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Investigation of leaching process heavy metals (Fe, Zn) in the soil under sewage sludge application by using hydrus-1D

Azita Behbahaninia*, Maryam Farahani

Department of Environment, Islamic Azad University, Roudehen branch, Iran

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Abstract

Transport of dissolved material in the soil is a dynamic process which a set of differential equations that are solved with the help of computers. Hydrus-1D software is one of the most advanced software in conjunction with one-dimensional movement of water, salts in the soil. In the present study, changes in Fe and Zn concentrations were measured in soils and leaching water affected by wastewater and sludge. Concentrations of heavy metals were considered in wastewater and sludge as boundary conditions on the flow of incoming water, the concentration of iron and zinc were identified in leaching water as the downstream boundary. Considering the equilibrium model Crank Nicholson and Galerkin finite element by software Haydrus, concentration changes of Iron and Zinc was simulated over irrigation period. For this purpose the software Haydrus numbered 1 to number of soil layers, Depth of 100 cm, the depth of groundwater. Time period of irrigation was 150 days. Distribution curves of Fe and Zn concentrations in the soil profile shows the experimental results with computational results obtained from the model are similar. And it can be applied in order to determine the concentration distribution of metals in the non- Unsaturated zone soil and anticipated arrival time to a specified depth. It can be used to control metal concentrations in the soil and groundwater. Accuracy and validity of the model by experimental results has been confirmed for iron and zinc and it can be used in practical cases safely.

***Corresponding Author:** Azita Behbahaninia ✉ Azitabehbahani@yahoo.com

Introduction

Many or most subsurface pollution problems at the field scale involve such simultaneous processes as water flow, multicomponent solute transport, heat transport and biogeochemical processes and reactions. Process-based models that integrate these various processes can be valuable tools for investigating the mobility of a wide range of inorganic and organic contaminants subject to different hydrologic and geochemical conditions (Jacques *et al,* 2008).Process-based reactive transport models can be valuable tools for studying the subsurface fate and transport of organic or inorganic contaminants, including radionuclides, nutrients and pesticides, that may be subject to a large number of often simultaneous interactive physical, chemical, mineralogical, geological, and biological processes (Mayer *et al*, 2002; Lichtner *et al,* 2004; Appelo, 2005).

Transport in soil systems is often further complicated by a multitude of coupled biogeochemical reactions, the presence of spatially and temporally variable flow velocities, and spatial heterogeneities at different scales (Mallants *et al.,* 2011). A numerical tool that integrates these various processes offers unique possibilities for advanced model building and improving process understanding, ranging from sensitivity analyses of processes and parameters, designing experiments, hypothesis testing by running virtual laboratories, conceptual model validation using experimental data, and evaluation of different management or remediation practices. Based on the Hydrus suite of models for one-dimensional (Šimůnek *et al.,* 2008) and multi-dimensional problems (Šimůnek *et al.,* 2011),

The Hydrus software package also includes modules for simulating carbon dioxide and major ion solute movement. Diffusion in both liquid and gas phases and convection in the liquid phase are considered as CO_2 transport mechanisms. The CO_2 production model is described. The major variables of the chemical system are Ca, Mg, Na, K, SO_4, Cl, NO_3, H_4SiO_4, alkalinity, and CO_2 (Šimůnek *et al.,* 2013). The model accounts for equilibrium chemical reactions between these components such as complexation, cation exchange and precipitation-dissolution. For the precipitation-dissolution of calcite and dissolution of dolomite, either equilibrium or multicomponent kinetic expressions are used which include both forward and back reactions. Other dissolution-precipitation reactions considered include gypsum, hydromagnesite, nesquehonite, and sepiolite. Since the ionic strength of soil solutions can vary considerably with time and space and often reach high values, both modified Debye-Hückel and Pitzer expressions were incorporated into the model as options to calculate single ion activities. The fate and transport of nutrients and contaminants in variably-saturated porous media is influenced by a range of interacting physical, chemical, and biological processes (Jacques *et al.,* 2013). Heavy metals present in sludge pose the risk of human or phytotoxicity from land application. Groundwater of south of Tehran is polluted because of ten years irrigation agricultural land with wastewater. IN pilot study research after irrigating with wastewater, the columns were cut and the soils separated from sectioned pieces and their heavy metal concentrations (Pb, Cd and Ni) were measured. Because of high sorption capacity of these elements by soils, these metals were accumulated in surface layer of the soils. Ni has had the most accumulation or the least vertical movement, and Pb the opposite ones (salmasi, *2006).* The main reason for the research is soil and groundwater pollution by heavy metals in south of Tehran In the present study, changes in Fe and Zn concentrations were measured in soils and leaching water affected by wastewater and sludge. Sewage wastes from different parts of Tehran city transfer to south areas of this city by 2 small rivers. Water deficiency at this area is one reason to use wastewaters to irrigate agricultural lands. Because of sludge-borne metals, and vertical movement to groundwater of these waste waters; and high water table in these areas; there is a question that how can wastewater irrigation cause these area's groundwater pollution with heavy metals in long-terms? In other words, how are these elements, movement in the soil profiles with attention to irrigation with wastewater? Answer to these questions is the aim of this study. For this purpose, Fe and Zn were selected and their movement evaluated in this soils. Transport of metals was predicted by using hydrus 1D. There is a need to study and understand further the situation of the retention of heavy metals in soil, especially those which are of particular concern because of their relations with certain human health problems.

Material and methods

The equations

One-dimensional equilibrium water movement in a partially saturated rigid porous medium is described by a modified form of the Richards equation using the assumptions that the air phase plays an insignificant role in the liquid flow process and that water flow due to thermal gradients can be neglected:

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial x}\left[K(\theta)\left(\frac{\partial h}{\partial x} + \cos\alpha\right)\right] - S$$

Where *h* is the water pressure head, θ is the volumetric water content, *t* is time, *x* is the spatial coordinate (positive upward), *S* is the sink term, α is the angle between the flow direction and the vertical axis and *K* is the unsaturated hydraulic conductivity function. In these models to describe soil hydraulic properties such as water retention curve and unsaturated hydraulic conductivity relationships several have been defined. The most common relationship between the van Genuchten - Moallem (Šimůnek, 2006) as follows:

$$\theta(h) = \theta_r + \frac{\theta_s - \theta_r}{\left[1 + (\alpha h)^n\right]^m} \qquad m=1-1/n \qquad n>1$$

$$K(h) = K_s Se^l\left[1 - (1 - Se^{\frac{1}{m}})^m\right]^2$$

Θ_r: residual moisture, Θ_s: saturation moisture, l, m, n: experimental parameters, Ks is the saturated hydraulic conductivity and S_e: relative saturation

Hydrus-1D model for the simulation of water movement in soil, Richards equation solve using linear finite elements. Since in the case of one-dimensional, linear finite elements plan and finite difference is similar, in this respect, using an indirect plan finite difference is as follows:

$$\frac{\theta_i^{j+1,k+1} - \theta_i^j}{\Delta t} = \frac{1}{\Delta x}\left(K_{i+1/2}^{j+1,k}\frac{h_{i+1}^{j+1,k+1}}{\Delta x_i} - K_{i-1/2}^{j+1,k}\frac{h_i^{j+1,k} - h_{i-1}^{j+1,k+1}}{\Delta x_{i-1}}\right) + \frac{K_{i+1/2}^{j+1,k} - K_{i-1/2}^{j+1,k}}{\Delta x} - S_i^j$$

$$\Delta t = t^{j+1} - t^j$$

$$\Delta x = \frac{x_{i+1} - x_{i-1}}{2} \qquad \Delta x_i = x_{i+1} - x_i \qquad \Delta x_{i-1} = x_i - x_{i-1}$$

$$K_{i+1/2}^{j+1,k} = \frac{K_{i+1}^{j+1,k} + K_i^{j+1,k}}{2} \qquad K_{i-1/2}^{j+1,k} = \frac{K_i^{j+1,k} + K_{i-1}^{j+1,k}}{2}$$

Inputs of Model

From main process window of software, part of simulate, water flow and sloute transport parameters was selected. For geometry information, cm unit was considered as length.

Number of soil materials :1

Number of layers for mass balances: 1

Decline from vertical axes: 1 (Cos 0 ° =1)

Depth of the soil profile: 100 cm

Time information: Day was selected as time unit.

Time discretization: start 0 irrigation time: 150 days

Soil Hydraulic model: Van Genuchten-Mualem

Hysteresis: No hysteresis

Soil catalog : loam

Water flow parameters

Qr Residual soil water content : 0.078

Qs Saturated soil water content : 0.43

Ks Saturated hydraulic conductivity: 0.01733

l: Tourtuosity parameter in the conductivity function 0.5

n: Parameter in the soil water retention function 1.56

Water flow boundary condition

- upper boundary condition : contant flux, heavy metals concentration in sewge sludge

-Lower boundary condition: free drainage, heavy metals concentration in leached water
-Constant boundary flux: 150 lit (water for irrigation) , plot size: 2* 10 : 0.00416 cm/day
-Solute transport: 2 kind of metals , 150 days , absorbtion cofficent: 1.7
-Profile information: 19 different of depth in the simulated zones

Wastewater treatment plant Shush is located in south of Tehran. Wastewater and sludge were sampled at different times in during three months. Concentrations of heavy metals were determined in wastewater and sludge samples by atomic absorption Varian model 200. Three experimental pilots of agricultural land, each size 2 by 10 m on a silt loam soil at the South Tehran wastewater treatment plant Shush were prepared. (Behbahaninia *et al.*,2009). Some chemical physical properties of the soil measured before the sludge and wastewater application and the results are summarized in table 1. A pvc drainage pipe was installed in one meter depth of each pilot to collect the leached water. The first pilot was irrigated with pipe water as control pilot, second pilot with wastewater and third one added sludge of wastewater treatment plant. Five months after the cultivation of crops sampling from the soils was performed, also after and during each irrigation period leached water from each pilot was sampled and heavy metals were measured. Soil samples were taken from the topsoil to 100 cm depth. Overall 50 soil samples were taken from all pilots. Heavy metals concentration in the soil samples were obtained by determining metal concentration in 4 normal HNO_3 extract (70 ^{o} C) by atomic absorption spectrometry (Black, 1965).

Table 1. Some physical and chemical properties of the silt loam soil in the study area.

Pilot number	pH	CEC meq/lit	CaCO3 percent	Organic matter percent	Soil moisture percent	Soil porosity percent
2	7.76	36.62	12.25%	0.36	35.7934	0.52
3	7.94	44.25	11.25%	0.37	40.9814	0.52
Mean	7.85	40.43	11.75%	0.365	38.3874	0.52

Results and discussion

Average concentrations of heavy metals in wastewater and sludge treatment plants Shush in Table 2 have been identified. Mean iron concentration in the wastewater 1.35 and zinc 0.15 mg/ l, in the sludge, mean iron concentration 259.8 and zinc 59.3 mg/kg in the sampling time. Results showed iron accumulation in the soil surface layers is more than down layers. As shown in Fig. 1, respectively, 229.8 mg/kg in the third pilot, 50.6mg/kg in the second pilot, 37.5 in the first pilot. The variation of Zn concentration was shown in Fig. 2, the highest concentration Zn in the third pilot on the surface soil layers 80.5, the second pilot 38.5 in the First Pilot 7.97 mg/kg. In the first pilot pipe water was used for irrigation and metals concentrations in surface and deep soil is low. Generally the highest concentrations were found in the surface soil. The highest concentrations were observed for the iron. Zinc and iron were seen in the down depths with low concentrations. Concentrations of metals decline in the second pilot under irrigation with wastewater treatment plant after 30 cm depth. The third pilot accumulation of metals in the soil levels increased substantially compared to second and first pilot. And metal concentration decreases gradually with depth. And the depth of 35 cm, the concentrations of most metals is stable. To investigate the leaching of iron and zinc in leaching water in the sinks, the concentration of heavy metals was also measured in leaching water and the results specified in table 3. Iron concentrations in the first and second pilot low and almost the same 0.090 and 0.094 mg/l but the third pilot was applied the sludge, iron concentration obtained 0.400 mg/liter. The zinc concentration in leaching water obtained in the first and second pilot 0.050 and 0.070 respectively but the third pilot 0.270 mg/l. The experimental results with computational results in the second pilot for iron in Fig. 3 and for zinc in Fig. 4 showed great consistency. Accuracy and validity of the model by experimental results has been confirmed for iron and zinc and it can be used in practical cases safely. The most important consideration for the material transfer is transferring of material which has a high absorption coefficient is taking place slowly over time. Much of this material was absorbed to soil particles near the soil surface. Therefore, the studies of substances into groundwater, the materials with low absorption coefficient are more important (Moshood, 2009). This research should also considered, transfer of heavy metals into groundwater is so complicated because of organic matter percent of soil and Percent calcium carbonate of soil and formation of salts and various complexes with different absorption coefficient. Therefore the absorption coefficient as an important parameter in determinant, Lack of precision in the choice of the coefficients, can be one of the reasons that the simulation results are not fully consistent with the experimental results. Increase of diffusion coefficient is caused faster transmission of material. On the other hand with increase diffusion coefficient, Concentration distribution in the soil profile depth increases and the upper parts of the profiles decreases (Camobreco *et al.,* 1996) although zinc and iron are not toxic metals but Knowledge of the concentration in the soil profile and the possibility of leaching can be effective in maintaining quality of soil and groundwater. And knowledge of the transition metals can be used in the field of operation of sources. Using computer models considering the hydraulic properties of soil and groundwater hydrology predict the influence of metal can in a short time give experts and researchers valuable results for assessing the potential contamination of soil and water resource (Richards *et al.,* 1998). Long term sewage sludge application scenarios showed that heavy metals can move to the deeper depths of soil profiles and enter the relatively shallow groundwater. Consensus of modeling and experimental results show usage Hydrus software can predict situation of transport heavy metals in soil profile.

Table 2. The average of heavy metals concentration in wastewater and sludge mg/l.

sample	Fe	Zn
wastewater	1.35	0.15
sludge	259.8	59.3

Table 3. The average of heavy metals concentration in leaching samples mg/l.

Pilot number	Fe	Zn
1	0.0900	0.050
2	0.094	0.070
3	0.40	0.270

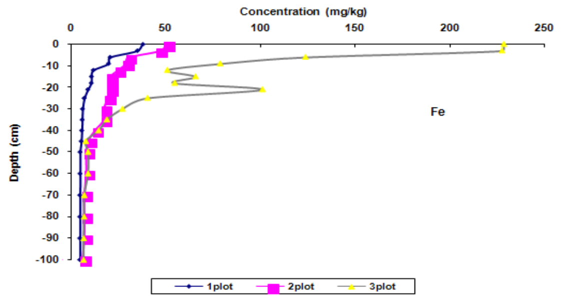

Fig.1. The average concentration of Fe in different depth of pilots.

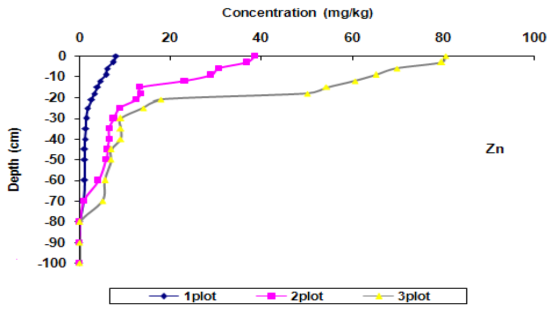

Fig. 2. The average concentration of Zn in different depth of pilots.

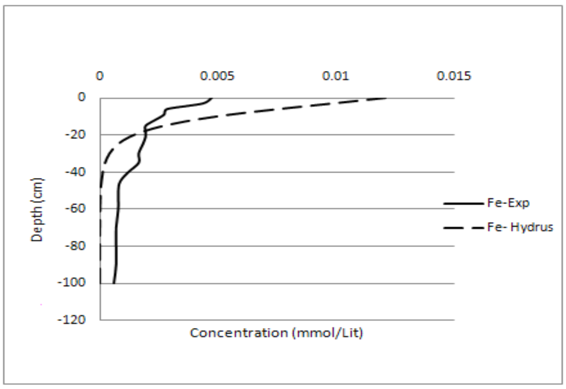

Fig. 3. Experimental and simulation results of Fe concentration in pilot 2.

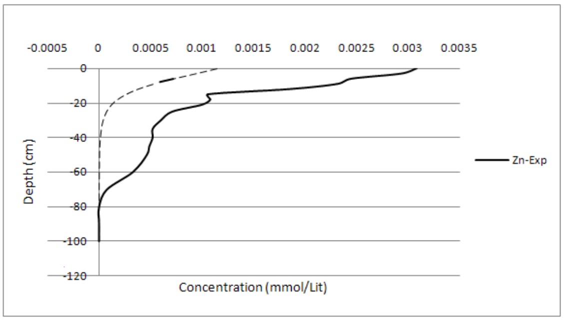

Fig. 4. Experimental and simulation results of Zn concentration in pilot 2.

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