different effects on groundwater, and have to be monitored with specific systems.

It will be clear that the solution for each type of pollution source will be different; for example point sources often relate to one or few individuals, and can be handled with specific laws (e.g. landfills), while diffuse sources mostly relate to the general development of the entire region (e.g. agriculture).

Besides these man-made sources of pollution, there are also other causes of pollution, e.g. mineral addition to groundwater from geological formations, intrusion of salt water from the sea. The concept of protection of groundwater quality does not include these features.

It is certain that much research still has to be done before one can draw firm conclusions in the science of "contaminant hydrogeology", and to give soundly justified recommendations for protection practices. However, if we do not start to protect groundwater today, it will be too late.

Hydrogeologists handling protection problems in practice will also have to deal with the legislative aspects. Legislation offers several possibilities to regulate groundwater protection:

- Adaption to groundwater protection of several specific existing laws, which are meant for quite other purposes, e.g. mining acts, laws on transport and storage of dangerous goods, laws on waste disposal. In this way only certain facets of the protection problem can be regulated; a satisfactory solution will not be found. Where no specific legislation exists, action on physical planning and nuisance law are very useful tools.
- The best solution, of course, will be a law on soil and groundwater protection to be included in a water management act. Only a few countries have such specific legislation.

To solve the problem of groundwater protection, the Commission for Groundwater Protection of the International Association of Hydrogeologists could stimulate the hydrogeologist:

- to describe satisfactory technical systems for protection, based as much as possible on the knowledge of contaminant hydrogeology;

- to communicate with other sciences and interests to exchange knowledge and experience, and to balance all interests;
- to communicate with legislators on the possibilities of groundwater protection legislation, and to provide technical guidelines for protection on which legislation can be based.

CHAPTER 7

GROUNDWATER POLLUTION PROTECTION IN DEVELOPING COUNTRIES

by

S.S.D. Foster

7.1. Introduction

7.1.1. Significance of Groundwater Supplies

Groundwater is being widely and increasingly exploited for potable water-supply in developing countries, especially in smaller towns and rural areas, because it is normally the cheapest, safest source. In many regions, development now involves very large numbers of low-yielding (0.5-5 l/s) boreholes, often drilled on an uncontrolled basis, tapping unconfined (water-table) aquifers and providing untreated, unmonitored, and often unreticulated, supplies. Shallower dug wells, of traditional design but improved completion, continue to be constructed under some conditions.

Much higher-yielding (10-100 l/s) boreholes are also quite widely developed to provide piped supplies in many larger towns, including such urban major conurbations as Mexico DF, Lima, Bangkok, Cairo, Manila and Djakarta. Even in such cases, raw water surveillance and treatment are often limited and intermittent.

7.1.2. Situation on Groundwater Protection

In view of the significance of groundwater supplies, aquifer protection to prevent degradation of potable groundwater quality ought to be of paramount importance in many developing countries. This especially because of the increasingly widespread practice of on-site disposal of untreated domestic and industrial effluents to the ground, due to prohibitive cost of more satisfactory handling and disposal. Moreover, the rehabilitation of polluted aquifers will generally be expensive, protracted and even impracticable.

Some hydrogeological environments virtually unique to tropical countries (notably coastal and island limestones of Tertiary/Quaternary geological age), appear to be exceptionally vulnerable to many polluting activities, because of their thin soils, shallow groundwater table, high transmissivity aquifers and often exceptional rates of rainfall infiltration.

Groundwater protection has, however, not yet received much attention. Related legislative provisions are also rare. Moreover, the state of groundwater quality in most major potable water-supply aquifers in the developing countries is poorly described and the controls on this quality impartially understood.

In view of generally-limited institutional resources and prevailing complacency in this context, the most urgent need is a consistent and relatively simple technique for rapid assessment, or classification, of groundwater pollution risk. Such a classification would serve to identify and focus attention upon those areas most at risk and to act as the basic framework within which measures for pollution protection or control could be specified and implemented.

Hereafter, only the protection of groundwater against pollution is discussed and such important issues as natural quality problems aggravated by over-abstraction, saline water encroachment, recharge reduction, excavation for mining or quarrying, are not considered. In many developing countries effluents are discharged directly to surface watercourses without treatment, and in the dry season with little or no dilution. In arid regions especially, such watercourses will frequently be influent with respect to shallow aquifers and as such constitute a serious but indirect pollution threat, which is also not considered further here.

In any discussion of groundwater protection the fundamental need for sound sanitary completion of groundwater sources to prevent direct ingress of surface drainage should not be forgotten. This is the commonest cause of serious microbial pollution of water supplies in developing countries, especially where they are not reticulated and the population and their livestock have direct access to the wellhead.

A reasonably comprehensive list of potentially-polluting activities is presented (Table 7.1.). Many of those activities producing serious groundwater pollution risks in the industrialised countries are present to lesser degree in similar form in some parts of the developing world. They include oil and chemical spillages, mining processes, sewer leakage, aerial deposition, landfill waste disposal, intensive livestock practices, highway drainage. The most important risks require more detailed discussion.

7.2. Principal Groundwater Pollution Risks

7.2.1. Unsewered Sanitation

Improvements in sanitation are extremely widely and very urgently needed in many developing countries, as was defined

Tab. 7.1: Categorisation of activities causing groundwater pollution risk.

Activity causing Pollution Risk (*relative importance in developing countries)	Characteristics of Activity	Pollutant Characteristics	
		(bio)chemical type (s)	hydraulic surcharge
(1) URBANISATION			
(a) unsewered sanitation **	u/r P-D	F,N,µO,s	+(+)
(b) leaking sewers ^a	φ u P-D	µO,f,o,n	+
(c) unlined sewage oxidation lagoons ^a *	u/r P	F,µO,O,n,s	++(+)
(d) sewage land discharge ^a **	φ u/r P-D	F,N,µO,O,s	++(+)
(e) sewage discharge to influent rivers ^a *	φ u/r P	N,µO,o,f,s	+++
(g) leaching refuse landfill/tips ^a	u/r P	n,s,µo,o,m,f	
(h) aerial deposition ^a	φ u/r D-RG	s,µo,m	
(i) highway drainage soakaways	u/r P-D	s,µo	+
(2) INDUSTRIAL DEVELOPMENT			
(a) leaking storage tanks/pipelines ^b	φ u P-D	O,µO,s,m	+
(b) chemical spillages	φ u P-D	µO,o,m,s	+
(c) unlined process water/effluent lagoons *	u P	µO,O,m,s	++(+)
(d) effluent land discharge **	φ u P-D	O,µO,M,s	++(+)
(e) effluent discharge to influent rivers *	φ u P	µO,O,m,s	+++
(f) leaching residue tips	u/r P	µO,M,O,s	
(3) AGRICULTURAL PRACTICE ^c			
(a) soil cultivation			
(i) with agrochemicals *	r RG	N,µo,s	
(ii) with agrochemicals + irrigation **	r RG	N,µO,S	+
(iii) with sewage sludge/animal slurry	r D	N,µO,s,m	
(iv) with wastewater irrigation *	r D	N,µO,s,m,f	+
(b) livestock rearing/crop processing			
(i) unlined effluent lagoons	r P	F,O,N,µO,s	++(+)
(ii) land discharge of effluent/residues *	φ r P-D	O,N,s,µo,f	(++)
(iii) effluent disposal to influent rivers	φ r P	O,n,f,s,µo	+++
(4) MINERAL EXTRACTION			
(a) hydraulic disturbance *	r/u P-RG	s,m	
(b) drainage water discharge *	r/u P-RG	M,s	+++
(c) unlined process water/sludge lagoons *	r/u P	M,S	+(++)
(d) leaching residue tips	r/u P	S,m	

- a can include industrial components
 b can also occur in non-industrial areas
 c intensification presents main pollution risk
 φ activity frequently uncontrolled
 u/r urban, rural
 P/D/RG point source/diffuse source/regional effect
 n nutrient compounds
 f fecal pathogens
 µo micro-organic compounds
 o organic load causing elimination of dissolved oxygen,
 mobilisation of trace metals and inorganics
 s salinity, major inorganics
 m trace metals and inorganics
 (capital letters indicated major risk)

for the UN International Drinking Water-Supply & Sanitation Decade. It has become accepted that unsewered (on-site) sanitation (such as ventilated and pour-flush pit latrines) can provide adequate service levels for excreta disposal in rural areas, villages, small towns, and even in parts of larger urban areas, at much lower cost than main sewerage systems. As a result of this policy, sooner or later, an enormous expansion of excreta disposal into the ground will occur, especially in many Asian countries where much of the population are currently without any form of sanitation.

Current knowledge of the migration and attenuation of the associated water pollutants is derived largely from investigations, in North America and Europe, on the effectiveness of septic tank soakaways. It is important to recognise that there are significant differences between septic tanks and pit latrines including:

- a) Septic tank soakaways discharge at significantly higher levels in the soil profile than pit latrines, where conditions are more favourable for pathogen elimination.
- b) The hydraulic surcharge from septic tank soakaways is normally designed not to exceed about 30 mm/d, whereas substantially higher rates of surcharging (around 100 mm/d) may occur from pour-flush latrines.
- c) Septic tanks are mainly restricted to low-density urban or rural settlements, whereas latrines are being installed at very high density at many locations in developing countries.
- d) Septic tanks are lined and their solid effluent, of high nitrogen content, is periodically removed, while in the case of pit latrines it may remain in the ground.

The water laws and codes of practice of many countries make reference to a minimum acceptable spacing between excreta disposal unit and potable groundwater source of 15 m (50 feet), under favourable (hydrogeological) site conditions. This criterion appears to have been selected rather arbitrarily from the results of some painstaking, but limited, early research. Some countries have selected a much larger distance (for example, 200 m (220 yards)) in Malawi water law). There is considerable pressure to reduce this permitted spacing to as little as 5 m in some developing countries, such as Bangladesh and parts of India and Sri Lanka. This often results from lack of space in very densely populated settlements, but can also occur in more prosperous and well organised urbanisations served by latrine sanitation, with the tendency for individuals to construct private dugwells or tubewells to replace, or to augment, properly-sited communal groundwater sources.

It has long been recognised that the natural soil profile can be an effective medium for the purification of human wastes, including the elimination of fecal microbes, the adsorption, breakdown or immobilisation of many chemicals. However, not all soil profiles (and their associated hydrogeological environments) are equally effective in this respect, and under certain conditions there is risk of the direct migration of pathogenic microbes, especially viruses, to neighbouring groundwater sources. Contamination of groundwater supplies by unsewered sanitation has been the proven vector of pathogen transmission in numerous disease outbreaks in developing countries.

The nitrogen compounds in excreta do not represent as immediate a hazard to groundwater but can cause widespread and persistent problems. An indication of the potential

groundwater nitrate pollution from on-site excreta disposal units can be derived from the following considerations: a population density of 100 persons/ha represents a discharge of 500 kg/ha/a to the ground which, if oxidised and leached by 100 mm/a of infiltrating rainfall, would result in the local groundwater recharge containing 500 mg $\text{NO}_3\text{-N/l}$ (2.214 mg $\text{NO}_3\text{/l}$). Dilution and reduction due to various processes will occur but, nevertheless, it must be expected that unsewered sanitation schemes will cause quite widespread groundwater nitrate problems even in relatively humid climates. There will be major buildup of nitrate in groundwater of arid areas without significant regional aquifer flow.

If sullage waters are also discharged to latrines, this will lead, in the long-term, to risk of groundwater contamination by organic micropollutants, in view of the progressive spread of household products and community chemicals containing an increasing range and concentration of synthetic organic compounds.

Numbers of measures can be considered to reduce the risk, or the scale, of groundwater pollution from on-site excreta disposal schemes, where they can be technically and economically justified. However, under the most unfavourable hydrogeological conditions, the two low-cost techniques, potable unreticulated groundwater supplies and unsewered on-site sanitation, may be incompatible.

7.2.2. Agricultural Land-Use

The impact of modern agricultural land-use and management practices on groundwater quality became fully apparent in Europe and North America during the latter part of the 1970s. In particular, high rates of leaching of nitrate and other mobile ions, from many soils under continuous arable

cultivation sustained by large applications of inorganic fertilisers, were demonstrated.

The use of inorganic fertilisers in the developing countries is now expanding rapidly with efforts to secure self-sufficiency in food production. Numerous cultivated soils will, in result, change from being almost invariably nutrient-deficient (except where moisture limits crop growth) to having intermittent nutrient excess. It is not yet established which soil conditions, climatic regimes and cropping systems will be the more vulnerable to nutrient leaching, although the aforementioned coastal limestone regions, are likely, amongst others, to be very sensitive in this respect.

Where a significant or major part of crop moisture requirements are provided by irrigation, as will be the case in many developing country situations, there is possibility of closely-controlling soil drainage and infiltration, and therefore nutrient leaching losses. However, this will require detailed understanding of soil water regimes, specific crop selection, and more advanced irrigation water management and technology, which will take many years to effect practice at field level, and in the meantime there is risk of major nutrient leaching especially from the thinner, coarser-textured soils.

Agricultural land-use practices can present further problems for groundwater quality including:

- a) Saline infiltration during the initial habilitation and subsequent management of irrigated soils by application of excess water.
- b) Increasing groundwater salinity and nitrate concentrati-

ons, and possibly organic micropollutant levels, resulting from irrigation with (stabilised) wastewater, especially where this is the major source of soil moisture.

- c) Increasing nitrate, chloride and possibly trace element concentrations consequent upon excessive land applications of sewage effluent/sludge or animal slurry.

In overall terms, the use of increasing numbers and quantities of pesticides has not led to major and widespread groundwater contamination in Europe and North America, at least as far as is presently understood. There are, however, significant exceptions, especially the more soluble and weakly-absorbed products (such as aldecarb, its related compounds and their metabolites) which have given rise to serious groundwater pollution locally.

In the developing countries, growth in the use of pesticides for agricultural cropping is closely following that of inorganic fertilisers. Geohydrological environments with shallow water-table and coarse-textured soils of low organic content, (such as the coastal limestones and some alluvial deposits) would appear vulnerable to some pesticide leaching to groundwater. Concern must be expressed about the use of weakly-absorbed compounds of significant solubility in these environments, especially where irrigation is practised, even if their quoted biodegradation rate (in a fertile aerobic soil) is relatively rapid.

7.2.3. Urban and Industrial Effluents

In many developing countries, extensive sectors of urban areas are unsewered. In these areas increasing numbers of, mainly small-scale, industries (such as textiles, metal pro-

cessing, vehicle maintenance, paper making) are located, often on a highly dispersed basis. Most of these industries generate small quantities of liquid effluent (such as spent oils and solvents). In the absence of directives or controls these will generally be discharged to the soil, primarily because of the cost of such alternatives as on-site treatment or secure storage and transport to safe disposal sites. Bigger industrial plants generating large volumes of process water will also commonly have unlined surface impoundments for the handling or concentration of liquid effluents.

With increasing availability and complexity of synthetic chemicals being used in all these enterprises, some activities may represent a serious long-term threat to local groundwater quality in respect of variety of constituents.

In those urban areas with main sewerage systems, a favoured economical method of sewage treatment, if any is considered or practised, is wastewater stabilisation by retention in shallow unlined oxidation lagoons, prior to discharge into rivers or the ground or to reuse for irrigation. These lagoons may have high rates of seepage loss, especially after initial construction and subsequent cleaning, and if so, will also have considerable impact on local groundwater quality, especially in relation to trace organic compounds.

7.3. Selected Examples of Field Conditions

7.3.1. General Background

It is considered that the introduction of some examples will help to focus on the special circumstances pertaining in developing countries in relation to groundwater supply, pollution risk and protection policy. The three examples selected come one from each continent, Africa, Asia and South

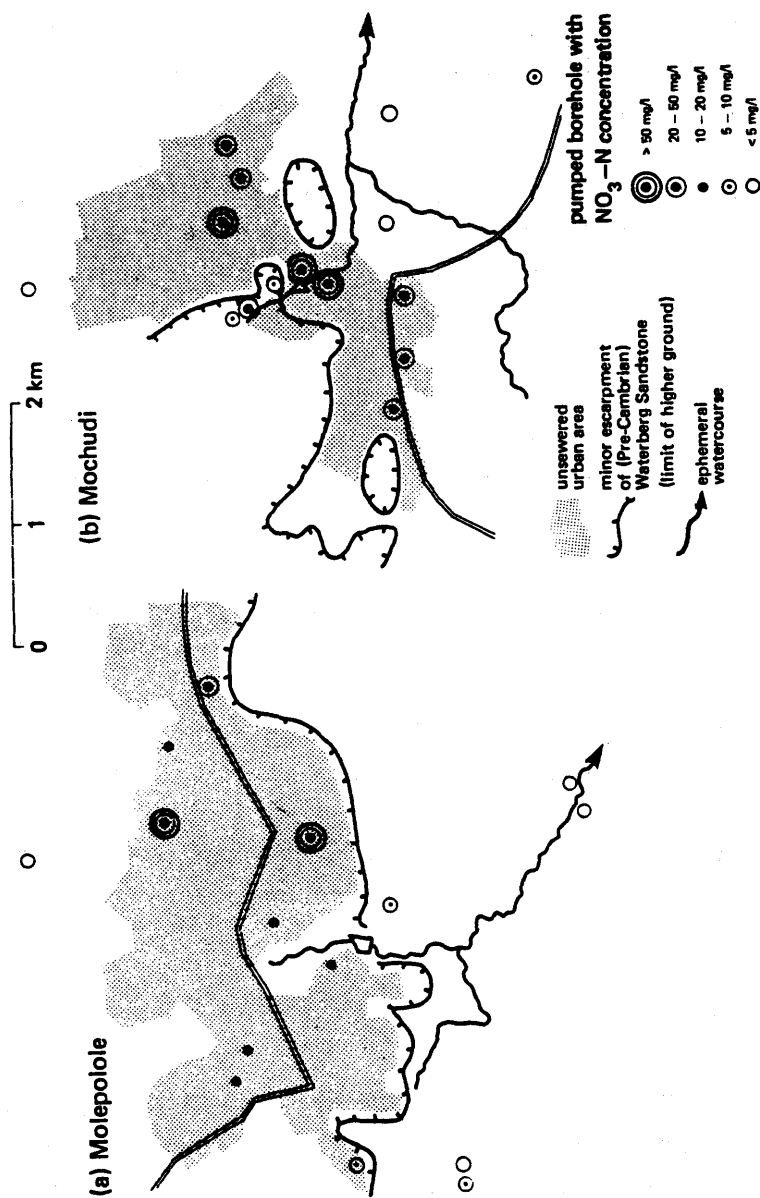


Fig. 7.1: Groundwater nitrate concentrations in water-supply boreholes of two Botswana towns (1976)

America, and cover a wide range of groundwater conditions and of economic development. They are not claimed to be representative, nor is their description comprehensive.

Various technical and economic factors prevailing in many developing countries lead the scheme of groundwater development to be significantly different from that in most industrialised countries in certain respects:

- a) Groundwater sources will generally be sited as close as possible to individual demand locations because of the pressing need to eliminate or minimise reticulation costs.
- b) This results in very large numbers of small, unmonitored and untreated sources (owned and/or operated by individuals or small communities) predominating over municipal wellfields.
- c) Near surface aquifers with shallow groundwater tables are preferred to reduce development and operating costs.
- d) Rapid rates of growth in the numbers of water-supply boreholes and in borehole pumping rates are commonplace.

These factors are extremely pertinent when considering the most practicable approach to groundwater pollution protection.

7.3.2. Botswana Towns

Molepolole and Mochudi (Fig. 7.1.) are so-called traditional villages situated in a sparsely populated region on the fringe of the Kalahari in eastern Botswana. The population of each has grown from less than 10,000 in 1971 to about

20,000 in 1981. There is very little industrial activity and most people seasonally cultivate some surrounding land (without irrigation) and rear livestock over much larger areas.

Minor aquifers are developed in weathered and/or fissured zones of the Pre-Cambrian Waterberg Sandstone and underlying Metamorphic Basement Complex of low transmissivity (less than $20 \text{ m}^2/\text{d}$) and relatively shallow groundwater table (generally at 5-20 m depth). During the last 40 years, their groundwater resources have been developed by low-yielding boreholes, which provide unreticulated, untreated and unmonitored supplies. The average annual rainfall is about 500 mm/a, and with thin soils and frequent bedrock exposures, some diffuse groundwater recharge occurs fairly regularly in most wet seasons. However, low aquifer transmissivity and the generally flat terrain mean that there is little regional groundwater flow.

Ventilated-dry pit latrines have been progressively introduced by sanitation programmes in the 1960s and 1970s. With population densities up to 100 capita/ha, latrines generally occur at densities of up to 10 capita/ha, and at distances of down to 15 m from water-supply boreholes. During the latter half of the 1970s, very high groundwater nitrate concentrations were discovered in many of the potable water-supply boreholes in numerous Botswana villages, including Molepolole and Mochudi (Fig. 7.1.). These, together with significant fecal contamination of some supplies, have been attributed to the installation of unsewered sanitation. The problem is being solved, at quite considerable cost and in some cases technical difficulty, by drilling substitute water-supply boreholes outside the populated area for the worst-affected sources, and reticulating to standpipes in the towns.

7.3.3. Jaffna Peninsula, Sri Lanka

The Jaffna Peninsula is a low flat area of about 800 km² in the extreme north of Sri Lanka. In the island's dry zone, it is characterised by very high temperatures and evaporation rates all the year round, with an average rainfall of some 1000 mm/a, most of which occurs in the Maha season, associated with the north-east monsoon. It is entirely composed of Miocene Limestone, which varies from a honeycombed mass of coral material to a massive cemented rock with interbedded coral debris, mantled only by a thin layer of soil, generally less than 1 m thick. The soils are characteristically light-textured and well drained, with high terminal infiltration capacity (over 5.0 m/d) and available moistures of only about 20 %. There is thus no surface water, except flood channels and ephemeral ponds. Depth to groundwater table varies mainly in the range 2-10 m, depending on season and location.

The population of Jaffna Town is put at a little over 100,000 with a very high and rapidly growing rural population density in many parts of the peninsula. Everywhere there are large numbers of dug wells, rarely exceeding 10 m depth. Estimates of the numbers vary widely but it is likely that there are more than 10,000 domestic wells, used for potable supply and watering (so-called) home gardens, and probably a similar number of irrigation wells. At Kondavil and Thirunevely (Fig. 7.2.), groundwater has been developed for reticulated public supply, but the proportion of the population receiving mains water, either direct to the house or via external standpipes, is small.

The urban areas (Fig. 7.2.) are virtually unsewered. For public buildings and more prosperous dwellings, domestic effluents are discharged to the ground via septic tanks and

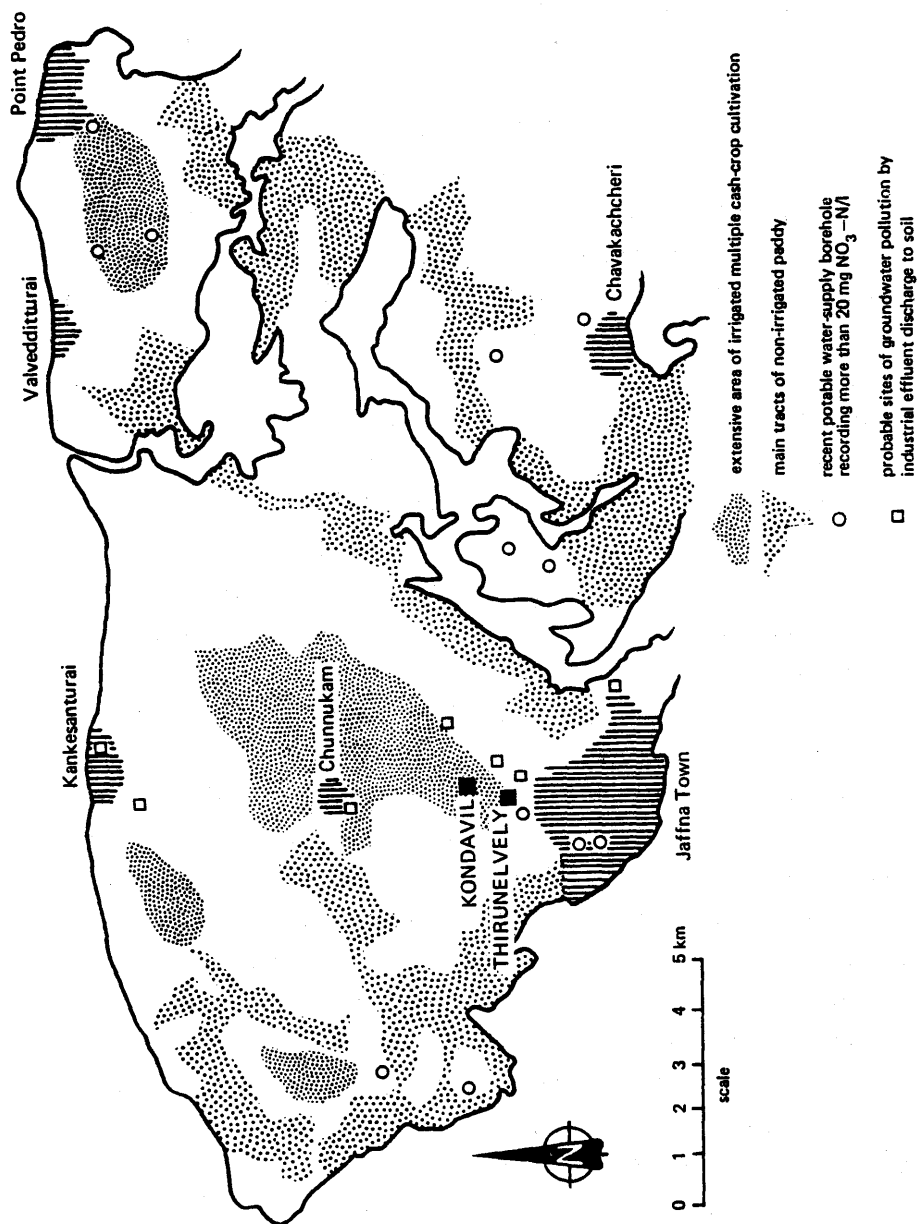


Fig. 7.2: Generalized land-use in the western part of the Jaffna peninsula, Sri Lanka

during the 1970s there has been a major campaign for household pour-flush latrine installation. These latrines are typically 2.0 m deep and may be situated at distances down to less than 10 m from potable water-supply wells. They are believed to be the cause of the widespread microbiological (fecal coliform) contamination and are thought to have been the cause of a number of incidents of pathogen transmission.

Shallow groundwaters quite widely contain in excess of 20 mg NO₃-N/l (88,5 mg NO₃/l) and locally 30-50 mg NO₃-N/l (133 - 221 mg NO₃/l). Latrines will contribute to this increasing groundwater nitrate problem, but it is suspected that the major source is irrigated cultivated land. Parts of the Jaffna Peninsula are amongst the most intensively cultivated and highest yielding land in Asia (Fig. 7.2.). During the 1970s, major increases in chilli, onion and other cash-crop yields have been achieved, with intensification to double or triple cropping by the introduction of inorganic fertilisers, pesticides and the use of increasing quantities of groundwater for irrigation. Fortnightly fertiliser applications, totalling 80 kg N/ha/crop, coupled with a similar quantity in the form of organic manure applied on seeding are now commonplace. With existing forms of basin and channel irrigation (on a weekly schedule outside of the wet season), losses by deep infiltration are relatively high on the thin permeable soils. It is believed that these irrigation return waters are primarily responsible for the usually very high groundwater nitrate concentrations, and the sometimes high ammonium concentrations (more than 5 mg NH₄-N/l = more than 5,7 mg NH₄/l), that are observed. There must also be a significant risk of migration of any weakly-absorbed pesticides to groundwater. A further problem results from the build-up of salinity in the shallow groundwater in years of below average monsoon rainfall and associated groundwater recharge.

The urban areas of the peninsula also have significant numbers of small-scale industries, some of whose effluent disposal may constitute a threat to groundwater quality unless it is properly controlled (Fig. 7.2.).

7.3.4. Metropolitan Lima, Peru

Lima is an exceedingly rapidly growing city, with present population approaching 5.0 million, situated in one of the world's driest regions. With little or no precipitation, virtually all fresh water in the area originates in a series of transverse rivers whose flow derives from tropical rainstorms and snowmelt in the immediate hinterland of the Andean Cordillera. The sediments of the alluvial system in the associated valleys are of the alluvial braided plain and alluvial fan type and commonly form important, highly-permeable, aquifers, recharged by the rivers themselves, by seepage losses from irrigation canals, from excess irrigation, and by leakage from water services in urban areas.

Groundwater represents a major source of potable water-supply. Some 250 municipal production boreholes (Fig. 7.3.) provide a supply in excess of 6000 l/s and tap unconfined and semiconfined alluvial aquifers, with widely falling groundwater table generally at depths considerably in excess of 10 m. Most of the borehole supplies are reticulated to private connections throughout the longer-established urban areas and to some of the newer suburbs, the remainder are used for tanker supplies to areas without water reticulation.

All of the longer-standing urban areas and some of the newer suburbs are sewered and the water-supply boreholes will, as a result of rapid urban development, generally be located within 100 m of a sewer line. The hydrogeological conditions are, in general, relatively favourable for the self-elimination of microbiological and biodegradable chemical contami-

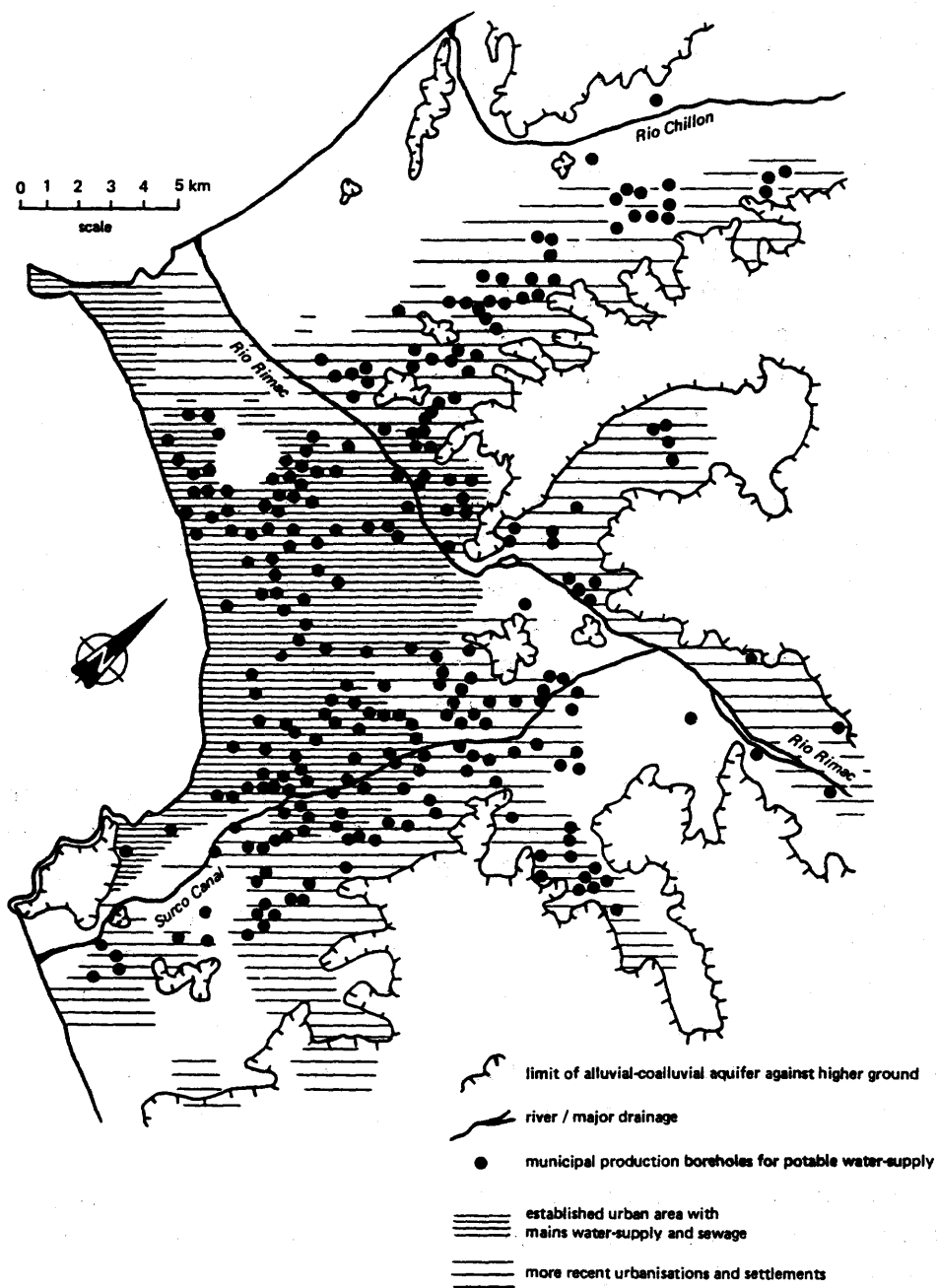


Fig. 7.3: Distribution of potable groundwater supply sources in the Lima metropolitan area, Peru

nants, although there is no detailed groundwater quality data available to substantiate the picture. Some small-scale industries in recently-settled unsewered areas may be discharging effluent to the ground, but in other areas most industrial effluents are believed to be discharged via the sewerage system or to surface watercourses. Wastewater recycling for agricultural and amenity irrigation is rapidly increasing. In a proposed major scheme, certain large out-fall sewers will be diverted to unlined stabilisation lagoons of 200 ha area on the fringe of the alluvial areas and (after 10 or more days retention) wastewater will be used to bring 5000 ha of neighbouring desert land into agricultural production. Elsewhere small sewerage systems may discharge into unengineered lagoons. Such activities constitute a significant risk to local groundwater quality and may require systematic investigation and control.

7.4. Strategy for Pollution Risk Assessment and Protection Policy

7.4.1. General Approach

One policy option for groundwater pollution protection would be to attempt to impose varying levels of appropriate restrictions in a series of "protection zones" surrounding individual water-supply sources with groundwater delay (or residence) times, with respect to those sources, of from 50 days up to many years. Such source-oriented policies are best suited to the more uniform aquifers exploited by a relatively small, fixed number of high-yielding, municipal, water-supply boreholes with stable pumping regimes. Despite the administrative convenience and legislative simplicity of the protection zone approach, it cannot be readily and widely applied in developing countries because of:

- a) The frequently very large and rapidly growing number of sources, which renders consideration of individual sources and the establishment of fixed zones, especially those of large residence time, impracticable.
- b) Data deficiencies and technical uncertainties, especially in heterogenous aquifers, which make the estimation of required dimensions of protection zones, (particularly those of longer residence time), problematic or inadequate without intensive field work.

Protection zones around individual groundwater supply sources can, thus, play only a limited role in the overall protection policy. A possible exception will be in sparsely populated regions where protection zones could be applied conservatively without producing serious conflict with other interests. The protection zone concept could also usefully be incorporated into a broader-based groundwater pollution control policy in the form of a) minimum separations (corresponding to given hydrogeoloical environments), between groundwater supply source and excreta disposal unit, for microbiological protection and b) dilution zones of modified land-use to alleviate the impact of unavoidably polluting land-use activities.

A more comprehensive, flexible, and universally-applicable policy to groundwater protection is, however, needed. The approach should be aquifer-oriented and activity-related.

7.4.2. Technical Basis for Pollution Risk Assessment

A relatively rapid assessment of groundwater pollution risk requires the classification or evaluation of the following two distinct components and consideration of their interactions:

- a) Pollutant loading on the subsurface environment generated by the given activity, the way in which this load is applied to the soil, unsaturated zone or aquifer and the associated fluid or hydraulic loading.
- b) Aquifer pollution vulnerability in respect of (i) delay time for access of mobile pollutants under natural hydraulic loading, (ii) capacity for physicochemical reaction of less mobile (inorganic and organic) pollutants, (iii) overall dilution potential of hydrogeological environment.

Such assessment would serve to focus attention on the higher risk areas and activities, and to provide a logical framework within which pollution control measures could be selected or to designate priorities for groundwater field investigation and quality monitoring. This approach is also a sounder basis from which to confront the problem of dispersed source pollution, which, in the longer run, may prove a more serious threat to groundwater quality than that associated with identifiable point sources.

The first criterion for pollution vulnerability - delay time for access to the saturated zone of mobile (soluble, non-reactive weakly-absorbed) pollutants - would be estimated approximately on a logarithmic scale, from less than about 20 days to more than 50 years, on the basis of a) the thickness and character of any confining beds or of the unsaturated zone, b) the average rate of diffuse rainfall recharge.

The criterion relates primarily to the risk of contamination by pathogenic microbes and by degradable (mobile) organic micro-pollutants, but indirectly also to the likelihood of

significant filtration, sorption or dispersion. The other criteria would be rated on a qualitative basis, for example to indicate areas with negligible regional groundwater flow and limited inorganic adsorption or organic absorption capacity, and relate to the dilution and attenuation of persistent pollutants respectively.

7.4.3. Formulation and Implementation of Protection Policy

Reconnaissance mapping of aquifer pollution vulnerability on a regional basis, at scales of 1:500 000 or larger may be required for overall national planning, but evaluation of aquifer pollution vulnerability for operational protection purposes would concentrate on those more densely-populated or intensively-utilised areas with significant use of groundwater for potable supply. Except in areas of very uniform hydrogeological conditions, classification on a map basis at a scale of 1:50 000 or 1:100 000 would probably be most convenient, although numerous problems of data presentation have to be resolved and no existing system is considered adequate in all respects.

The classification of pollutant and hydraulic loading associated with an existing or proposed activity, such as urban sanitation, industrial areas, intensive agriculture, mining processes, etc., can be undertaken by the categorisation and ranking of various elements of the activity. In the case of industries, for example, categorisation of the quantity, solubility, toxicity and persistence of the effluent is required, together with effluent handling and disposal practice.

The type of groundwater pollution protection measures feasible will vary with the polluting activity. In relation to unsewered sanitation schemes, aquifer vulnerability should

be capable of being interpreted in terms of compatibility with the use of groundwater for potable supply, in relation to possible contamination by either pathogenic microbes or nitrates, of the need for modification to the design of excreta disposal units and of the preferred separation between these units and potable groundwater sources. The latter is equivalent to the innermost protection zone, calculated on a residence time of 20, to 50 or more, days as further scientific research finds appropriate. It would, however, be evaluated primarily on the basis of the delay time for access to the saturated zone and only secondarily in respect of groundwater flow in the saturated zone, and its dimension could range from a sensible minimum of perhaps 5 m, up to the practicable maximum of perhaps 200 m.

Similarly in relation to agricultural land-use, aquifer vulnerability should be capable of interpretation to indicate those areas most vulnerable to serious groundwater contamination by leaching of some nutrients and salts, and certain pesticides, and thus to suggest investigation priorities. Where confirmed, measures involving some control over crop selection, irrigation, fertilisation and other practices would have to be considered, perhaps through a system of subsidy or compensation.

The zones most vulnerable to pollution by soil discharge of industrial effluents, should also be apparent and the need for restrictions can be assessed. Accidental oil and chemical spillages also represent a significant threat in zones of high groundwater pollution vulnerability. An inventory is required of all sites at which certain ('black-list') chemicals are manufactured, stored or used, with enforcement of appropriate controls to minimise the risk of soil discharge. Such installations as wastewater stabilisation lagoons, main sewer lines, storage tanks, and landfill solid waste dispo-

sal are theoretically containable, but in practice frequently leak and represent significant hazards. They should not normally be permitted in zones of high groundwater pollution vulnerability used for potably supply, without special precautions.

In those circumstances where alternative water supplies are readily available, it may be considered politic to allow groundwater quality degradation, but appropriate restrictions should be enforced on the use of groundwater for potable supply.

At present, substantial difficulties will arise with the implementation of any groundwater protection policy:

- a) In many developing countries there is shortage of personnel with appropriate training in environmental engineering and in hydrogeology, and experience in groundwater pollution problems, to undertake the pollutant loading and aquifer vulnerability evaluations, and work would be hampered by unavailability of relevant field data.
- b) The level of intersectorial consultation and collaboration required will not be easily attained and the legal reinforcement needed in relation to some aspects may be difficult to formulate or to ratify.
- c) As discussed herewith, the current state of scientific knowledge is such as to limit the ability to interpret the scale of groundwater pollution risk associated with some activities and the effectiveness of some possible remedial measures. This will tend to result in conservative interpretations, which may disproportionately prejudice other social or economic interests and produce

7.4.4. Scientific Limitations

7.4.4.1. Aquifer Flow Regimes

Certain aspects of groundwater flow, important in the evaluation of pollution vulnerability, often prove problematic to estimate and expensive to investigate in a given field situation, notably:

- a) The scale of hydraulic heterogeneity in non-uniform formations, and especially in fissured aquifers.
- b) Actual mean groundwater flow rates (and residence times), which (even for radial flow) may depart widely from the values derived from simple, analytical equations or digital models, given for example, the presence of 'permeability layering'.
- c) Groundwater migration rates and residence times when a component of vertical saturated flow is induced as a result of wellscreen settings.
- d) The magnitude of dispersion and its effect on the first arrival time of mobile pollutants and on the peak concentration after a given residence time.

7.4.4.2. Groundwater Pollutant Behaviour

Similar can be said of many aspects of pollutant behaviour in groundwater systems, notably:

- a) The residence time, and other factors, for the effective elimination of pathogenic bacteria; survivals of more than 200 days have been demonstrated, 50 days is commonly used as the basis for the innermost microbiological protection zone but rarely, if ever, has more than 20 days been proved in actual cases of disease transmission.

- b) Whether it is justified to assume similar behaviour for pathogenic viruses as for the indicator bacteria, bearing in mind the somewhat different processes controlling their survival and migration, and differences in migration and persistence of the common indicator bacteria themselves.
- c) The complex behaviour of pollutants exhibiting immiscibility and/or density effects.
- d) The rates of biodegradation of organic micropollutants, which may be highly variable even for various isomers of the same compound and also for differing groundwater pH and Eh conditions.
- e) The degree of retention of organic micropollutants by absorption/adsorption mechanisms in formations of low organic content, as opposed to the soil profile.
- f) The mobility of some trace elements, such as Cr^{6+} and As^{3+} , in certain aquifer types and under certain groundwater conditions.
- g) The differing rates of migration of various pollutants in fissured porous ('double porosity') aquifers, dependent solely on variations in aqueous diffusion coefficient and in fissure flow velocity.
- h) The rates of natural dissolved oxygen consumption and denitrification in groundwater systems.

7.4.4.3. Unsaturated Zone

- a) The unsaturated zone (including the soil profile) represents the first, and by far the most important, line of defence against aquifer pollution. This is because of the generally low water velocities in this zone, the fact

that water movement is normally concentrated in the smaller pores with large specific surface and its overall generally aerobic condition.

- b) It is thus important that the unsaturated zone is taken into consideration for the assessment of aquifer pollution vulnerability. Should it be ignored, evaluations will be excessively conservative. However, water and pollutant movement in the unsaturated zone are complex and thus its role in pollution protection requires careful consideration.
- c) The unsaturated vertical hydraulic conductivity is a function of prevailing pore-water tension and moisture content. In the case of heterogeneous formations, and especially of fissured rocks, the variation can be dramatic, since the fissures can only contain and conduct water at very low tensions. Thus, whilst natural water flow rates in the unsaturated zone of most formations generally do not exceed 0.3 m/d, even in the short term, under conditions of artificial hydraulic surcharging, and sometimes of exceptionally high intensity infiltrating rainfall, they may be very much higher.
- d) It is generally reasonable to assume that under conditions of natural rainfall infiltration, transit or residence time (for a given unsaturated zone thickness) is a function of the annual infiltration rate and an average moisture content approaching the specific retention (Fig. 7.4.). Since the latter varies little amongst soil and rock types compared with the climatic variations of the former, the unsaturated zone transit time under these conditions is essentially controlled by infiltration regime and, therefore, climatic type. Exceptions will occur in lower permeability soil profiles where a significant proportion of the excess rainfall may be rejected as sur-

face run-off and if very high intensity infiltration intermittently occurs.

- e) An indication of the sensitivity of the unsaturated zone transit time to hydraulic loading (or surcharging) may be obtained by assuming that, under conditions of gross surcharging, it is a function of effective porosity and saturated in situ vertical hydraulic conductivity (Fig. 7.4.). In practice this case will rarely be reached under field conditions, but it is clear that in all soil and rock types, other than fine-grained unconsolidated sediments, gross sensitivity may be exhibited and unsaturated zone transit time radically reduced. The degree of hydraulic loading associated with any source of pollution is thus a key factor when considering the pollution protection afforded by the unsaturated zone.
- f) Moreover, when we consider pathogen elimination and biodegradation of organic micropollutants, the effective soil thickness below the depth of effluent discharge (Fig. 7.5.) is another key factor, since the proportion of organic carbon and the level of biological activity is by far higher in this upper part of the unsaturated zone. Losses of potential pollutants by volatisation will also decrease rapidly with depth below surface of the discharge point and with increasing hydraulic surcharge.
- g) A further problem arises in fissured porous rocks where interaction by diffusive exchange between mobile fissure water and matrix pore water is a very important process controlling the rate of downward migration of miscible water pollutants. This process is sensitive to aqueous diffusion coefficient, to fissure flow velocity and to porous-media geometry. Under certain flow conditions,

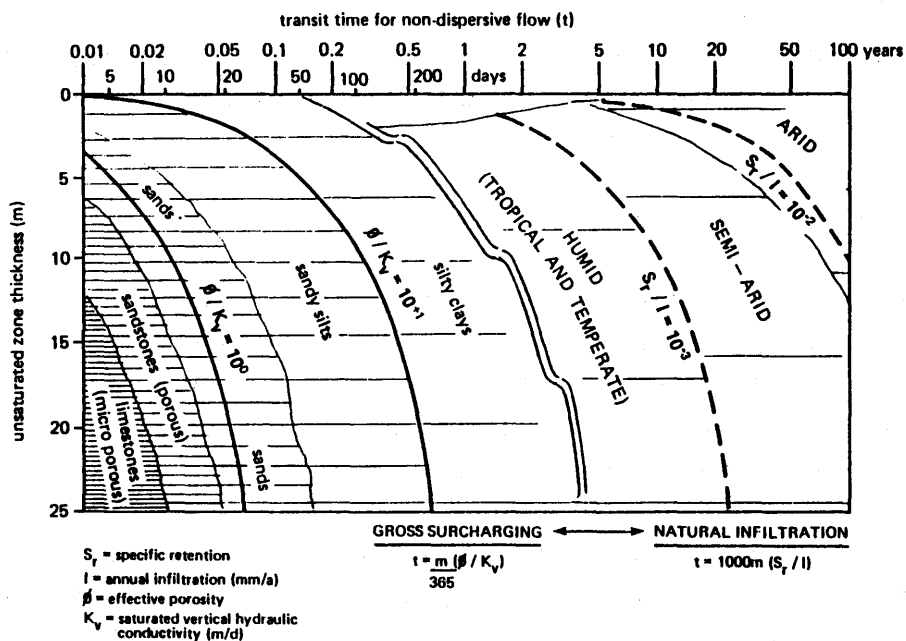


Fig. 7.4: Highly simplified estimation of unsaturated zone transit times

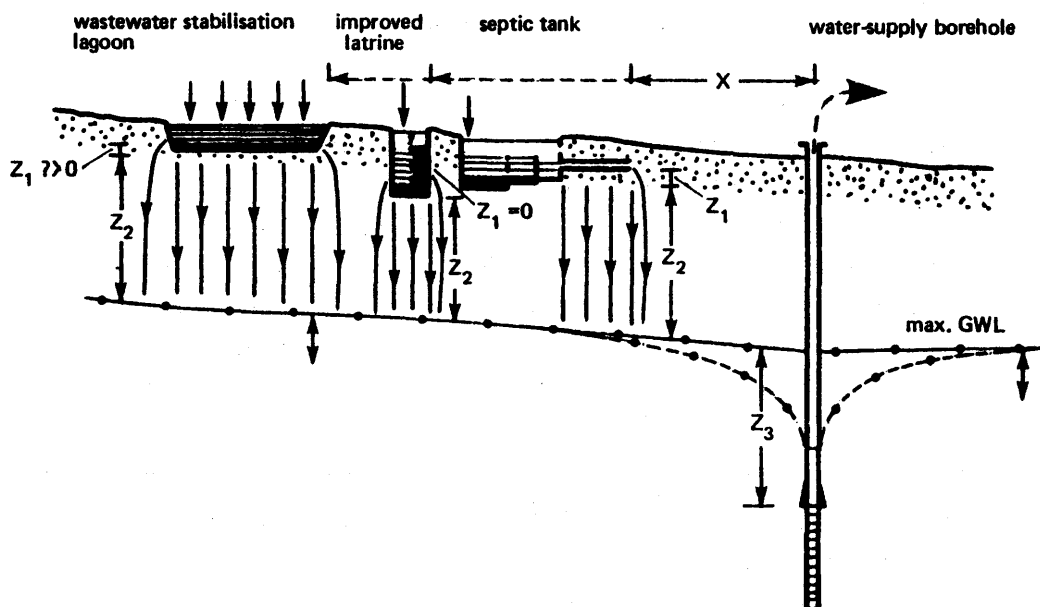


Fig. 7.5: Groundwater fecal pollution risk from common sanitary installations: key dimensions

pollutants with low aqueous diffusion coefficients, and microbial and immiscible organic pollutants, will migrate more rapidly than readily-diffusing soluble species.

7.4.4.4. Standards for Water Potability

- a) In most instances a groundwater protection policy will not be able, nor will be intended, to prevent all contamination. Thus the question of how much contamination is tolerable arises. The standards for water potability are, in effect, the design criteria for the case of groundwater pollution protection. In some cases, notably amongst the organic micropollutants, the corresponding limits are not yet precisely defined because of inadequate medical evidence. In others, the existing WHO recommendations are increasingly questioned as being unnecessarily stringent for adoption in developing countries, because of the disproportionate cost of attaining such standards in relation to other public health risks. Two particularly relevant parameters are the microbiological quality standard and that for nitrate.
- b) For the former, the appropriate current WHO standard relates to the pathogenic indicator organisms and stipulates that a potable supply should not regularly exceed 10 TC/100 ml and never exceed 0 FC/100 ml. Some public health specialists consider that this should be revised to allow from 10 to 50 FC/100 ml in small untreated unreticulated groundwater supplies and the relevance of the TC count is also contested.
- c) In relation to nitrate, the WHO recommended limit for potable supplies in tropical countries is currently set at 10.0 mg $\text{NO}_3\text{-N/l}$ (44 mg $\text{NO}_3\text{/l}$), but many argue that concentrations up to 22.6 mg $\text{NO}_3\text{-N/l}$ (100 mg $\text{NO}_3\text{/l}$) are permissible under most circumstances.

- d) Potable water quality standards and targets for developing countries require amplification and qualification so that they can be viewed on a cost-benefit basis in relation to improvements in other areas of public health.

7.5. Conclusions

Substantial resources and considerable efforts continue to be put into groundwater development for potable water-supply in many regions of the developing countries. In view of the characteristics of groundwater systems, this investment needs to be protected by implementation of realistic groundwater pollution control policies.

In many areas the groundwater pollution risks associated with unsewered sanitation schemes, changing agricultural land-use and management practices, some urban and industrial effluent storage and disposal, will be more important than those other activities of major concern in the industrialised world.

In some areas serious deterioration of groundwater quality has been proven and in numerous others significant groundwater pollution risks may exist and need to be quantified.

The sparsity and/or unreliability of groundwater quality data, and the frequent lack of analytical surveillance of potable water supplies, prevent a comprehensive appraisal of the current groundwater quality situation internationally. Such data, however, would not in any case be adequate to indicate the current groundwater pollution risk because of the very slow groundwater flow rates and pollution response times of most aquifers.

In consequence of the style of groundwater development, the nature of the principal pollution threats, and the variety of hydrogeological conditions involved, it is not realistic or practicable to adopt source-oriented protection zones as the main thrust of aquifer pollution protection policy in developing country situations.

A consistent, detailed, procedure for the rapid assessment of groundwater pollution risk needs to be drawn-up, under the patronage of the international organisations, from increasing experience and costly mistakes in the industrialised countries. It should be based on the classification of pollutant and hydraulic loading from specific activities and of aquifer pollution vulnerability and developed to cover the presentation of results, and the formulation and implementation of pollution protection policy in relation to such activities.

Evaluation and classification of aquifer pollution vulnerability based on delay time for access of mobile pollutants, capacity for physicochemical reaction of less mobile pollutants and overall dilution potential of hydrogeological environment is required.

In this context, the role of the unsaturated zone will be most important in unconfined aquifers, but such factors as the sensitivity of unsaturated zone behaviour to hydraulic surcharging and the distinction between the role of the soil profile and the remainder of the unsaturated zone in pollutant retention and degradation need careful consideration.

Such classifications of aquifer pollution vulnerability can be used to frame groundwater protection policy specifically in relation to each potentially-polluting activity.

Implementation of these policies will involve such measures as some degree of land-use and development control, restriction on the use or discharge of certain 'black-list' chemicals, and as such will require extensive intersectorial consultation and collaboration, with legal reinforcement as appropriate.

Limitations of scientific knowledge and shortage of experienced personnel will produce difficulties in the formulation and implementation of any type of groundwater pollution protection policy in many developing countries.

Intensive investigations and/or careful monitoring of groundwater quality in selected field situations in developing countries needs to be promoted, so as to improve knowledge of, and to focus attention upon, potential pollution problems.

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No bibliography is given, since the contribution is based upon the author's overall knowledge of aquifer pollution and of groundwater conditions in developing countries, but extensive reference was made to two major composite texts:

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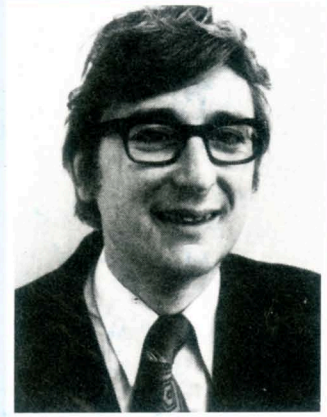
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The International Association of Hydrogeologists (IAH), which has established a Commission for Groundwater Protection in 1979, constituted in the same year a Working Group „Drinking Water Protection Areas in Humid Climates“ to compare the various national groundwater protection systems and to define criteria for a scientific based system for groundwater protection zones. This publication includes the papers presented at the IAH/UNESCO workshop at Koblenz in September 1, 1983 which was held in the framework of the International Hydrological Programme (Project A.3.7.).

This volume published with financial support of UNESCO is the result of international cooperation of groundwater scientists. It includes three parts:

Part I 2 chapters dealing with the theoretical background of protection zones

Part II 3 chapters dealing with groundwater protection in different aquifer types

Part III 2 chapters dealing with the practice of groundwater protection zones

This publication, partly written in English and partly in French, summarizes the scientific background and the practical experience and knowledge on protection zones for water supply systems.

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