



# Vadose zone oxygen (O<sub>2</sub>) dynamics during drying and wetting cycles: An artificial recharge laboratory experiment



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## SUMMARY

Vadose zone oxygen dynamics control all subsurface redox reactions and play a decisive role in maintaining groundwater quality. Although drying and wetting events are common in artificial recharge, their effects on subsurface oxygen distribution are poorly documented. We monitored oxygen concentration in the unsaturated zone in a mid-scale (1 m high) laboratory soil lysimeter, which was subjected to short wetting and drying cycles that simulated a highly permeable shallow aquifer recharged by river water. Ten cycles of varying duration were performed for a period of 85 days. Measurements of oxygen in the liquid and the gas phases were recorded every 20 s using non-invasive optical fibers (PreSens). The results provided high-resolution (in time) oxygen concentration maps. The infiltration rate revealed a decreasing trend during wetting cycles associated with biological clogging. Such a decrease with time was accompanied by a depletion of O<sub>2</sub> concentration, occurring within the first few hours of the infiltration. During drying, O<sub>2</sub> concentrations recovered rapidly at all depths owing to air flushing, resulting in a stratified vertical profile consistent with the biological consumption of O<sub>2</sub> along the air infiltration path. Furthermore, drying periods caused a potential recovery of the infiltration capacity while preserving the soil biological activity. Scraping also led to the recovery of the infiltration capacity of the soil but was less effective than drying. Our experiment suggests that the small-scale heterogeneity played a key role in accurately mapping pore-scale O<sub>2</sub> concentrations and should be considered in modeling O<sub>2</sub> fluxes of unsaturated soils under natural or managed recharge conditions.

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## 1. Introduction

Subsurface redox zonation is driven by the spatial and temporal distribution of oxygen that serves as the primary terminal electron acceptor during the degradation of organic carbon (Greskowiak et al., 2006). Understanding oxygen zonation in artificial recharge is important for two reasons: (a) biodegradation of hydrocarbons demands aerobic conditions (Rifai et al., 1995; Christensen et al., 2000), and (b) biodegradation of halogenated compounds requires reducing conditions (Bouwer, 1994; McCarthy and Semprini, 1994; Vogel, 1994). Moreover, bio-denitrification takes place preferentially under anaerobic conditions (Schmidt et al., 2011; Rubol et al., 2012). Thus, in order to better understand organic and inorganic contaminants in aquifer systems, it is necessary to carry out a detailed mapping of the subsurface distribution of oxygen and to elucidate the transport processes.

Artificial recharge of groundwater from available surface water is an important management strategy for replenishing groundwater supplies while improving water quality (Dillon, 2005; Fox et al., 2006). The extent to which conditions in these managed systems can be optimized to achieve an adequate water supply (involving both quantity and quality aspects) depends largely upon the physical, geochemical and biological processes that occur in the vadose zone.

Application of surface water or wastewater in rapid infiltration systems is cyclic and typically consists of a period of water application (flooding) followed by days or weeks of drying (Bouwer and Rice, 1984; NRMRI, 2006; US EPA, 1984). Drying is often accompanied by scraping the low-permeability layer on the pond's floor (e.g., Mousavi and Rezai, 1999). This is a way to recover aerobic conditions of the topsoil surface and the infiltration capacity, and to renew the soil's capability of biodegradation (e.g., Bouwer, 2002). The hydraulic loading rate within each wetting cycle affects oxygen availability, pore-fluid velocity and retention time. However, it is not possible to assess the optimal ratios of

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flooding/drying periods owing to the complexity and difficulty in accurately measuring and mapping the spatial and temporal distribution of oxygen concentrations in the field (Akhavan et al., 2013).

Soil pore oxygen concentration depends on the interaction between transport and consumption processes. The former is regulated by advection and diffusion, whereas the latter occurs via chemical reactions and microbial activity. Hot-moments, which are defined by McClain et al. (2003) as short periods that display very high reaction rates with respect to longer intervening periods, affect microbial activity directly (Schimel et al., 2007) or indirectly by changing the redox conditions and O<sub>2</sub> availability (e.g., Rubol et al., 2012). High resolution O<sub>2</sub> maps of soil metabolic activity revealed the presence of these hot-moments in riverbed sediments (Dutta and Rubol, 2014). Microbes can adapt to fluctuations of redox conditions by changing their function or composition (DeAngelis et al., 2010), or protecting themselves by forming biofilms (Romani et al., 2004). Biofilm formation may result in biological clogging, which could reduce the hydraulic conductivity of the soil by several orders of magnitude (Baveye et al., 1998; Bouwer and Rice, 2001) and alter the soil's water retention capacity (Rubol et al., 2014). Infiltration rates may also be affected by temperature, either directly due to the dependence of water viscosity on temperature (Constantz, 1982; Jaynes, 1990), or indirectly as temperature affects biological activity (Le Bihan and Lessard, 2000). Notwithstanding, a poor knowledge of the pore-scale processes and the lack of instrumentation techniques to capture small-scale heterogeneity prevent us from proper upscaling from micro to larger macro scale (Krause et al., 2014).

In the present study, using a 1 m high lysimeter equipped with an array of sensors, we monitored a number of physical and chemical parameters continuously over a period of 85 days in order to link the spatial and temporal dynamics of fluid flow, water retention, and pore-scale O<sub>2</sub> concentration distribution with depth and time during the succession of wetting and drying cycles. The experimental setup also allowed us to assess the changes in the infiltration capacity resulting from the drying phases.

## 2. Materials and methods

### 2.1. Soil collection, preparation and characterization

Soil was collected from the prodelta region of the Llobregat River in a Managed Aquifer Recharge facility located in Sant Vicenç dels Horts, Spain (418446.63 N, 4581658.18 E, zone 31T). Pebbles and coarse grains were removed by passing the soil through a sieve of size 0.2 cm. As a result, the amount of fine grains increased, facilitating permeability and the soil's capacity to support bacterial activity. By using a sieve of size 0.2 cm, the architecture of soil aggregates was preserved.

Representative soil samples were analyzed for particle size distribution, pH and dissolved organic carbon (DOC, organic carbon analyzer model Shimadzu TOC-V-CSH 230V). A small composite sample was extracted using 2 M KCl for the determination of ammonium and nitrate. The extractants were analyzed using an Ionic chromatograph (model-DIONEX IC5000) equipped with an autosampler AS-AP (Injection volume: 25 µL), a gradient pump with an eluant flow rate 1 mL/min, an eluant generator for the production of the mobile phase: 22–40 mM KOH (for anions) and 30 mM MSA (for cations), conductivity detector (suppressed conductivity), variable wavelength detector (not used for this application), an IonPac® AS18 anion-exchange column (4 × 250 mm) with an AG Guard column (4 × 50 mm) and an IonPac® CS16 cation-exchange column (5 × 250 mm) with a CG Guard column (5 × 50 mm).

Analysis of particle size distribution (ASTM, 2000) indicated that the soils were largely composed of medium sand (>80%, see Table 3). X-ray powder diffraction analysis of a sample of soil located nearby revealed that the soil consisted mostly of quartz, with an observable amount of calcite and traces of dolomite, albite, clinchore, muscovite and orthoclase (exact proportions not known). Chemical characteristics of the sediment are listed in Table 3.

### 2.2. Lysimeter set-up, packing and chemical sink addition

A rectangular shaped lysimeter was constructed following the method described in Rubol et al. (2014) (see Fig. 1 for the complete setup). The lysimeter was made of plexiglass, and was 1.2 m high, 0.46 m long, and 0.15 m wide to minimize boundary effects and to allow for some degree of heterogeneity in both the vertical and horizontal directions. All the instrumentation and materials (except the soil) were autoclaved.

The lower 15 cm of the tank was filled with silica sand (0.7–1.8 mm diameter, supplied by Triturados Barcelona, Inc.) and was covered with a geo-synthetic membrane to prevent flushing of the smaller particles out of the system. The upper 85 cm of the tank was filled with the sieved soil to minimize perturbations both in soil conditions and existing cellular biodiversity. Granular materials were placed layer by layer (10 cm thick) and were packed manually to attain an adequate consistency. Additional compaction occurred owing to the hydrostatic forces created by successive filling and emptying of the device but no additional shrinkage was observed. Filling took place from the bottom to avoid bubble retention. Porosity values were determined from the saturated water content, ranging from 0.27 to 0.38 depending on the sensor location. An initial dry bulk density of 1.38 g/cm<sup>3</sup> was measured.

The top 20 cm of the tank was left empty to allow ponding and infiltration conditions to develop during wetting periods. The height of the water above the soil surface was maintained at a fairly constant level by means of a regulated peristaltic pumping system. Using this device, the system was fed with chemically controlled (synthetic) water with no recirculation. The synthetic water was made up of the chemical signature compounds of the river Llobregat (Fernandez-Turiel et al., 2003) (see Table 1). Organic and inorganic compounds (see Table 1 for compositions) were mixed with deionized water daily in order to minimize variations in its chemical composition with time. To monitor the infiltration rate, pumped water and water level at the pond were recorded. Infiltration as a function of time was estimated by applying water balance considerations at the pond. The rate of evaporation during the wetting phases was negligible compared with the infiltration rate. During drying periods, the water feeding system was substituted by an array of five 15 W light bulbs. These bulbs were placed 15 cm above the surface to mimic the effect of the sun directly on the soil surface during the drying phases. Only the surface of the lysimeter was directly exposed to light, while the lateral walls were covered by a black plastic bag to prevent autotrophic activity inside the system. The bottom of the tank was connected to an external water reservoir to fix the water table level.

### 2.3. Experimental protocol and data collection

The infiltration experiment lasted 85 days. By changing the top boundary condition, the lysimeter was subjected to five wetting (W) cycles alternating with five drying (D) cycles of variable duration (see Table 2). The cycles can be classified according to their duration as: medium (W1–D1, W2–D2), long (W3–D3, W5), and short (W4–D4, D5). In the middle of the longest wetting cycle, W3, infiltration was discontinued for a short time, followed by

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