

General Overview of the Chosen Processes along with Their Impact on the Concentration Reduction Values for Contaminants Moving in Groundwater

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Abstract- This paper addresses the general overview of 2D mathematical models both for inorganic and organic contaminants moving in an aquifer, taking into consideration the most important processes that occur in a ground. These processes impact in different extent on the concentration reduction values for the moving contaminants in a groundwater. In this analysis the following processes have been taken into consideration: reversible physical non-linear adsorption, chemical and biological reactions (as biodegradation-biological denitrification) and radioactive decay (for moving radionuclides). Based on this 2D contaminant transport models it has been possible to calculate numerically the dimensionless concentration values with and without all the chosen processes in relation both to the chosen natural site (piezometers) and the chosen contaminants. In this paper, it has also been possible to compare all the numerically calculated concentration values to the measured concentration ones (in the chosen earlier piezometers) in relation both to the new unpublished measurement series of May 1982 and to the new set of parameters used in these 2D contaminant transport models.

Keywords- Advection; Dispersion; Adsorption; Biodegradation; Radioactive Decay; Numerical Calculations

I. INTRODUCTION

One can say that many more or less complicated mechanisms (processes) appear during the flow of contaminants in a groundwater. These processes appear and overlap in a ground during contaminant movement and should also be mathematically described as practical transport models which can be used in practice to simulate contaminant concentrations in a groundwater. In spite of the phenomenon of contaminant transport in groundwater flow system is quite well-known and described in literature as the partial differential equations; however, the main problem is related with the proper selection of the optimum numerical values of all the parameters being considered in these equations. These all parameters representing the interactive processes in ground system are strongly connected among other things both with a kind of climate (temperature) and with a kind of ground system with moving in it contaminants. It is especially related to the parameters representing the biological and chemical (radioactive decay) reactions – processes (on aqueous – solid surface) being considered in transport equations as the last term of these equations (see also the last term $\sum R_n$ in the presented below general Equation 1).

In various climatic and ground conditions all the numerical parameters using both in author's models and in the many literature models are quite different even for the similar (but never the same) contaminants moving in ground media (Spitz and Moreno, 1996; Chiang, 2005; Javadi and AL-

Najjar, 2007; Weiss and Cozzarelli, 2008; Kraft et al., 2008; Aniszewski, 2009; Taniguchi and Holman, 2010; Aniszewski, 2011). This is connected, among other things, with negative or positive charging of the solid phase of natural soils (as the "anionic" or "cationic" exchange) (Spitz and Moreno, 1996; Chiang, 2005; Javadi and AL-Najjar, 2007; Weiss and Cozzarelli, 2008; Kraft et al., 2008; Taniguchi and Holman, 2010). It is also to be note that in various climatic and ground conditions, the physico - chemical and biological properties both of moving groundwater and contaminants may differ considerably (Spitz and Moreno, 1996; Chiang, 2005; Javadi and AL-Najjar, 2007; Weiss and Cozzarelli, 2008; Kraft et al., 2008; Taniguchi and Holman, 2010). The more detailed description of impact of these climatic and ground conditions on contaminant flow in a ground is given, among other thing, in Spitz and Moreno, 1996; Chiang, 2005; Weiss and Cozzarelli, 2008; Kraft et al., 2008; Aniszewski, 2009; Taniguchi and Holman, 2010; Aniszewski, 2011.

The first important topic of this paper was a presentation of differences between the numerically calculated concentration values with and without all the chosen processes proceeding in groundwater based on the presented, in the further part of this paper, the reduced to 2D domain transport equations. In the further analysis differences between the numerically calculated concentration values with all the chosen processes and the measured concentration values was also made in relation to the other later and unpublished measurement series of May 1982 (compared to the earlier published measurement series of November 1981 given in Aniszewski, 2009 and Aniszewski, 2011). The abovementioned differences (marked generally as Δ values in Table II) are also allowed to calculate the dimensionless values of concentration reductions as the ratios of these differences both to the numerically calculated concentration values with all the chosen processes and to the measured concentration values. All the calculated concentration reductions (given in Table II) were treated in the further analysis as the standard errors ($\Delta / C_{\max c}^*$ and $\Delta / C_{\max m}^*$, respectively) (just like in Aniszewski, 2009 and Aniszewski, 2011). One can say that the presented numerical standard errors are also allowed to estimate an importance of the chosen processes in relation to concentration reduction values (see results given in Section III-C-Table II and general summary and conclusions in Section IV).

The calculated numerically $C_{\max c}^*$ concentrations ($C_{\max c}^* = C_{\max c} / C_o$) are treated in the further analysis as maximal dimensionless ones along the main x (ξ) axis for the

$y(\eta)$ and z axes = 0 (see given below Fig. 1 in Section II-A). The explanations of the dimensional $C_{\max c}$ and C_o concentrations are given in the further part of Section II-A.

II. MATERIALS AND METHODS

A. General Description of the Contaminant Transport and Fate Mechanisms

The partial differential governing equation in 3D domain describing the contaminant transport and fate mechanisms of species k_o is taken into consideration (as mass balance equation with the terms of advection, dispersion, adsorption, physico-chemical and biological reactions) (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005):

$$\frac{\partial(mC^{k_o})}{\partial t} = \frac{\partial}{\partial x_i} (mD_{ij} \frac{\partial C^{k_o}}{\partial x_j}) - \frac{\partial}{\partial x_i} (mu_i C^{k_o}) + q_s C_s^{k_o} + \sum R_n \quad (1)$$

where C^{k_o} is the solute concentration in flowing groundwater in aqueous phase (in the local equilibrium conditions) of species k_o , u_i is the component of the so called pore groundwater velocity in pore space, m is the effective porosity of the porous medium, x_i is the distance along the Cartesian co-ordinate axis, D_{ij} is the hydrodynamic dispersion coefficient symmetrical tensor, q_s is the volumetric flow rate per unit volume of aquifer representing fluid sources (positive) and sinks (negative), $C_s^{k_o}$ is the concentration of the source or sink flux for species k_o , t is the co-ordinate of time and $\sum R_n$ is the function describing generally biological and

chemical reactions treated as aqueous-solid surface ones (see that $\sum R_n$ function in Sections II-D and II-E). All the further adopted assumptions along with the adopted new set of parameters being considered in Equation (1) are presented below and in the further part of this paper (see Section II-E).

In this analysis one-dimensional flow of groundwater along the $x(\xi)$ axis (with respect to natural groundwater slope along this axis, see also Fig. 1) was assumed hence, both the components of the pore groundwater velocities ($u_y = u_z = 0$) and the advection terms ($u_y \partial C / \partial y = u_z \partial C / \partial z = 0$) can be neglected. Taking also into consideration both real slotted influx that is often used in practice of contaminants into the chosen aquifers along the vertical z axis and the concentration equalization along this direction ($\partial C / \partial z = 0$), the dispersion term along this vertical z axis can be set equal to zero ($D_z \partial^2 C / \partial z^2 = 0$). In this analysis, assuming an isotropic ground medium being considered in the chosen real aquifer (in the y and z axes), the simplified form of the hydrodynamic dispersion coefficient symmetrical tensor D_{ij} was accepted. Such assumption is often used in literature for practical engineering applications (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005). So, taking into consideration both the abovementioned assumption and one-dimensional flow of groundwater along the $x(\xi)$ axis, the D_{ij} tensor has simplified form, as the longitudinal and transverse dispersion coefficients D_x and D_y (under assumption $D_y \equiv D_z$).

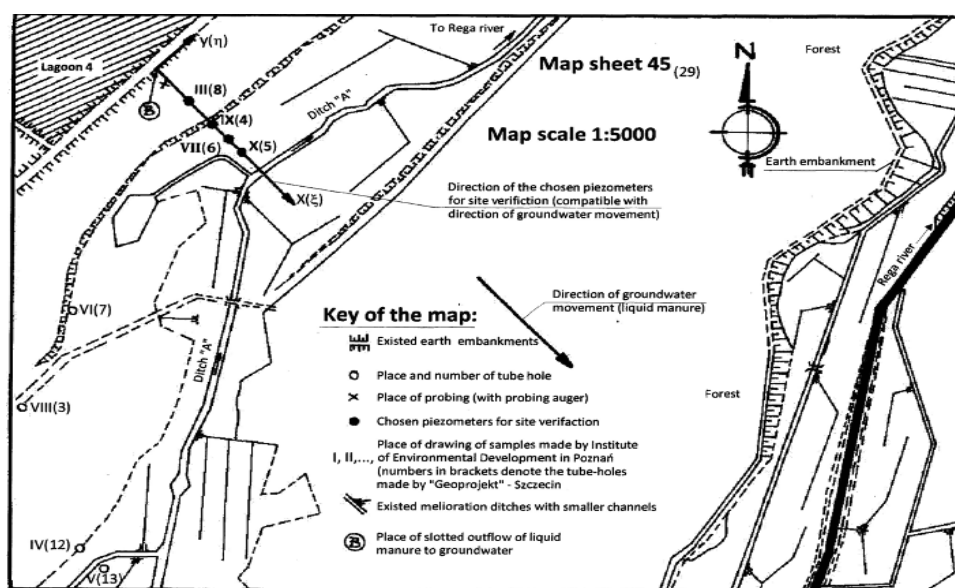


Fig. 1 The illustrative map of the chosen site (piezometers) adopted for numerical calculations and practical verification of the presented here 2D transport models (equations)

The presented above assumptions were also related to the site research made by the Institute of Environmental Development in Poznań as the capital of the Greater Poland Province. The source of groundwater contamination in the chosen site (aquifer) was the real ground lagoon (marked by its user as the Lagoon 4) filled with the liquid manure from the pig breeding farm "Redło" nearby Świdwin in the West Pomeranian Province of Poland. For the exact examination of contamination transport into groundwater a certain number of

piezometers were installed nearby the existing ground Lagoon 4 (Aniszewski, 2009; Aniszewski, 2011). The illustrative map only of the chosen natural site is presented in Fig. 1 along with the detailed numeration of the existed and chosen ones in these analysis four piezometers. These four piezometers were used in the analyzed numerical calculations of the presented here transport models (equations) along with their practical verification (see, in Table II, the exemplary dimensionless standard error values $\Delta / C_{\max m}^*$ in relation to measured

concentrations for chlorides and sulfates as Footnote 4 in parentheses). However, the distances from the ground Lagoon 4, as the liquid manure leakage source, to the particular piezometers are presented in Tables I, II as well as in Fig. 2 (see Sections III-B, III-C and II-E, respectively).

The visual observations of this Lagoon 4 conducted by its user confirmed the vertical slotted cracking of geotextile and direct leaking of the liquid manure into groundwater and afterwards movement of this liquid manure in the direction of Rega river along the main axis x (ξ) (see Fig. 1). Basing on installed piezometers, the Institute of Environmental Development in Poznań measured concentration values of the chosen contamination indicators in the natural groundwater stream (treated as the C_m concentrations), collecting the water samples in these piezometers located nearby the existing ground Lagoon 4. In the further analysis the numerical calculations were related only to the four above-mentioned piezometers located along the main axis x (ξ) for axis y (η) = 0 (see Fig. 1), taking also into consideration the maximal measured concentration values in these four piezometers (treated here as the $C_{\max m}$ concentrations).

The calculated numerically dimensional and maximal ($\eta = 0$) concentrations in the chosen four piezometers are treated in the further analysis as the $C_{\max c}$ ones. During these testing, the concentration values of the chosen contamination indicators were also measured in the samples collected directly in the considered Lagoon 4 and treated as the initial C_o concentrations. In relation to the real (observed) conditions of the source of the liquid manure influx into the natural aquifer, the infinitesimal width of the geotextile slotted vertical cracking was assumed in the further analysis (for the numerical solution and calculations of the presented transport equations).

In the further analysis, taking into consideration the relatively short ground medium $L \approx 105 \text{ m}$ (as the total length between the liquid manure leakage source in lagoon 4 and the last chosen piezometer X (5) – see Fig. 1, Tables I, II), the lack fluid sources (positive) and sinks (negative) for species k_o in Equation (1) was also assumed ($q_s C_s^{k_o} = 0$). In the governing Equation (1) the species index k_o , for simplicity of presentation the presented in the further parts equations, was also dropped. So, finally, taking into consideration all the above mentioned assumptions, the simplified forms of Equation (1) in 2D domains were presented in the further parts of this paper in relation to the x (ξ) and y (η) axes (see Equations 3, 8, 14 and 18 in Sections II-B, II-C, II-D and II-E, respectively).

B. Detailed Description of Contaminant Transport in Groundwater Theoretically without Adsorption Process

Dropping, for chloride and sulfate indicators, both the adsorption process and the biological and chemical reactions in a ground ($\sum R_n = 0$), the most simplified well-known 2D form of Equation (1) can be expressed as follows (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005):

$$\frac{\partial C}{\partial t} + u_x \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2}, \quad (2)$$

where u_x is the component of the so called pore groundwater velocity in pore space along the x axis, D_x , D_y are the components of the longitudinal and transverse dispersion coefficients along the x and y axes that depend on the longitudinal and transverse dispersivities (α_L , α_T) and (x , y) are the Cartesian co-ordinates of the assumed reference system. Dropping, in the first stage of numerical calculations, the adsorption process (proceeding with more or lower intensity in natural ground systems) was aimed at the expected defining concentration reduction values caused theoretically by this process only. In the further analysis the adsorption process, proceeding always in reality in natural ground water streams, was taken also into consideration (see also Equations 8, 14 and 18 with adsorption terms). So, under such assumption, the well-known retardation factor resulting with the adsorption process was also neglected in Equation (2)

($R = 1 + \frac{\rho}{m} \cdot \frac{\partial S}{\partial C} = 1.00$), where the explanation of the parameters appearing in this retardation factor are given in the further part of this paper (after Equation 7). The dimensionless form of Equation (2) is written below:

$$\frac{\partial C^*}{\partial \tau} + \frac{\partial C^*}{\partial \xi} = D_x^* \frac{\partial^2 C^*}{\partial \xi^2} + D_y^* \frac{\partial^2 C^*}{\partial \eta^2}, \quad (3)$$

with the auxiliary parameters:

$$\frac{x}{L} = \xi, \quad \frac{y}{L} = \eta, \quad \frac{C}{C_o} = C^*, \quad \frac{u_x t}{L} = \tau, \quad \frac{D_x}{u_x L} = D_x^*, \quad \frac{D_y}{u_x L} = D_y^*. \quad (4)$$

where L is the measured distance from the source of the contaminant outflow (injection) to the last ground-cross section (piezometer) in the chosen ground, treated as the known length of the solution area in the numerical solution (calculations) ($L \approx 105 \text{ m}$, see also Tables I, II) and C_o is the initial measured contaminant concentrations in the source of the outflow (injection) into the chosen natural aquifer, along the vertical slot with assumed infinitesimal width of this slot, treated as the known initial concentrations in the numerical solution (calculations). The exemplary initial numerical C_o concentration values for the chloride and sulfate indicators are given in Section II-C for the analyzed later measurement series of May 1982. So, taking into account the above assumptions for numerical solution of Equation (3), the initial and boundary dimensionless conditions were adopted in the form:

– initial condition:

$$\begin{aligned} C^*(0 < \xi \leq \infty, 0 < \eta \leq \infty, \tau = 0) &= 0, \\ C^*(\xi = 0, \eta = 0, \tau = 0) &= 1. \end{aligned} \quad (5)$$

– boundary conditions:

$$\begin{aligned} C^*(\xi = 0, 0 < \eta \leq \infty, \tau > 0) &= 0, \\ C^*(\xi = 0, \eta = 0, \tau > 0) &= 1, \\ C^*(\xi \rightarrow \infty, 0 < \eta \leq \infty, \tau > 0) &= 0. \end{aligned} \quad (6)$$

These conditions as Equations (5) and (6) are related to the considered semi-confined plane of groundwater flow. In this analysis, taking into account the symmetry, the area of flow was considered for ($0 < \xi \leq \infty$) and ($0 < \eta \leq \infty$). In

further numerical calculations the asterisk symbol for the concentration values were also neglected ($C^* = C$).

C. Detailed Description of Contaminant Transport in Groundwater with Adsorption Process

Taking into consideration the adsorption process and neglecting the biological and chemical reactions in a ground ($\sum R_n = 0$), the simplified well-known 2D form of Equation (1) can be expressed as follows (Spitz and Moreno, 1996; Seidel-Morgenstern, 2004; Chiang, 2005):

$$\frac{\partial C}{\partial t} \left(1 + \frac{\rho}{m} \frac{\partial S}{\partial C} \right) + u_x \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2}, \quad (7)$$

where S is the mass of the solute species adsorbed on the grounds per unit bulk dry mass of the porous medium (in the local equilibrium conditions), ρ is the bulk density of the porous medium and $[1 + (\rho/m) \cdot (\partial S / \partial C)]$ is the constant in time retardation factor ($R \neq 1.00$) resulting from sorption process (dimensionless) (Chiang, 2005). The dimensionless form of Equation (7) is written below:

$$\frac{\partial C^*}{\partial \tau} + A_1^* \frac{\partial C^*}{\partial \xi} = A_1^* \cdot D_x^* \frac{\partial^2 C^*}{\partial \xi^2} + A_1^* \cdot D_y^* \frac{\partial^2 C^*}{\partial \eta^2}, \quad (8)$$

with the auxiliary parameters:

$$\begin{aligned} \frac{x}{L} = \xi, \quad \frac{y}{L} = \eta, \quad \frac{C}{C_o} = C^*, \quad \frac{u_x t}{L} = \tau, \quad \frac{D_x}{u_x L} = D_x^*, \\ \frac{D_y}{u_x L} = D_y^*, \quad \frac{m}{m + \rho N K C_o^{(N-1)} C^{*(N-1)}} = A_1^*. \end{aligned} \quad (9)$$

where K , N are the parameters of the non-linear Freundlich isotherm (K as the Freundlich constant and N as the Freundlich exponent, that depend on the solute species, the nature of the porous medium and the other condition of the system). The numerical values of the K and N parameters were calculated based on the updated author's laboratory research for all the chosen indicators in relation to the analyzed here other later measurement series of May 1982. According to this presented here later measurement series, the exemplary and updated numerical values of the K and N parameters are: $K = 0.4091 \text{ m}^3 / \text{g}^{-1}$ and $N = 0.6103$ for chloride indicator and $K = 1.2648 \text{ m}^3 / \text{g}^{-1}$ and $N = 0.8368$ for sulfate indicator, respectively. These numerical values of the K and N parameters presented in this paper insignificantly differ from the K and N ones related to the earlier presented measurement series of November 1981, given in Aniszewski, 2009 and Aniszewski, 2011 (as smaller values than those given in the earlier presented measurement series of November 1981). So, the general equation describing dimensionless retardation factor (R) for all the accepted indicators takes the form (Seidel-Morgenstern, 2004):

$$R = 1 + \frac{\rho}{m} \cdot \frac{\partial S}{\partial C} = 1 + \frac{\rho}{m} \cdot N \cdot K \cdot C^{(N-1)}, \quad (10)$$

It should also to be note that the retardation factor (R) is a constant value in time for the constant porosity of natural aquifers but depends on the concentration values. Taking into consideration the updated numerical values of the K and N

parameters, the lower calculated retardation factors based on Equation (10) (treated as the average values R_a) for all the chosen indicators are given in Table II (in Footnote 2). According to the presented series here later measurement, the exemplary initial measured contaminant concentration in the source of the outflow into the chosen natural aquifer (as the Lagoon 4 with liquid manure) are: $C_o \approx 298.0 \text{ g} / \text{m}^{-3}$ for chlorides and $C_o \approx 394.0 \text{ g} / \text{m}^{-3}$ for sulfates, respectively. These initial numerical C_o concentration values presented in this paper insignificantly differ from the C_o values related to the earlier measurement series, given in Aniszewski, 2009 and Aniszewski, 2011 (as greater values than those given in the earlier presented measurement series). The dimensionless initial measured contaminant concentrations in relation to the initial measured contaminant ones in the Lagoon 4 with liquid manure, for all the presented here contaminants, are given also in Table I as Footnote a (as the values equal to 1.00). All the measured concentration data (C_o and $C_{\max m}$) related to the analyzed here later measurement series were described in unpublished report (in Polish) entitled: "Results of physicochemical and bacteriological analyses of water samples together with documentation and conclusions resulted from these analyses for agricultural complex "Redło" in Redło nearby Świdwin", prepared by the Institute of Environmental Development in Poznań (this document may be obtained from the author).

It should also be noticed that the measured chloride and sulfate concentration values ($C_{\max m}^*$) between the particular piezometers are burdened with certain measurement errors and maybe for this reason the variation between them does not have a character of an exponential curve (just like for the numerically calculated concentrations both with and without adsorption process for all the presented indicators). So, these measured concentrations for the exemplary two indicators were not combined between the particular piezometers in Fig.

2. The other measured concentration values ($C_{\max m}^*$) for all the rest analyzed here contaminants in relation to the other later measurement series of May 1982 may be obtained from the author.

D. Detailed Description of Contaminant Transport in Groundwater with Biodegradation - Biological Denitrification

Taking into consideration both adsorption process and the proceeding biological and chemical reactions in a ground (as biodegradation - biological denitrification process) ($\sum R_n \neq 0$), the well-known 2D form of Equation (1) can be expressed as follows (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005; Joekar-Niasar et al., 2008):

$$\frac{\partial C}{\partial t} \left(1 + \frac{\rho}{m} \frac{\partial S}{\partial C} \right) + u_x \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} - \sum R_n, \quad (11)$$

where $\sum R_n$ is the function describing in this case biodegradation (biological denitrification) process being considered in the chosen ground. The ($\sum R_n$) function for biodegradation (biological denitrification) process can be expressed as follows (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005; Joekar-Niasar et al., 2008):

$$\sum R_n = -\rho \frac{\partial S}{\partial t} - k_1 m C - \rho k_2 S, \quad (12)$$

where k_1 is the first-order reaction rate for the kinetically-controlled biodegradation process for the dissolved (aqueous) phase and k_2 is the first-order reaction rate for the kinetically-controlled biodegradation process for the sorbed (solid) phase. In this analysis, for biodegradation process, the same first-order reaction rate k was assumed for both the dissolved and the sorbed phases. However, basing on the literature, it should also be noticed, for biodegradation process, that certain reactions can occur only in the dissolved phase with a different rate constant (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005; Javadi and AL-Najjar, 2007; Weiss and Cozzarelli, 2008; Joekar-Niasar et al., 2008). So, taking into consideration the abovementioned remarks and substituting Equation (12) into Equation (11), Equation (11) can be rearranged and written as:

$$\frac{\partial C}{\partial t} \left(1 + \frac{\rho}{m} \frac{\partial S}{\partial C} \right) + u_x \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} - k \left(C - \frac{\rho S}{m} \right), \quad (13)$$

where k is the first-order reaction rate for the kinetically-controlled biodegradation process (biological denitrification) for both the dissolved (aqueous) and the sorbed (solid) phases. Based on the literature analysis, one can say that the nitrate biodegradation rate degree (biological transformation) proceeding in natural groundwater strongly depends on such physico-chemical parameters as: groundwater temperature (T), water reaction (pH) and oxidation-reduction potential called in short as redox potential (Eh). Different, every time, values of these abovementioned parameters (T , pH , Eh) existing in natural aquifers, strongly impact on different numerical values of the biodegradation first-order rate constants (k_1 and k_2) in these aquifers for various moving contaminants.

The nitrate ions (NO_3^-) and BOD indicator (as biochemical oxygen demand, that can also be used for determining indirectly concentration of substances susceptible to biodegradation process) were chosen in this analysis (Javadi and AL-Najjar, 2007; Weiss and Cozzarelli, 2008; Joekar-Niasar et al., 2008). Based on the literature, in the case of nitrate biodegradation, one can rather speak of biological transformation (reaction), treated in a saturated zone mainly as biological denitrification. More detailed explanations concerning the complex biodegradation process proceeding in a natural groundwater along with description of the physical and chemical parameters depending on this process are given among other things in Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005; Javadi and AL-Najjar, 2007; Weiss and Cozzarelli, 2008; Kraft et al., 2008; Joekar-Niasar et al., 2008; Aniszewski, 2011.

The numerical optimum literature values of the chosen indicators in relation to biodegradation process (biological denitrification) and used in numerical calculations are: $k = 0.18 \times 10^{-5} \text{ s}^{-1}$ for nitrates and $k = 0.67 \times 10^{-5} \text{ s}^{-1}$ for BOD, respectively. The abovementioned numerical values, k , were adopted after detailed analysis of these parameters based on the unpublished Polish literature in relation to the chosen Polish (cold) climatic and ground conditions (in the analyzed here the West Pomeranian Province of Poland). The dimensionless form of Equation (13) is written below:

$$\frac{\partial C^*}{\partial \tau} + A_1^* \frac{\partial C^*}{\partial \xi} = A_1^* D_x^* \frac{\partial^2 C^*}{\partial \xi^2} + A_1^* D_y^* \frac{\partial^2 C^*}{\partial \eta^2} - k^* C^* (A_1^* + A_2^*), \quad (14)$$

with the auxiliary parameters:

$$\begin{aligned} \frac{x}{L} = \xi, \quad \frac{y}{L} = \eta, \quad \frac{C}{C_o} = C^*, \quad \frac{u_x t}{L} = \tau, \quad \frac{D_x}{u_x L} = D_x^*, \quad \frac{D_y}{u_x L} = D_y^*, \\ k^* = k \frac{L}{u_x} = k t, \quad \frac{m}{m + \rho N K C_o^{(N-1)} C^{*(N-1)}} = A_1^*, \\ \frac{\rho K C_o^{(N-1)} C^{*(N-1)}}{m + \rho N K C_o^{(N-1)} C^{*(N-1)}} = A_2^*. \end{aligned} \quad (15)$$

E. Detailed Description of Contaminant Transport in Groundwater with Radioactive Decay

Taking into consideration both adsorption process and the proceeding radioactive decay in a ground ($\sum R_n \neq 0$), the well-known 2D form of Equation (1) can be expressed just like Equation (11) (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005). However, in this case, the ($\sum R_n$) function for radioactive decay can be expressed as follows (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005):

$$\sum R_n = -\rho \frac{\partial S}{\partial t} - \lambda_1 m C - \rho \lambda_2 S, \quad (16)$$

where λ_1 is the first-order reaction rate for the radioactive decay for the dissolved (aqueous) phase and λ_2 is the first-order reaction rate for the radioactive decay for the sorbed (solid) phase. Basing on the literature, it should also be noticed for radioactive decay that the reaction generally occurs at the same rate in both the dissolved and the sorbed phases (Spitz and Moreno, 1996; Zheng and Wang, 1999; Chiang, 2005; Konikow et al., 1996; Javadi and AL-Najjar, 2007). So, taking into consideration the abovementioned remark and substituting Equation (16) into Equation (11), Equation (11) can be rearranged and written as:

$$\frac{\partial C}{\partial t} + u_x \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} - \frac{\rho}{m} \frac{\partial S}{\partial t} - \lambda \left(C - \frac{\rho S}{m} \right), \quad (17)$$

where λ is the first-order decay rate usually expressed as a half-life ($t_{1/2}$) for both the dissolved (aqueous) and the sorbed (solid) phases. The numerical optimum literature value of the chosen theoretical radionuclide λ in relation to radioactive decay and used in numerical calculations is: $\lambda \cong 0.01 \text{ s}^{-1}$ (for the relatively short the half-life time $t_{1/2} \cong 70 \text{ s}$) (see also text for given references – see Konikow et al., 1996 and Footnote 4 in Table I). More detailed explanations concerning the radioactive decay in a natural groundwater for various radionuclides is given also in Konikow et al., 1996; Spitz and Moreno, 1996; Zheng and Wang, 1999; Javadi and AL-Najjar, 2007; Aniszewski, 2009. The dimensionless form of Equation (17) is written below:

$$\frac{\partial C^*}{\partial \tau} + A_1^* \frac{\partial C^*}{\partial \xi} = A_1^* \cdot D_x^* \frac{\partial^2 C^*}{\partial \xi^2} + A_1^* \cdot D_y^* \frac{\partial^2 C^*}{\partial \eta^2} - \lambda^* \cdot C^* (A_1^* + A_2^*), \quad (18)$$

with the auxiliary parameters:

$$\begin{aligned} \frac{x}{L} &= \xi, \quad \frac{y}{L} = \eta, \quad \frac{C}{C_o} = C^*, \quad \frac{u_x t}{L} = \tau, \quad \frac{D_x}{u_x L} = D_x^*, \quad \frac{D_y}{u_x L} = D_y^*, \\ \lambda^* &= \lambda \cdot \frac{L}{u_x} = \lambda \cdot t, \quad \frac{m}{m + \rho \cdot N \cdot K \cdot C_o^{(N-1)} \cdot C^{*(N-1)}} = A_1^*, \\ \frac{\rho \cdot K \cdot C_o^{(N-1)} \cdot C^{*(N-1)}}{m + \rho \cdot N \cdot K \cdot C_o^{(N-1)} \cdot C^{*(N-1)}} &= A_2^*. \end{aligned} \quad (19)$$

All the numerical values of the calculated dimensionless and maximal ($\eta = 0$) concentrations ($C_{\max c}^* = C_{\max c}/C_o$) based on Equations (3), (8), (14) and (18) (without and with

all the chosen processes) are given in Table I and referred to appropriate footnotes in the chosen earlier piezometers. However, the chosen only calculated dimensionless and maximal ($\eta = 0$) concentrations ($C_{\max c}^* = C_{\max c}/C_o$) based on Equations (3) and (8) (without and with adsorption processes) are given also in Fig. 2 and referred to the appropriate colours (see Fig. 2). The exemplary dimensionless measured chloride and sulfate concentrations ($C_{\max m}^* = C_{\max m}/C_o$) (marked as Δ symbol in the colour brown for chlorides and as \bullet symbol in the colour black for sulfates, respectively) are given also in Fig. 2 in relation to the other later measurement series of May 1982. The rest numerical values of both the calculated dimensionless concentrations ($C_{\max c}^*$) and the measured dimensionless ones ($C_{\max m}^*$) were not presented in Fig. 2, taking into consideration preservation of its legibility.

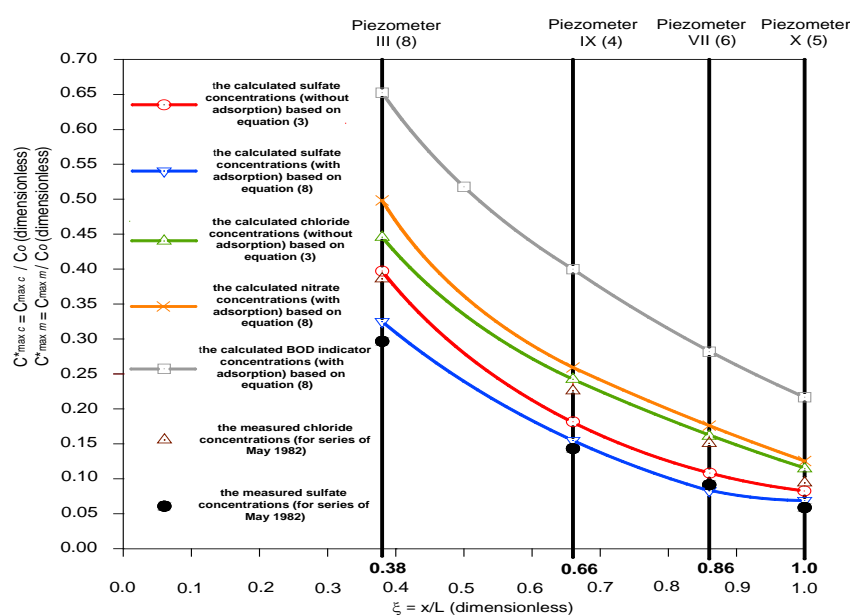


Fig. 2 Maximal values of the dimensionless and numerically calculated concentrations ($C_{\max c}^*$) for the analyzed and chosen indicators in the chosen earlier piezometers along with the exemplary dimensionless measured concentrations for chlorides and sulfates ($C_{\max m}^*$) in the chosen piezometers (in relation to the measurement series of May 1982)

At the same time the numerical optimum values of all the rest required parameters being considered in the presented Equations (3), (8), (14) and (18) as updated and new parameterization of all these equations in relation both to the chosen contaminants and to the considered ground medium are the following:

– the parameters

($u_x = 1.05 \times 10^{-3} \text{ m/s}$, $\rho = 1.71 \text{ g/m}^3$, $m = 0.36$) as the real ground ones adopted based on unpublished technical reports (in Polish),

– the parameters

[$D_x = 7.35 \times 10^{-3} \text{ m}^2/\text{s}$ ($\alpha_L = 7.0 \text{ m}$), $D_y = 5.88 \times 10^{-3} \text{ m}^2/\text{s}$ ($\alpha_T = 0.56 \text{ m}$)]

as the optimum dispersion ones calculated by the author of this paper.

The abovementioned new set of parameters used in the presented here Equations (3), (8), (14) and (18) along with the

new K and N parameters of the non-linear Freundlich isotherm (see Section II-C) are related to the analyzed here other later measurement series of May 1982. These all the used parameters insignificantly differ from the parameter values related to the earlier measurement series of November 1981, given in Aniszewski, 2009 and Aniszewski, 2011 (as smaller values than those calculated in the earlier presented measurement series). General comment concerning of both all the final results of numerical calculations and the general conclusions resulting from these calculations is given in Sections III-C and IV, respectively.

III. RESULTS AND DISCUSSION

A. Description of Difference Scheme Used in All the Presented Numerical Calculations

In numerical calculations the “upwind” scheme (as “explicit” finite difference one, Szymkiewicz, 2010) and the author's modified computational, “PCCS – 2.1” program were used, respectively. This program allows generally to receive

the dimensionless values of the contaminant concentrations (without and with all the chosen processes) in the dimensionless range of $< 0, 1 >$. However, the method of linearization of the presented here Equations (3), (8), (14) and (18) along with approximation of the first and second derivatives in all these equations was presented in detailed in (Aniszewski, 2009 and Aniszewski, 2011). In the presented numerical solution of Equations (3), (8), (14) and (18) the values of dimensionless steps of the difference scheme grid (h^* , k^*) were determined in relation to the real range scale $L \approx 105.0$ m of contamination transport in the chosen earlier natural aquifer (Aniszewski, 2009 and Aniszewski, 2011). The presented below numerical values of dimensionless steps of the adopted in this paper difference scheme grid ($h^* = \Delta x / L \approx 0.01$ and $k^* = \Delta y / L \approx 0.001$) were determined under the assumption that the numerical value of the Peclet number should not be larger than two:

$$Pe = \frac{u_x \cdot \Delta x (\Delta y)}{D_x (D_y)} = \frac{u_x \cdot \Delta x (\Delta y)}{\alpha_L \cdot u_x (\alpha_T \cdot u_x)} = \frac{\Delta x (\Delta y)}{\alpha_L (\alpha_T)} \leq 2,$$

that is, the difference scheme grid should also be no larger than twice the dispersivities (α_L, α_T) as the characteristic length of heterogeneities of the ground systems (Szymkiewicz, 2010). In practice, the Peclet number constraint is often relaxed outside the area of interest, where lower predictive accuracy is acceptable.

Such numerical Peclet number values lower than two ($Pe \leq 2$) minimize simultaneously the “numerical

dispersion” and “artificial oscillations” in relation to Equations (3), (8), (14) and (18), where the contaminant transport in ground is dominated not only by advection process. For such an assumption, the well-known “upwind” scheme (as “explicit” finite difference scheme) is particularly suitable in numerical calculations (Szymkiewicz, 2010). In the further analysis, the numerical value of dimensionless time step of the adopted difference scheme grid ($w^* = u_x \cdot \Delta t / L \approx 0.01$), that minimizes also the “numerical dispersion” and “artificial oscillations”, was determined under the assumption that the numerical value of the Courant number, well-known in the literature, should not be larger than one ($Ca = u_x \cdot \Delta t / \Delta x \leq 1$) (Szymkiewicz, 2010). In these numerical calculations (for the adopted dimensionless values of difference scheme grids), the important consistency, stability and convergence conditions were also preserved to minimize so called “numerical dispersion” and “artificial oscillations” in relation to these all the above mentioned equations describing the particular processes (Szymkiewicz, 2010). More details concerning the applied “upwind” scheme is given also in (Aniszewski, 2009 and Aniszewski, 2011).

B. Presentation of All the Numerically Calculated Concentration Values for the Chosen Contaminants

All the results of the presented numerical calculations of dimensionless concentration values according to appropriate Equations (3), (8), (14) and (18) in the chosen piezometers along with exemplary dimensionless measured concentrations ($C^*_{max m}$) for chlorides and sulfates are given below in Table I.

TABLE I. MAXIMAL DIMENSIONLESS CALCULATED VALUES OF THE CHOSEN CONTAMINANT CONCENTRATIONS ($C^*_{max c}$) ACCORDING TO APPROPRIATE EQUATIONS ALONG WITH THE EXEMPLARY DIMENSIONLESS MEASURED CONCENTRATIONS ($C^*_{max m}$) (FOR CHLORIDES AND SULFATES IN THE CHOSEN EARLIER PIEZOMETERS FOR MEASUREMENT SERIES OF MAY 1982)

Chosen Contamination in Relation to Considered Processes	Numbers of Chosen Piezometers with Dimensionless and Dimensional Distances from the Leakage Source in Lagoon 4 [Total Distance L to the Last Piezometer X (5) – $L \approx 105.0$ M]			
	III (8)	IX (4)	VII (6)	X (5)
	0.38 X \approx 40.0 M	0.66 X \approx 70.0 M	0.86 X \approx 90.0 M	1.00 X (L) \approx 105.0 M
Chlorides (NaCl) [Adsorption Process]	0.4457 ¹⁾	0.2425 ¹⁾	0.1627 ¹⁾	0.1057 ¹⁾
	0.4097 ²⁾	0.2283 ²⁾	0.1553 ²⁾	0.1027 ²⁾
	1.00 ^{a)} ($C^*_{max m} = 0.3862$)	1.00 ^{a)} ($C^*_{max m} = 0.2249$)	1.00 ^{a)} ($C^*_{max m} = 0.1531$)	1.00 ^{a)} ($C^*_{max m} = 0.1014$)
Sulfates (Na ₂ SO ₄) [Adsorption Process]	0.3879 ¹⁾	0.1818 ¹⁾	0.1122 ¹⁾	0.0753 ¹⁾
	0.3250 ²⁾	0.1550 ²⁾	0.0975 ²⁾	0.0660 ²⁾
	1.00 ^{a)} ($C^*_{max m} = 0.2968$)	1.00 ^{a)} ($C^*_{max m} = 0.1435$)	1.00 ^{a)} ($C^*_{max m} = 0.0916$)	1.00 ^{a)} ($C^*_{max m} = 0.0625$)
Nitrates (NO ₃ ⁻) [Biodegradation Process]	0.4995 ²⁾	0.2648 ²⁾	0.1678 ²⁾	0.1147 ²⁾
	0.4934 ³⁾	0.2625 ³⁾	0.1667 ³⁾	0.1146 ³⁾
	1.00 ^{a)}	1.00 ^{a)}	1.00 ^{a)}	1.00 ^{a)}
BOD Indicator [Biodegradation Process]	0.6563 ²⁾	0.4010 ²⁾	0.2819 ²⁾	0.2131 ²⁾
	0.6231 ³⁾	0.3882 ³⁾	0.2750 ³⁾	0.2056 ³⁾
	1.00 ^{a)}	1.00 ^{a)}	1.00 ^{a)}	1.00 ^{a)}
Chosen Radionuclide [Radioactive Decay]	0.3606 ²⁾	0.1760 ²⁾	0.1067 ²⁾	0.0701 ²⁾
	0.3467 ⁴⁾	0.1705 ⁴⁾	0.1037 ⁴⁾	0.0689 ⁴⁾
	1.00 ^{a)}	1.00 ^{a)}	1.00 ^{a)}	1.00 ^{a)}

1) = The values of the maximal dimensionless concentrations $C^*_{max c}$ (MDC) according to Equation (3) without adsorption ($R = 1.00$ for chlorides and sulfates).

- 2) = The MDC according to Equation (8) with adsorption ($R_a \approx 1.03$ for chlorides, $R_a \approx 2.11$ for sulfates, $R_a \approx 1.75$ both for nitrates and for BOD indicator – accepted just like for nitrates and $R_a \approx 1.03$ for chosen radionuclide – accepted just like for chlorides).
- 3) = The MDC according to Equation (14) both with adsorption ($R_a \approx 1.82$ both for nitrates and for BOD indicator – accepted just like for nitrates) and biodegradation ($k^* \approx 0.17$ for nitrates and $k^* \approx 0.62$ for BOD indicator).
- 4) = The (MDC) according to Equation (18) with adsorption ($R_a \approx 1.03$ for chosen radionuclide – accepted just like for chlorides) and with radioactive decay ($\lambda^* \approx \lambda \cdot t$) (as literature value of λ – see also text for references).
- a) = The initial dimensionless concentrations in relation to the initial measured ones in the Lagoon 4 with liquid manure (for the presented later measurement series of May 1982).

C. Presentation of All the Calculated Standard Error Values for the Chosen Contaminants

In the next turn all the dimensionless calculated standard error values ($\Delta / C^*_{\max c}$) in relation to the numerically

calculated concentrations along with the exemplary dimensionless standard error values ($\Delta / C^*_{\max m}$) in relation to measured concentrations (for chlorides and sulfates as Footnote 4 in parentheses) are given below in Table II in the chosen piezo meters.

TABLE II DIMENSIONLESS CALCULATED STANDARD ERROR VALUES ($\Delta / C^*_{\max c}$) IN RELATION TO THE NUMERICALLY CALCULATED CONCENTRATIONS ALONG WITH THE EXEMPLARY DIMENSIONLESS STANDARD ERROR VALUES ($\Delta / C^*_{\max m}$) IN RELATION TO THE MEASURED CONCENTRATIONS (FOR CHLORIDES AND SULFATES AS FOOTNOTE 4 IN PARENTHESES)

Chosen Contamination in Relation to Considered Processes	Numbers of Chosen Piezometers with Dimensionless and Dimensional Distances from the Leakage Source in Lagoon 4 [Total Distance L to the Last Piezometer X (5) – L ≈ 105.0 M]			
	III (8) 0.38 x ≈ 40.0 M	IX (4) 0.66 x ≈ 70.0 M	VII (6) 0.86 x ≈ 90.0 M	X (5) 1.00 x (L) ≈ 105.0 M
Chlorides (NaCl) [Adsorption Process]:				
$\Delta / C^*_{\max c}$ Equation (8) $\times 100\%$	8.8 ¹⁾	6.2 ¹⁾	4.8 ¹⁾	2.9 ¹⁾
$\Delta = C^*_{\max c} [\text{Equations (3) – (8)}] $				
$\Delta / C^*_{\max m}$ for Footnote 4 in Parentheses	(6.1) ⁴⁾	(1.5) ⁴⁾	(1.4) ⁴⁾	(1.3) ⁴⁾
$\Delta = C^*_{\max c} \text{ Equation (8)} - C^*_{\max m} $				
Sulfates (Na ₂ SO ₄) [Adsorption Process]:				
$\Delta / C^*_{\max c}$ Equation (8) $\times 100\%$	19.3 ¹⁾	17.3 ¹⁾	15.1 ¹⁾	14.1 ¹⁾
$\Delta = C^*_{\max c} [\text{Equations (3) – (8)}] $				
$\Delta / C^*_{\max m}$ for Footnote 4 in Parentheses	(9.5) ⁴⁾	(8.0) ⁴⁾	(6.4) ⁴⁾	(5.6) ⁴⁾
$\Delta = C^*_{\max c} \text{ Equation (8)} - C^*_{\max m} $				
Nitrates (NO ₃ ⁻) [Biodegradation Process]:				
$\Delta / C^*_{\max c}$ Equation (14) $\times 1000^{\circ}/_{\infty}$	10.8 ²⁾	8.7 ²⁾	6.2 ²⁾	0.8 ²⁾
$\Delta = C^*_{\max c} [\text{Equations (8) – (14)}] $				
BOD indicator [Biodegradation Process]:				
$\Delta / C^*_{\max c}$ Equation (14) $\times 1000^{\circ}/_{\infty}$	4.7 ²⁾	3.3 ²⁾	2.5 ²⁾	2.0 ²⁾
$\Delta = C^*_{\max c} [\text{Equations (8) – (14)}] $				
Chosen Radionuclide [Radioactive Decay]:				
$\Delta / C^*_{\max c}$ Equation (18) $\times 100\%$	4.0 ³⁾	3.2 ³⁾	2.9 ³⁾	1.8 ³⁾
$\Delta = C^*_{\max c} [\text{Equations (8) – (18)}] $				

- 1) = The standard errors (SE) between the (MDC) according to Equations (3) and (8) with ($R_a \approx 1.03$) and without ($R = 1.00$) adsorption process for chlorides and with ($R_a \approx 2.11$) and without ($R = 1.00$) adsorption process for sulfates in [%] in relation to the (MDC) according to Equation (8).
- 2) = The (SE) between the (MDC) according to Equations (8) and (14) with ($k^* \approx 0.17$ for nitrates and $k^* \approx 0.62$ for BOD indicator) and without ($k^* = 0$) biodegradation (with adsorption $R_a \approx 1.75$ both for nitrates and for BOD indicator – accepted just like for nitrates) in [$^{\circ}/_{\infty}$] in relation to the (MDC) according to Equation (14).

- 3) = The (SE) between the (MDC) according to Equations (8) and (18) with ($\lambda^* \approx \lambda \cdot t$) (as literature value of λ – see also text for references) and without ($\lambda = 0$) radioactive decay (with adsorption $R_a \approx 1.03$ for chosen radionuclide – accepted just like for chlorides) in [%] in relation to the (MDC) according to Equation (18).
- 4) = The (SE) between the (MDC) according to Equation (8) with ($R_a \approx 1.03$) adsorption process for chlorides and with ($R_a \approx 2.11$) adsorption process for sulfates in [%] in relation to the measured concentrations $C^*_{max m}$ in the chosen piezometers.

All the results as the standard errors of the numerical concentration calculations (in % and ‰), given in Table II are as follows:

– based on Equations (3) and (8) (with and without adsorption process in relation to the calculated contaminant concentrations with adsorption): from 2.9 to 8.8 for chlorides and from 14.1 to 19.3 for sulfates (as the maximal errors, see also Fig. 2) (values referred to Footnote 1 in Table II, in %). At the same time, one can say that all these abovementioned maximal standard errors are also various to the standard errors based both on Equation (8) (with adsorption process) and the measured contaminant concentrations in relation to the measured contaminant ones in the chosen piezometers. The numerical values of the calculated earlier standard errors in relation to the measured contaminant concentrations are as follows: from 1.3 to 6.1 for chlorides and from 5.6 to 9.5 for sulfates (values in the parentheses referred to Footnote 4 in Table II, in %). Additionally, it should also be emphasized that the lower measured concentration values (compared to the calculated ones), for all the presented here contaminants, cause a greater margin of safety for prognosis and simulation of concentration values calculated based on the presented transport models (equations). In opposite case practical use of all the presented transport models could be questionable for calculations of contaminant concentrations in ground systems. The exemplary lower measured concentration values for chlorides and sulfates in relation to the calculated concentration values are presented in Fig. 2 (the symbols of these indicators were given also in Section II-E).

– based on Equations (8) and (14) (with and without biodegradation process and with adsorption in both cases): from 0.8 to 10.8 for nitrates and from 2.0 to 4.7 for the BOD indicator (as the minimal errors) (values referred to Footnote 2 in Table II, in ‰),

– based on Equations (8) and (18) (with and without radioactive decay and with adsorption in both cases): from 1.8 to 4.0 for the chosen radionuclide (values referred to Footnote 3 in Table II, in %) (see also text for references – Konikow et al., 1996).

The presented here both numerical calculations of dimensionless concentration values (given in Table I) and the standard errors (given in Table II) confirmed also importance hierarchy of the most important and chosen here processes in relation to the concentration reduction values for all the moving contaminants in the analyzed ground (from sulfates as the highest reduction to nitrates and BOD indicator as the lowest one). It should be note that the abovementioned remark is also confirmed based on the more or less similar earlier calculations of both the concentration values and the standard error values in relation to the earlier measurement series of November 1981, presented in Aniszewski, 2009 and Aniszewski, 2011.

Additionally, it should also be noted that all the measured concentration values from the presented here measurement series of May 1982 in all the chosen piezometers are greater

than the measured concentration values in the earlier measurement series of November 1981 (given in Aniszewski, 2009 and Aniszewski, 2011). It can show a gradual depletion in time of the adsorbing capacity in the sandy aquifer presented here with fine sand filling in relation to adsorption process, as the process that causes the greatest concentration reduction values. This fact is connected at the same time with decreasing in time the retardation factors (R) that depend on contaminant concentration values in ground systems (see Equation 10 and remarks given in Section II-C). We can also say that these calculated retardation factors (R) will be gradually approaching the values $R \approx 1.00$ (for total depletion of the adsorbing capacity of porous media).

Simultaneously, it means that the S value as the mass of the solute species adsorbed on the grounds per unit bulk dry mass of the porous medium will be gradually approaching the zero value $S \approx 0$ (see Equation 10 in Section II-C). The lower numerical values of (R) (treated as the average values R_a) are given in Table I as Footnote 2 compared to the greater R_a values given in Aniszewski, 2009 and Aniszewski, 2011. Nevertheless, to confirm all the above mentioned remarks, it seems to be necessary to carry out further practical verifications of the presented author's numerical transport models (equations) in relation to next ground regions (with installed piezometers) with both the same or other climatic and the same or other ground conditions, compared to the presented here ones (as other sets of parameters using in these models).

IV. SUMMARY AND CONCLUSIONS

Final conclusions resulting from all these numerical calculations, given in this paper, are as follows:

1. One can say that the non-linear adsorption process is of very great importance in terms of the concentration reduction, especially for the sulfates (see Fig. 2 and Table I) and in less range for the chlorides moving in a natural aquifer (see the standard errors referred to Footnote 1 in Table II). This is connected with significantly various adsorption capacities for both these inorganic compounds chosen in this paper (various numerical parameter values of non-linear adsorption isotherms describing this process for these indicators).

2. One can say that the biodegradation of the chosen compounds (as nitrates and BOD indicator) moving in a natural groundwater is of very small importance in terms of the concentration reductions (see the standard errors referred to Footnote 2 in Table II). This is connected with very low numerical values of the first-order biodegradation rate constants, k , for both compounds chosen in this paper and the other inorganic ones taken from the literature. The biodegradation of the chosen indicators moving in a natural groundwater to fully mineral forms proceeds very quickly at a relatively short ground medium [in this paper this distance was referred from the source of the contaminant outflow to the last existed piezometer X (5) – $L \approx 105.0$ m] (Tables I, II). As it results from the literature (see the text for references and Aniszewski, 2011) one can say also of very small importance

of the biodegradation process in relation also to organic compounds susceptible to this process because of very low numerical values of the first-order biodegradation rate constants, k , for this type of contaminants.

3. One can say that the radioactive decay is of small importance in terms of the concentration reductions (see the standard errors referred to Footnote 3 in Table II). At the same time, it should be noted that the first-order decay rate constants λ (half-life times $t_{1/2}$) of various radionuclides vary in huge ranges what simultaneously, can cause great differences in the concentration reductions, but generally in low limited ranges as low numerical values of the last term describing the radioactive decay (see Equations 17 and 18 in Section II-E).

4. One can also say that the presented importance hierarchy of the most important and chosen here processes in relation to the concentration reduction values for all the moving contaminants was confirmed for the two different sets of parameters used in the analyzed equations in relation also to the two different measurement series of November 1981 (Aniszewski, 2009 and Aniszewski, 2011) and of May 1982, respectively.

5. It can also be said that the presented author's transport models in the chosen groundwater stream are a certain contribute and supplement in relation to numerous literature transport models (equations), using for calculations of contaminant concentrations moving in natural ground systems. Especially as it is very hard to define concentration values based on the numerous more or less similar literature models (equations), taking also into consideration several the most important factors given below:

- various every time climatic and ground conditions (as negative or positive charging of various natural ground media in relation to the solid phases – see also text in Section I),

- various every time scales (lengths) of contaminant propagation plumes with the necessity of determining the geometric scale-dependent similarity for these plumes (with various micro- or macro-scopic ground heterogeneities in them) (Joekar-Niasar et al., 2008),

- various every time selections or calculations of all the required parameters being considered both in the presented here equations and in the literature ones, based on laboratory or field measurements (as various numerical values of all these parameters changing in huge ranges).

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REFERENCES

- [1] Aniszewski, A., 2009. Mathematical modeling and practical verification of groundwater and contaminant transport in a chosen natural aquifer. *Acta Geophys.* 57, 435–453, DOI:10.2478/s11600-008-4.
- [2] Aniszewski, A., 2011. Particular application of a mathematical transport model incorporating sub-surface reactive pollutants. *Acta Geophys.* 59, 110–123, DOI: 10.2478/s11600-010-0040-7.
- [3] Javadi, A. A., AL-Najjar, M. M., 2007. Finite element modeling of contaminant transport in soils including the effect of chemical reactions. *Journal of Hazard. Mater.* 143, 690–701, DOI: 10.1016/j.jhazmat.2007.01.016.
- [4] Joekar-Niasar, V., Ataie-Ashanti, B., Sattari, Z. 2008. Large scale modeling of nitrogen transformation in the unsaturated zone – A case study of Tehran City, Iran. Taylor & Francis Group, London, UK, 169–179, DOI: 10.1201/9780203894569.ch 17.
- [5] Seidel-Morgenstern, A., 2004. Experimental determination of single solute and competitive adsorption isotherms. *Journal Chromatogr. A* 1037, 255–272, DOI: 10.1016/j.chroma.2003.11.108.
- [6] Chiang, W.-H., 2005. 3D-Groundwater modeling with PMWIN. A simulation system for modelling groundwater flow and pollution. Springer Verlag, Heidelberg - New York, 397.
- [7] Konikow, L.F., Goode, D. J., Homberger, G. Z. 1996. A three-dimensional method-of-characteristics solute-transport model (MOC3D). U. S. Geological Survey, Water-Resources Investigations. Report 96-4267, Reston, VA, Virginia, USA, 87.
- [8] Kraft, G.J., Browne, B. A., De Vita, W. M., Mechenich, D. J. 2008. Agricultural pollutant penetration and steady state in thick aquifers. *Ground Water* 46, 41–50, DOI: 10.1111/j.1745-6584.2007.00378.
- [9] Spitz, K. H., Moreno, J. 1996. A practical guide to groundwater and solute transport modelling. New York: John Wiley and Sons, 461.
- [10] Szymkiewicz, R., 2010. Numerical modelling in open channel hydraulics. *Water Sci. Technol. Library*, Vol. 83, 370, Springer Dordrecht, DOI: 10.1007/978-90-481-3674-2.
- [11] Taniguchi, M., Holman, I. M. 2010. Groundwater response to changing climate. CRC Press 2010, DOI: 10.1201/b10530-18, 237.
- [12] Weiss, J. V., Cozzarelli, I. M. 2008. Biodegradation in contaminated aquifers: Incorporating microbial/molecular methods. *Ground Water* 46, 305–322, DOI: 10.1111/j.1745-6584.2007.00409.x.
- [13] Zheng, Ch., Wang, P. P. 1999. MT3DMS: A modular transport three-dimensional multispecies transport model for simulation of advection, dispersion, and chemical reactions of contaminants in groundwater systems; Documentation and user's guide. Dept. of Geol. Sci., University of Alabama, Tuscaloosa, AL 35487, Contract Reports SERDP-99-1, 169.