



Variation of the hydraulic properties and solute transport mechanisms in a silty-clay soil amended with natural zeolites



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ABSTRACT

This study aims to quantify changes in flow and transport parameters induced by the addition of zeolites in a silty-clay soil. Hydraulic and physical processes governing solute transport under variably saturated flow conditions were studied in a laboratory scale analog model (AM) irrigated with canal water for seven weeks. One plot of the AM was fertilized with 270 kg-N/ha of urea while the other plot was amended with 5 kg/m² of NH₄⁺-enriched chabazite. The observed water flow was inversely simulated using the single porosity (SP) and the dual porosity (DP) approaches; solute transport was inversely simulated using the convection–dispersion equation (CDE) and the mobile–immobile (MIM) approaches using HYDRUS-1D. Total domain reflectometry (TDR) probes were used to track soil water content and salinity while moisture sensors allowed obtaining the matric potential. The transport of bulk solutes through the soil could be coherently described using a simple approach (SP + CDE). Inverse parameter estimation suggested that percolation and solute front can be confidently predicted in silty-clay soils in case of low precipitation intensity using a combination of TDR and matric potential monitoring techniques. This study shows that NH₄⁺-enriched zeolites increase the water retention capacity even in silty-clay soils, thus limiting water and solute losses.

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1. Introduction

Intensive agriculture and farming impact soil and water quality bearing concerns over its long time sustainability (Foley et al., 2011). Therefore, nowadays research is focused on innovative management strategies for improving soil fertility, reducing water consumption and nutrient loss to surface and groundwater that can ultimately lead to eutrophication (Edmeades, 2003).

Urea has been the most used Nitrogen (N) source worldwide (Glibert et al., 2006; Heffer and Prud'Homme, 2008) due to lower cost per unit of N. However, N use efficiency of urea applied on soil surface may be decreased due to volatilization of ammonia (Rochette et al., 2009; Zhang et al., 2011). Soil conditions increasing Nitrogen (N) use efficiency, such as organic and integrated farming, alleviate these problems (Reganold, 1995). Application of organic and inorganic amendments is a valuable technique for reducing compaction and leaching, while increasing available water content and nutrient-holding capacity of the soils (Waltz et al., 2003). Addition of organic amendments leads to an increase in soil water retention, root zone aeration and nutrient retention (Bigelow et al., 1999; Juncker and Madison, 1967; McCoy, 1992) but organic matter decomposition over time reduces hydraulic conductivity and air-filled porosity (Huang and

Petrovic, 1995; McCoy, 1992). Inorganic amendments, retaining physical properties for extended period, are more appropriate (Aksakal et al., 2012; Truman et al., 2010). Among these zeolites, crystalline hydrated aluminosilicates having a three-dimensional crystal structures (Rehakova et al., 2004), have been recognized to improve the waterholding capacity (Huang and Petrovic, 1994; Xiubin and Zhanbin, 2001), the drainage control (Bigelow et al., 2004) and the retention and release of ammonium (NH₄⁺) due to their high cation exchange capacities (CECs) (Bish and Ming, 2001; Ferguson and Kissel, 1986; McGilloway et al., 2003). Recently, the application of natural zeolites has been reported to diminish nutrient leaching and increase crop water use efficiency (Coltorti et al., 2012; Gholamhoseini et al., 2013; Githinji et al., 2011; Saadata et al., 2012) and to reduce ammonia loss, because their small internal channels can protect ammonium ions (NH₄⁺) from excessive nitrification by microbes (Latifah et al., 2010). Chabazite is one of the most useful natural zeolites due to its high CECs (Mumpton, 1999; Sheta et al., 2003), selective reversible sorption for NH₄⁺ (Gualtieri and Passaglia, 2006) and structure stability over long period (Baerlocher et al., 2001). The soil physical properties, like the water retention capacity and the cation exchange capacity, can be positively altered by the addition of Chabazite but quantitative studies are still lacking except for few recent examples (Coppola et al., 2002; Hong et al., 2011).

Calibration of flow and transport parameters through simulation of the observation data helps to understand the effect of zeolite addition

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to the soil. HYDRUS-1D (Šimůnek et al., 2006) is a physically based model that solves the Richards equation for water flow and a convection–dispersion equation (CDE) for solute transport (Šimůnek et al., 2008). HYDRUS-1D has been recently used to simulate the transport of soil water (Hassan et al., 2010; Kandelous and Šimůnek, 2010), salts (Roberts et al., 2009; Wang et al., 2010) and organic contaminants (Cheviron and Coquet, 2009). HYDRUS-1D also addresses the impacts of preferential water flow and physical non-equilibrium solute transport on soil and water resources. Methods for estimating parameters of the steady-state Mobile-Immobile approach (MIM) involve parameter fitting under optimized experimental conditions (Köhne et al., 2009). For example, the immobile water content, the solute transfer coefficient, and the dispersion coefficient were estimated using a vertical TDR method that was used to analyze resident tracer breakthrough (Lee et al., 2000, 2001, 2002). A flow interruption technique for intermittent solute leaching through soil analog model (AM) was applied to estimate the mobile and immobile water contents, and the mass transfer coefficient (Ilseman et al., 2002). Several other studies have reported similar analyses for the MIM (Ersahin et al., 2002; Shaw et al., 2000).

This paper describes the application of the Italian Chabazite-rich tuff from Sorano (Grosseto) as a soil conditioner and slow nutrient fertilizer to an irrigated silty-clay soil analog model. The objectives of the study were to: (1) evaluate and compare the physical and hydraulic properties of mixtures of silty-clay soil and natural zeolite (95:5% v/v) against the unamended soil; (2) determine the effects of applying NH_4^+ -enriched zeolites and urea on silty-clay soils and (3) model water and solute movement for scenarios with and without amendment incorporation. The rationale of the study lies in the belief that inorganic amendments, that improve the physical and hydraulic properties of soils, can lead to minimize irrigation water use and retard excess leaching of solutes.

2. Materials and methods

2.1. Soil collection and characterization

The soil material was sourced from the top layer (0–20 cm) of the ZeoLIFE experimental site (Coltorti et al., 2012), which consists of recent interfluvial silty-clay deposits (Bondesan et al., 1995; Mastrocicco et al., 2013). Two series of soil samples from two different locations were collected to minimize soil heterogeneity. The soil samples were then mixed and a physical characterization was performed for the resulting mixture in triplicates (see Table 1 for results).

The zeolite-bearing sample comes from a thick deposit of volcanoclastic products close to Sorano (Grosseto, IT) having chabazite ($68.5 \pm 0.9\%$) and phillipsite ($1.8 \pm 0.4\%$) as the main zeolites,

intertwined with a K-feldspar ($9.7 \pm 0.7\%$), mixed with volcanic glass ($11.2 \pm 1.0\%$) (Malferrari et al., 2013). The main utilization of these materials has been as dimension stones in the building industry; currently, they are also employed in animal farming and in agriculture (De'Gennaro and Langella, 1996) due to their easy availability, very low cost and their high-grade zeolites leading to a CECs of 2.17 meq/g (Malferrari et al., 2013). Permeability test using a 2800 K1 Guelph Permeameter was performed at the end of the experiment on the upper and lower soil horizons (at 0.1 and 0.4 m). The Guelph Permeameter measures the saturated hydraulic conductivity of unsaturated deposits with a steady-state constant head using a Mariotte bottle system constituted by plastic tubes (Elrick and Reynolds, 1992). The dry bulk density and the water content were determined gravimetrically. The gravimetric water content was measured for saturated condition, after elution of 10 pore volumes in triplicates columns of 100 ml. The residual water content was measured gravimetrically in triplicates on air dried sediments after heating for 24 h at 105°C . The organic matter content of the soil was measured by dry combustion (Tiessen and Moir, 1993).

2.2. Analog model setup and experimental settings

A tridimensional analog model (AM), 100×50 cm in size was realized and filled with soil up to 45 cm. The AM was divided in two plots of 50 cm each: in the unamended plot (Unamended) the whole soil profile was made of the unamended soil collected from the ZeoLIFE filed site while in the zeolite amended plot (Zeolite) the top 10 cm is a mixture of unamended soil and natural zeolites (Malferrari et al., 2013) having the sedimentological characteristics reported in Table 1 for the amended soil and the rest of the soil profile is made of unamended soil (Fig. 1).

The filling method was to manually compact successive layers of about 2 cm each. The first 20 cm was filled and irrigated with a precision irrigation sprayer Volpi Micronizer M3V (Gardenup Inc., Brescia, IT) for ten days and then let evaporate at room temperature for 17 days ($20 \pm 0.5^\circ\text{C}$). Afterwards, a drying step was carried out installing two lamps of 60 W each, to provide a constant temperature of $50 \pm 1.0^\circ\text{C}$ at the ground level. This procedure allowed estimating the soil retention curve and the percolation front timeframe (Guerzoni et al., 2013). After this preliminary trial the filling procedure went on and sensor probes were inserted at -2 cm, -9 cm, -20 cm, -29 cm and -37 cm below ground level (b.g.l.) in both plots and they were connected to two data loggers (Fig. 1). 270 kg N/ha was applied on the top of the Unamended, using synthetic urea (46% N) granules buried below 2–3 cm to prevent NH_3 volatilization. In Zeolite the first 10 cm of soil was mixed with 5 kg/m^2 of natural zeolite (chabazite) enriched with 10 mg/g NH_4^+ from pig slurry. To monitor the soil water movement, two different kinds of sensor probes were employed; the 200SS Watermark cylindrical soil moisture sensor (IRROMETER Company, Inc. Riverside, CA) measuring the matric potential with a range of 0–200 centibars and the 5TE (Decagon Devices, Inc, Pullman, WA) TDR sensor measuring volumetric water content, temperature and bulk soil electrical conductivity (EC_b). All probes were connected to data loggers recording every 15 min.

The three-dimensional AM was just monitored for the first 10 days of the experiment to check the invariance of the monitored parameters at constant boundary conditions. The AM was then gradually wet with water using a precision irrigation sprayer until its almost complete saturation from day 10 to day 52 of the experiment. Successively, from day 53 to 80 it was let evaporate at room temperature. The raw TDR data were calibrated using the experimental marsh soil, since TDR probes are pre-calibrated for a medium-fine mineral soil that was unsuitable for this kind of soil (Mortl et al., 2011). The calibration procedure was performed with a third order polynomial function; the fit was very good with a R^2 of 0.991. The EC_b was converted to porewater EC using the model of Vogeler et al. (1996). Finally the soil water EC data

Table 1
Sedimentological characteristics of the unamended and amended soils and of the zeolite-bearing sample (mean \pm standard deviation) from triplicate samples.

Parameter	Unamended soil	Zeolite-bearing sample	Amended soil
Grain size (%)			
Coarse sand (630–2000 μm)	0.0 ± 0.0	5.0 ± 2.0	0.6 ± 0.1
Medium Sand (200–630 μm)	0.6 ± 0.1	10.5 ± 3.1	1.6 ± 0.1
Fine Sand (63–200 μm)	7.4 ± 0.3	15.5 ± 1.4	8.3 ± 0.3
Silt (2–63 μm)	49.2 ± 3.1	45.4 ± 4.2	48.2 ± 3.1
Clay (<2 μm)	42.0 ± 3.4	23.6 ± 2.9	41.3 ± 3.4
Organic matter	8.1 ± 1.5	15.9 ± 1.4	8.4 ± 1.5
Hydraulic conductivity (cm/day)	6.7 ± 2.4	48 ± 3.6	7.3 ± 2.4
Bulk density (kg/m^3)	1.15 ± 0.05	1.05 ± 0.07	1.15 ± 0.05
Saturated water content (%)	58.5 ± 0.6	39.3 ± 0.4	58.2 ± 0.6
Residual water content (%)	13.0 ± 0.2	14.8 ± 0.3	13.0 ± 0.2

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