Journal of Hydrology (2007) **332**, 427–441

available at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/jhydrol

Solute transfer in the unsaturated zone-groundwater continuum of a headwater catchment

Cédric Legout [a,d,*], Jérome Molenat [a], Luc Aquilina [b], Chantal Gascuel-Odoux [a], Mikael Faucheux [a], Yannick Fauvel [a], Thierry Bariac [c]

[a] *IFR CAREN, Sol Agronomie Spatialisation, INRA Rennes, France*
[b] *IFR CAREN, Géosciences Rennes, CNRS-Univ. Rennes I, France*
[c] *Biogéochimie et Ecologie des milieux continentaux, CNRS-ENSCP-INA PG-Univ. Paris VI, Paris-Grignon, France*
[d] *Faculty of Bioengineering, Agronomy and Environment, Department of Environmental Sciences and Land Use Planning, UCL, Louvain La Neuve, Belgium*

Received 3 February 2006; received in revised form 25 July 2006; accepted 27 July 2006

KEYWORDS
Solute transfer;
Water table;
Fluctuation zone;
Groundwater;
Multitporosity;
Tracer

Summary This study deals with solute transfer in the vertical continuum between the unsaturated zone and shallow groundwater of a weathered granite aquifer in the Kerbernez headwater catchment of western France. The objectives are (i) to determine the mechanisms responsible for solute transfer in the unsaturated and water-table fluctuation zones of the aquifer, and (ii) to analyse the implications of these results on solute transfer times at the catchment scale. An experimental site located in the plateau area of the catchment was equipped with 6 tensiometers, 18 ceramic cups at depths from 0.25 to 2.5 m and 7 piezometers from 3 to 20 m. Measurements of hydraulic head and water sampling were carried out over a period of 2.5 years in the unsaturated zone (0–2 m), the water table fluctuation zone (2–9 m) and the permanently saturated zone (>9 m). Two tracer experiments were carried out by applying two pulses of water, one enriched with deuterium and the other with bromide. Natural chloride concentrations, as well as deuterium and bromide concentrations, were analysed from solution samples. From the artificial tracer concentrations, two porosity compartments can be identified and partly quantified: (1) the slow-mobile porosity (36% of the bulk volume), accounting for the slow piston-flow transfer (2–3 m per year), and (2) the rapid-mobile porosity, which transfers small quantities of bromide at a rate of 19 cm h^{-1} down to the water table. Natural

* Corresponding author. Address: Unité de génie rural, Croix du Sud 2, bte 2, B-1348 Louvain La Neuve, Belgium. Tel.: +32 10 47 36 90; fax: +32 10 47 38 33.
E-mail address: legout@geru.ucl.ac.be (C. Legout).

0022-1694/$ - see front matter
doi:10.1016/j.jhydrol.2006.07.017

chloride concentrations are characterised by a high temporal variability in the water-table fluctuation zone, whereas the concentrations remain steady in time in the permanently saturated zone (42 mg l^{-1} at 20 m depth). The temporal variability is related to the water-table fluctuations and follows the same pattern each hydrological year, i.e. low concentrations during rising water-table followed by a progressive increase in concentrations during the periods of high piezometric level and water-table recession. This pattern is explained in terms of the two mobile porosity compartments and groundwater hydraulics. Based on these findings, we propose a conceptual model of solute transfer along the hillslope of a headwater catchment. We conclude that mixing in the water-table fluctuation zone could occur at two spatial scales. Firstly, at the pore scale, with mixing of waters in slow mobile and rapid mobile porosity, and secondly, at the scale of the hillslope. The mixing at this latter scale would appear to result from differences of flow path geometry and velocity between the unsaturated zone and the groundwater.

Introduction

In watershed underlain by crystalline bedrock, most of the solutes and water fluxes in the stream come from shallow groundwater that accumulates above and within the fractured crystalline bedrock. One of the key characteristics of the hydrology and hydrochemistry of these catchments is that the stream flow and water-table variations are highly reactive to rainfall, whereas the solute concentrations remain steady in time or are strongly damped in the streams and the deep groundwater (Kirchner et al., 2001; Molenat et al., 2002; Martin et al., 2004). Recent reviews have indicated that solute transfer in headwater catchments are characterised by wide time scales ranging from days to years, and that we require a better understanding of the flow paths and transfer mechanisms involved on such time scales (Mc Donnell, 2003; Kirchner, 2003). Furthermore, Kirchner (2006) underlined the need for a better temporal resolution as well as spatial resolution to analyse correctly solute transfer.

Although groundwater recharge is a key link in the transfer of solutes from the soil surface to streams, the process is still poorly understood, particularly in the interaction between the unsaturated zone and the groundwater. These two zones have generally been analysed separately. Most studies focusing on groundwater recharge assume that groundwater chemistry depends only on the solute concentration and volume of water reaching the water table (Landon et al., 2000; De Vries and Simmers, 2002). In the same way, classical transport models assume that, once the water and solutes have reached the water table, they are completely and instantaneously mixed with the pre-event groundwater (Beaujouan et al., 2002; Wade et al., 2002). However, in recent studies, Silliman et al. (2002) and Berkowitz et al. (2004) pointed out that a steady water table is a highly active and complex zone in terms of water and solute mixing. These processes include not only vertical flow from the recharge but also lateral flow from upslope. Moreover, little field evidence is available to support the assumption of complete and instantaneous mixing between water flowing vertically from the unsaturated zone and pre-event groundwater. Such an assumption is therefore highly questionable, and all the more so in catchments such as those in Western France where the free groundwater surface is often very reactive to rainfall and exhibits large fluctuations within the weathered material on the year scale (Martin et al., 2004; Molenat et al., 2005). Weathered material differs from other multiporosity materials such as soils, since it retains much of the structure and regularity of the bedrock, including rapid flow through relict fractures (Van Der Hoven et al., 2003) and variable exchanges between the different porosity compartments. Currently, little is known about the water and solute transfer in such materials, as well as the mixing processes involved. Generally speaking, we do not have a good understanding of the impact of variably saturated conditions, such as those induced by water table fluctuations, on flow and solute transfer processes (Hinz, 1998; Sinke et al., 1998).

In this paper, we focus on vertical solute transfer in the unsaturated zone-groundwater continuum of a weathered granite aquifer in a headwater catchment. The study aims at assessing the water flow and solute transfer processes occurring above, at and below the water table. For this purpose, we carried out a field investigation to (i) determine the mechanism responsible for solute transfer in the unsaturated and water-table fluctuation zones, and (ii) analyse the implications of these results on solute transfer times at the catchment scale. The experimental site is located in the Kerbernez catchment, western France. The arrangement of instruments on this site allows water sampling and the measurement of hydraulic head at various depths in the unsaturated zone, the water table fluctuation zone and the permanently saturated zone of the aquifer. Solute transfer was analysed by monitoring natural chloride concentrations and performing two tracer experiments.

Materials and methods

The Kerbernez catchment

The Kerbernez catchment, previously described by Ruiz et al. (2002) and Martin et al. (2004), is located in south-western Brittany (Fig. 1a). It is an agricultural headwater catchment covering an area of 0.12 km^2. The climate is oceanic with a mean annual precipitation of 1167 mm over the last decade (Standard deviation, SD = *195 mm*) and a mean annual Pennman potential evapotranspiration (PET) of 616 mm (SD = *71 mm*). Kebernez is an headwater catchment where runoff in excess of saturation occurs locally in the valley bottom when groundwater is at the soil surface. In