

## EXPERIMENTAL RESEARCHES OF CONTAMINANT TRANSPORT ACROSS THE CAPILLARITY FRINGE

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*In this study we are interested in observations of water flow and solute transport within the capillary fringe (CF) and exchanges between the CF and the region below the water-table. A transport experiment of water and nitrates in a homogeneous soil was developed and the transfers between saturated and CF regions was measured. The capillary fringe defines the interface between the vadose zone and the groundwater where the water rises by capillarity due to surface tension. Water and nitrates fluxes to the groundwater due to infiltration could be influenced by the capillary fringe with respect to the physical and chemical conditions. Experimental investigations were conducted in order to observe the development of the capillary fringe under the condition of varying groundwater table position, and to study the flow and nitrates behavior within the capillary fringe.*

**Keywords:** groundwater, capillary fringe, mass fluxes, nitrates

### 1. Introduction

In an unconfined aquifer the upper boundary is the groundwater table. The position of the groundwater table is usually defined as the location with atmospheric pressure (zero relative pressure). The domain above the groundwater table is the capillary zone. In principle the capillary zone is hydraulically unsaturated by the simultaneous presence of water and air in a control volume. However, part of the capillary zone can still be hydraulically saturated (saturated capillary fringe) (Fig. 1a).

A substantial body of literature exists that deals with the role of capillary fringe (CF) as an interface between the vadose zone and the saturated zone below the water table [1]. CF may significantly affect the evolution of fluid flow and solute transport from the vadose zone to the saturated zone below the water table.

Except near regions of discharge to surface water bodies, fluid flow and solute transport in the sub-surfaces is commonly conceptualized as primarily vertical in the unsaturated zone. If fluid pressures are measured above the water table, they will be found to be negative with respect to local atmospheric pressure.

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Capillary pores in the vadose zone can draw up water from beneath the water table below which the pores are saturated with water. The zone above the water table, in which the pores are saturated is termed capillary fringe.

In a capillary tube water molecules at the water table subject to an upward attraction due to surface tension of the air-water interface and the molecular attraction of the liquid and the solid phases.

If a pore space could be idealized to represent a capillary tube, the capillary rise  $h_c$  can be derived from equilibrium between surface tension of water and the weight of water raised. Thus:

$$h_c = \frac{2\sigma}{r\gamma} \cos \alpha \quad (1)$$

where  $\sigma$  is surface tension,  $\gamma$  is the specific weight of water,  $r$  is the tube radius, and  $\alpha$  is the angle of contact between the meniscus and the wall of the tube. For pure water in a clean glass  $\alpha = 0$ , and at  $20^\circ\text{C}$ ,  $\sigma = 0.074\text{g/cm}$ ,  $\gamma = 1\text{g/cm}^3$ , so for  $r$  in cm, the capillary rise (in cm) is

$$h_c(\text{cm}) = \frac{0.15}{r(\text{cm})} \quad (2)$$

Assuming uniform spherical grains of cubic packing, the capillary rise in a soil could be approximated as a function of grain diameter. Fig.1b shows a typical arrangement of cubical packing with spherical grains of equal diameter. It can be shown, using geometry, that the radius of the pore space between the grains is 0.2 times the grain diameter ( $d$ ). Assuming that, this pore space could be idealized to represent a capillary tube,  $r = 0.2 d$ . The capillary rise will be:

$$h_c(\text{cm}) = \frac{0.15}{0.20d} = \frac{3}{4d} \quad (3)$$

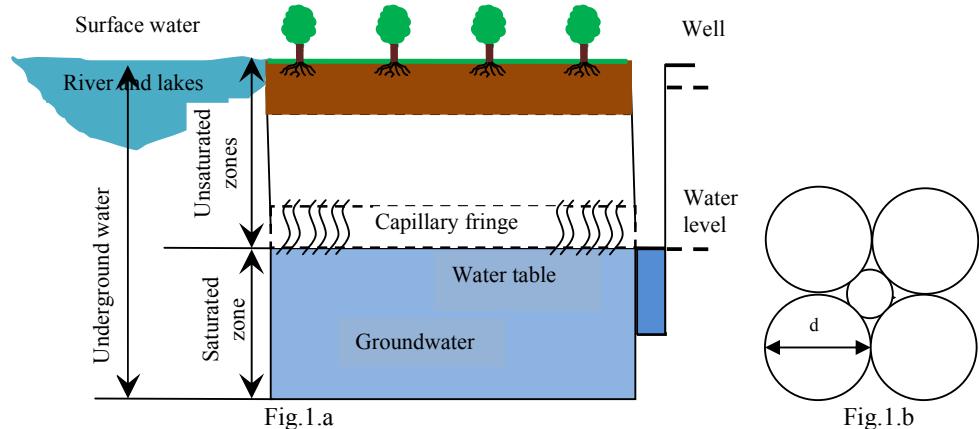


Fig.1a. Capillary fringe position in a soil profile. Fig.1b. Idealized pore diameter in a sediment with cubic packing. The equivalent capillary tube has a radius of 0.2 times the grains diameter ( $d$ )

By conclusion the thickness of capillary zone varies with the pore size of a soil and with the grain diameter. The visual capillary rise is invariably less than the theoretical zone as defined in equation (3). In real porous materials the grains are not identical and rarely are perfect spheres. In such conditions the observed experimental CF is usually different from the theoretical one.

**Table 1**  
**Capillary rise in samples of unconsolidated materials [2]**

Sediments	Grain diameter (d) mm	Estimated capillary rise cm	Actual capillary rise cm
Fine silt	0.05-0.02	150-375	200
Coarse silt	0.1-0.05	75-150	105.5
Fine sand	0.2-0.1	37.5-75	42.8
Medium sand	0.5-0.2	15-37.5	24.6
Coarse sand	1.0-0.5	7.5-15	13.5
Very coarse sand	2.0-1.0	3.75-7.5	6.5
Fine gravel	5.0-2.0	1.5-3.75	2.5

The CF has significant effects on the vertical motion of water [3], and is active biologically, geochemically [4], influences displacement of LNAPLs [5], root water uptake and plant growth.

## 2. Experimental researches concerning nitrates transfer within the capillary fringe

### 2.1 Experiment development

We consider the water flow and the dispersion of nitrogen compounds into a homogenous, isotropic, unconfined aquifer bounded by two lakes whose levels and qualities determine the flow in the aquifer and the pollutant's sources (Fig.2).

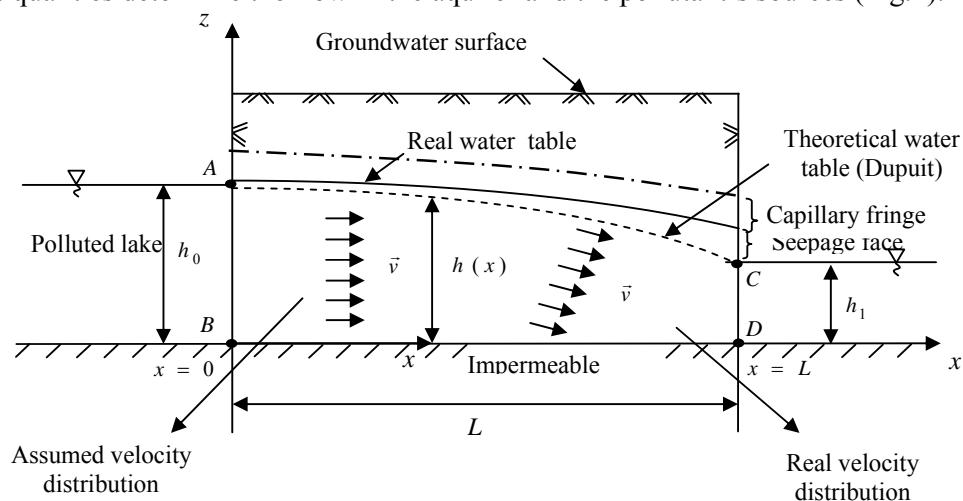


Fig.2. Steady flow in an unconfined aquifer

The experiments have been developed to described pollution and decontamination of an unconfined groundwater and the nitrates transfer within the capillary fringe.

The experimental bench is built to simulate, on a small scale, the hydrological principles of the groundwater flow and the pollutants behavior in saturated soils. The sand tank (fig. 3) is manufactured in glass and located is a steel frame which is designed for standing on a laboratory bench. A diffused water inlet (I) / outlet (O) with associated flow control level is installed at each end of the sand tank.



Fig. 3: Experimental bench

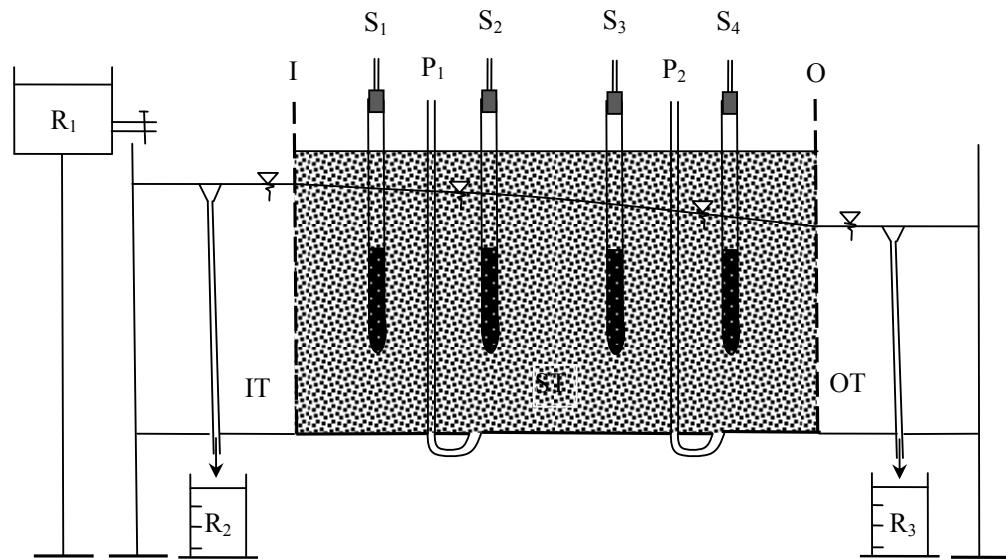


Fig. 4. The scheme of experimental bench. S1, S2, S3, S4 – soil solution extracting tube; P1, P2 – piezometer; IT – inlet water tank; OT – outlet water tank; ST – sand tank

By changing the position of water level in the inlet (IT) and outlet (OT) tanks, the desired water table can be established. Two toppings in the tank base are connected to piezometers ( $P_1, P_2$ ). The water levels in the piezometers indicate the water table position in the longitudinal groundwater profile. Four soil solution extracting tube ( $S_1, S_2, S_3, S_4$ ) are installed in the sand tank (ST). The discharge trough the groundwater can be volumetrically measured using calibrated reservoir  $R_3$  and can be modified controlling the levels position in the inlet and outlet tanks.

Groundwater can be polluted with contaminated water (from  $R_1$ ) having a known concentration. Soil solution is extracted at different time intervals with  $S_1 - S_4$  tubes, using a vacuum pump.

The contaminant used for our experiment was an ammonium nitrate,  $\text{NH}_4\text{NO}_3$ , solution with a concentration of 1000 mg  $\text{NH}_4\text{NO}_3/\text{L}$ . The intake reservoir was continuously filled with that solution during the pollution time (246 minutes). In the pollution experiment 176 samples were taken from six points, (the 2 tanks and 4 extraction wells).

For remediation (decontamination) the tank  $R_1$  was fed with clear water. During 444 minutes, 176 samples were taken in the same points mentioned above.

The samples were chemically analyzed to establish the ammonium,  $\text{NH}_4^+$  and nitrate,  $\text{NO}_3^-$  concentration, using spectrometric analysis (UV-VIS spectrophotometer).

Samples of soil extract were collected in 4 sampling position, before the pollution, after 250 minutes, and 1440 minutes from the beginning of pollution event, at 5 cm above the water-table. Also the soil was sampled after decontamination.

## 2.2 Aquifer characteristics

The unconsolidated aquifer's material can be defined like a mixture of fine gravel, and different types of sand. Its grain size distribution curve is described in Table 2.

Table 2

Grain size distribution curve of aquifer material

No.	Particle grain size [mm]	Soil classification (cfr. STAS1243/88)	Mass retained [g]	Percent finer by mass [%]
1.	3 – 4	Fine gravel	2,23	2,23
2.	2 – 3	Fine gravel	3,43	3,43
3.	1 – 2	Coarse sand	8,96	8,96
4.	0.5 – 1	Medium and fine sand	85,38	85,38
TOTAL			100	100

Aquifer boundaries levels (Fig.2) are:  $h_0 = 0.33 \text{ m}$ ,  $h_1 = 0.297 \text{ m}$ , aquifer length:  $L=0.38 \text{ m}$ .

Hydraulic measurement provided data about the flow in the aquifer:

- hydraulic gradient:  $I = 0.0868$ ,
- discharge through the aquifer:  $Q = 3.5000 \text{ [cm}^3/\text{s}]$ ,
- hydraulic conductivity:  $K = 0.0443 \text{ [cm/s]}$  was measured with a Kamenski permeameter,
- effective porosity of the aquifer:  $n_c = 0.22 = 22\%$ ,
- real interstitial water velocity in the groundwater:  $v = 0.6300 \text{ [m/h]}$ .

Extraction tubes were located at a distance  $x$  from the inlet boundary:  $S_1$  at  $x_1=0.05 \text{ m}$ ,  $S_2$  at  $x_2=0.15 \text{ cm}$ ,  $S_3$  at  $x_3=0.25 \text{ m}$ , and  $S_4$  at  $x_4=0.34 \text{ m}$ .

### **3. Materials and methods for determination of nitrate in water and soil extract**

Nitrate is a compound of nitrogen that occurs naturally in moderate concentrations in many environments. Nitrate ions usually originate from agriculture and are considered pollutants of groundwater bodies if the concentration is bigger than a maximum level. Also, major sources of nitrogen in aquifers are related to wastewater disposal and solid waste disposal (landfills). The lakes, rivers could be a pollution source if they are contaminated.

The Romanian Law 458/2002, regarding the quality of drinkable water, considers for the nitrates Maximum Contaminant Level, the value of 50 mg/l.

#### **3.1 Nitrogen nitrate concentration measurement**

The effect of capillarity on the nitrates transport was analyzed measuring the concentration of nitrate ions in the soil extract from 16 samples taken at 5 cm above the groundwater surface. We used the spectrometric method [6] to measure the nitrate concentration in soil extract solution.

The mineral soluble compounds represented by nitrates and ammonia salts represent the most mobile form for nitrogen in soil, and their quantitative determination is used to characterize the accessible quantity of nitrogen from soil [7].

##### *3.1.1 Sample preparation*

A mixture formed from 10 grams of soil and 50 mL of distilled water was stirred for one hour and then was filtered off. The obtained solution (extract) was used for quantitative determination of nitrate ions.

##### *3.1.2. Standards and calibration*

All chemicals used were of analytical reagent grade. Standard stock solution containing 100 ppm N- $\text{NO}_3^-$  was prepared by dissolving in distilled water 0,7219g of  $\text{KNO}_3$ , final volume being 100 mL.

Standard stock solution that contains 100 ppm  $\text{N-NH}_4^+$  was prepared using 0,4715 g of  $(\text{NH}_4)_2\text{SO}_4$  that were dissolved in distilled water and the final volume was 100 mL. Working standards were prepared by dilution of the stock solution. The calibration curve for nitrate nitrogen was linear for analyzed concentration ranges.

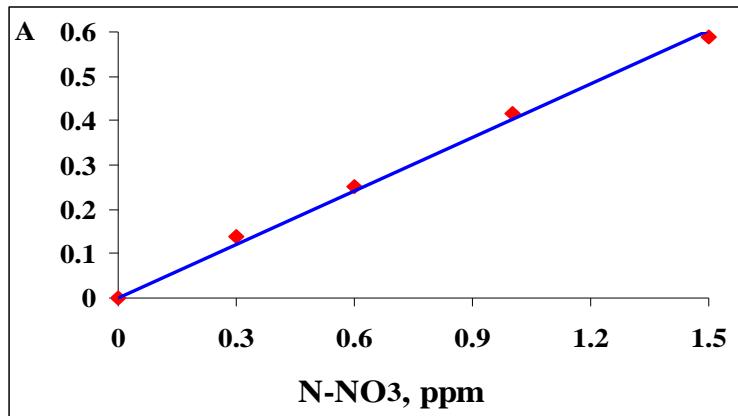


Fig. 5: Calibration curve

The nitrate nitrogen dosage from soil, water and plant were made through spectrophotometric method using phenol-2,4-disulphonic acid in basic medium. It was obtained yellow colored compounds named nitro derivates. These have absorption maxima at 410 nm and the intensity of color of nitro derivates is proportionally with nitrates concentration from analyzed samples.

All the measurements were made at 410 nm using VIS spectrophotometer Metertek SP830 Plus.

#### 4. Results and conclusions

We analyzed the water flow and the dispersion of nitrogen compounds into a homogenous, isotropic, unconfined aquifer above described, bounded by two lakes whose levels and qualities determined the flow and the pollution in the aquifer. We measured the concentration values  $C(x,t)$ , at different distances ( $x=5$  cm,  $x=15$  cm,  $x=25$  cm,  $x=35$  cm) in soil solution extracting tubes during the contamination and decontamination processes. The experimental results during the contamination case are depicted in Fig.6. After 250 minutes the nitrate's concentration in aquifer is 764.481 mg/L in S<sub>1</sub> ( $x=5$  cm) and 705.631 mg/L in S<sub>4</sub> ( $x=35$  cm).

Measuring the nitrate concentration in the soil extract, sampled at 5 cm above the water-table we can appreciate the transfer of nitrates from the saturated zone toward the soil surface, within the capillary fringe.

From Table 1 we can appreciate for our aquifer composition (Table 2) the capillary rise will be between 5 cm and 15 cm.

After 250 minutes since the experiment beginning we observed the porous material near the soil surface, was really moistened by capillarity. We appreciated the capillarity fringe raised toward the soil surface.

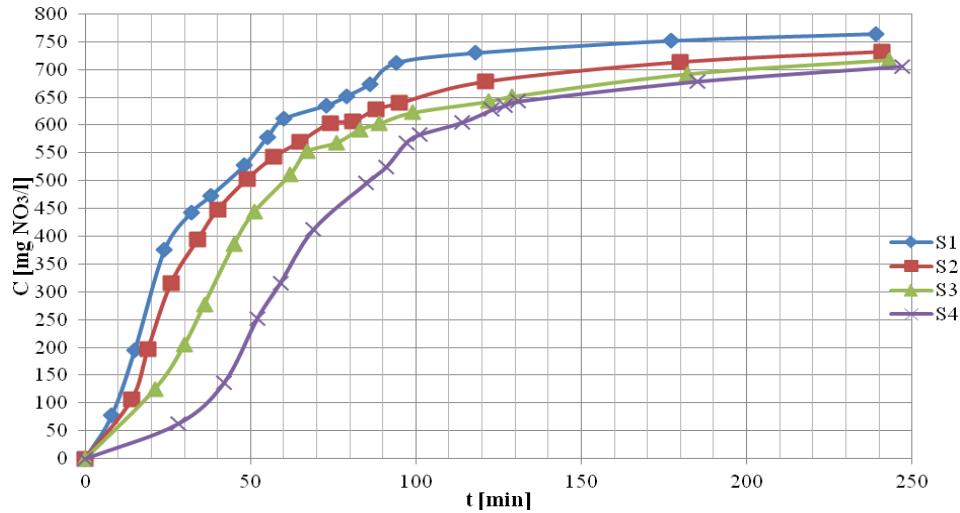


Fig.6. Measured values of concentration  $C(x,t)$ , at at different distances ( $x=5$  cm,  $x=15$  cm,  $x=25$  cm,  $x=35$  cm) in soil solution extracting tubes. Contamination case [8]

*Table 3*  
**NO<sub>3</sub><sup>-</sup> concentration of soil extract from CF (transported by capillarity between aquifer and the unsaturated soil), at 5 cm above water-table, in mg/L**

Sampling position	S <sub>1</sub> $x_1=5$ [cm]	S <sub>2</sub> $x_2=15$ [cm]	S <sub>3</sub> $x_3=25$ [cm]	S <sub>4</sub> $x_4=35$ [cm]
Sampling moment				
Before pollution ( $t=0$ )	0	0	0	0
250 minutes after pollution beginning ( $t=250$ min)	65.815	62.338	60.741	58.426
24h after pollution beginning ( $t=1440$ min)	106.517	86.671	78.879	74.779
After decontamination ( $t=1900$ min)	0	0	0	0

The concentration values of soil extract (Table 3) shows a vertical upward flux of nitrate NO<sub>3</sub><sup>-</sup> transported by capillarity between aquifer and the soil surface. After 250 minutes since pollution beginning the soil extract concentration was 65.815 mg/L at  $x=5$  cm, and 58.426 mg/L at  $x=35$  cm. After 24h the concentration in the capillary fringe was 106.517 mg/L and 74.779 mg/L in the mentioned sampling points.

Finally, in Table 4 the relationship between soil extract concentration of nitrates and the values of concentration in the aquifer, at the same distance from the inlet boundary is calculated in percent.

The experimental results showed the increasing concentration in CF, during the contamination process, for each distance  $x$ , the maximum value being 106.517 mg/L, after 24 hours since the beginning of pollution. That value represents 13.933 % from aquifer concentration.

**Table 4**  
**Percent (%) of NO<sub>3</sub>-concentration of soil extract from CF by the solution concentration in sampling tube**

Sampling moment	Sampling position $x_1=5$ [cm]	S <sub>1</sub>	S <sub>2</sub>	S <sub>3</sub>	S <sub>4</sub>
		$x_2=15$ [cm]	$x_3=25$ [cm]	$x_4=35$ [cm]	
250 minutes after pollution beginning ( $t=250$ min)	Soil extract concentration	65.815	62.338	60.741	58.426
	Concentration in sampling tube	764.481	732.16	717.641	705.631
	%	8.609	8.514	8.472	8.279
24h after pollution beginning ( $t=1440$ min)	Soil extract concentration	106.517	86.671	78.879	74.779
	Concentration in sampling tube	764.48	732.16	717.641	705.631
	%	13.933	11.837	11.003	10.597

By conclusion the soluble pollutants moving with water in an unconfined aquifer could be lifted above the water table by capillarity. The transfer process depends on the material's particle size. That aspect will be one of our next experimental subjects.

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