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Modeling sewage leakage and transport in carbonate aquifer using carbamazepine as an indicator

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ABSTRACT

The Western Mountain Aquifer (Yarkon-Taninim) of Israel is one of the country's major water resources and partially flows through a karst system. During late winter 2013, maintenance actions were performed on a central sewage pipe that caused sewage to leak into the creek located in the study area. Carbamazepine (CBZ) was used as an indicator for the presence of sewage in the groundwater. The research goal was to develop a mathematical model for quantifying flow and contaminant transport processes in the karst/fractured-porous unsaturated zone and groundwater system. The model was used to simulate CBZ transport during and after an observed sewage leakage event. A quasi-3D dual permeability numerical model represents the 'vadose zone – aquifer' system, by a series of 1D vertical flow and transport equations solved in a variably-saturated zone and by 3D-saturated flow and transport equation in groundwater. The results of simulation showed that after the leakage stopped, significant amounts of CBZ were retained in the porous matrix of the unsaturated zone below the creek. Water redistribution and slow recharge during the dry summer season contributed to a continuous supply of CBZ to the groundwater in the vicinity of the creek and hundreds of meters downstream.

1. Introduction

Sewage infiltration and pollution of groundwater is a worldwide problem, all the more so in carbonate aquifers where preferential flow paths exist. In order to better understand the potential threat from sewage pollution on vulnerable groundwater sources, there is a need to predict and quantify sewage infiltration processes. However, the complicated nature of the interactions between the matrix and conduits (Geyer et al., 2007) is the main obstacle to making predictions about sewage infiltration in flow and transport systems such as carbonate aquifers.

Only a few contaminants found in wastewater which originate from anthropogenic use can also serve as good indicators for anthropogenically-impacted water sources, such as untreated/treated sewage. Among those contaminants, the micro-contaminant Carbamazepine (CBZ) is both widely used and approved as a stable indicator for sewage contamination in groundwater (Clara et al., 2004; Fenz et al., 2005; Gasser et al., 2010). Moreover, CBZ's conservative nature has also been supported by sand column experiments, where partition coefficients values (K_D) were 0.21–5.32 l kg^{-1} (Scheytt et al., 2005). Rona et al. (2014) developed a 3D flow and transport model to simulate groundwater contamination due to wastewater recharge using CBZ and chloride as tracers. Furthermore, Gasser et al. (2014) used a combination of chloride, acesulfame and CBZ to quantify the ratio between pristine water and effluent in Karst area springs. In the aforementioned study, a good correlation was observed between carbamazepine and acesulfame abundances, which the authors attributed to a single source for both contaminant and minimal retardation of the two contaminants in the karst aquifer. Despite evidence that CBZ has been shown to sorb into the upper soil layers that are rich in organic matter on some level (Arye et al., 2011; Chefetz et al., 2008; Williams and McLain, 2012), the overall transport of CBZ is conservative even in soil aquifer treatment (SAT) system sites (Arye et al., 2011). As such, CBZ was found to be a

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suitable tracer for karst flow systems analysis.

The fate of CBZ in karst systems has not been adequately studied (Hillebrand et al., 2012a). Several studies of karst system springs have implied that CBZ is persistent in aquifers, given that CBZ was detected in groundwater at background level concentrations (Doummar et al., 2014; Gasser et al., 2014) (10.3 ng/l, up to 30 ng/l respectively). Manamsa et al. (2015) showed a link between surface water maximum CBZ concentrations and its maximum concentrations in a chalk aquifer. In another study, Doummar et al. (2014) presented CBZ introduction into the system through fast flow pathways with a rapid flow component. However, Hillebrand et al. (2012b) concluded that CBZ was ineffective as an indicator in the studied karst system since it was only detected during the spring season at very low concentrations that did not exceed the method quantification limit. The above studies indicate that CBZ flow and transport in carbonate systems is highly affected from interactions between the matrix and the karst/fracture (conduits) flow paths.

The major gap in current studies which limits the efficiency of evaluating CBZ as a stable indicator in karst systems is that the CBZ transport has yet to be studied with a model using the dual continuum approach. Dual continuum models integrate two (or more) different conceptual approaches (MacQuarrie and Mayer, 2005; Kurtzman et al., 2016). Such models may more accurately predict CBZ's fate in carbonate flow systems since the mechanism of exchange between the matrix and the conduits may have a significant effect on the CBZ transport.

The objective of this study was to develop and apply a dual permeability mathematical model for simulating water flow and CBZ transport processes in karst/fractured-porous unsaturated zone and groundwater system. The simulations were used to characterize the processes of CBZ natural attenuation from the initial input concentration at the ground surface into the vadose zone and throughout the aquifer. Our hypothesis was that CBZ can serve as an indicator for wastewater migration in the matrix and along conduits in the unsaturated zone and groundwater. This paper details the flow and transport model used to describe sewage infiltration through a thick, unsaturated zone and its dilution in the groundwater of a carbonate aquifer.

2. Materials and methods

2.1. Sewage discharge event

The study area is located on the Sorek creek west of the city of Jerusalem, Israel (Fig. 1). At this site, the Sorek creek drains a catchment area of approximately 88 km^2. 2.05 km upstream, a reservoir (Beit Zait) collects runoff water and limits downstream natural surface flow. Reservoir dam overflows take place every few rainy seasons. During the spring season water is regularly released from the dam downstream.

The local geology of the study area is composed of the carbonate Judea group (Fig. 2). The groundwater level is deep, ranging from tens of meters to a couple hundred meters. The general flow direction is south-west. The characteristics of the thick, unsaturated flow were described by Dvory et al. (2016), who revealed a relationship between precipitation, groundwater recharge and groundwater level.

The current research examines an event which occurred in 2013, when sewage was discharged from a major sewage pipeline located along the Sorek creek. This pipeline collects sewage from West Jerusalem and other local settlements and diverts it towards a treatment plant (Fig. 1). As a result of maintenance actions between April 2nd and April 19th, 2013, sewage was discharged on five separate occasions into the natural creek (Fig. 1; Fig. 3c). In order to limit additional contamination, a collection pipe was setup 2.45 km downstream of the discharge site. Five days later, the stream was flushed with fresh water which was released from the Beit Zait dam/reservoir. A total of 28,125 m^3 of sewage was released into the creek during this period, of which, 5553 m^3 was collected by the collection pipe back to the sewage system, while the rest infiltrated into the subsurface. Then, 32,069 m^3 of reservoir water flushed the stream bedrock and infiltrated into the subsurface. Infiltration volumes of both the sewage and the fresh water were used in the models to prescribe the boundary condition along the creek bed. CBZ was used as an indicator for identification and quantification of sewage entering the groundwater.

2.2. Sampling and monitoring

In order to monitor the fate of the discharged sewage, water sampling and hydrological monitoring of both surface water and groundwater was performed. Prior to the sewage discharge event, samples were taken from the Beit Zait reservoir and directly from the Sorek sewage pipeline. Groundwater sampling was carried out at the depth of 100 m below ground surface, in the EK11 observation well (Fig. 1). The local geology in the area of the observation well is characterized by an extensive conduit network that continues from the ground surface to the aquifer, therefore the groundwater level has very fast response to infiltration events (Dvory et al., 2016). Groundwater samples were collected over a period of 310 days. In total, twenty three groundwater samples were taken. The sampling interval varied between one and fifty six days. The sampling interval was shorter during the expected tracer breakthrough time, thus a high temporal resolution of the CBZ tracer breakthrough curve (BTC) was achieved. A dual groundwater level and temperature data logger was installed in the EK11 observation well in the Soreq creek to make measurements every 30 min (Solinst Levelogger). Data on measured hourly precipitation and evaporation rates were obtained from the Israel Meteorological Service (IMS) Tzuba Station. Sewage and surface water runoff discharges were acquired from gauging stations (Fig. 1).

2.3. Laboratory analysis

CBZ was analysed by GC/MS according to the EPA method 8270. D and by LC/MS/MS according to the EPA method 1694. For both methods CBZ was first extracted from natural samples by Solid Phase Extraction (SPE) using C18 Empore high performance extraction disks. Aliquots of 1 μL of the final extracts were separated at Agilent Gas Chromatograph 6890N (DB-5MS column of 0.25 × 30 m, 1μ) with MS detector 5973. Analyte was quantified in full-scan (m/z 44–450, 0.4 s/scan) or selected ion monitoring (SIM) mode using the ions 193 for quantification and 236, 165 and 192 ions for confirmation. The Limit of Quantification (LOQ) for GC/MS analysis was 100 ng/l. For samples with CBZ concentrations less than the GC/MS method LOQ, the analysis was performed using an Agilent G6410A Triple Quadruple mass spectrometer (QQQ) with an electrospray ionization source. For this type of analysis, analyte separation was conducted with Agilent ZORBAX Eclipse Plus C18 column (2.1 × 100 mm, 3.5 μm). The multi-reaction monitoring (MRM) transition of mass 237 to 194 was used for quantification and mass 192 to 179 for confirmation were used for CBZ determination. Quantification of the analyte was done with respect to CBZ-d_{10} (transitions of mass 247 to 204) purchased from the C/D/N Isotopes Ltd, Pointe-Claire, Canada. The LOQ of CBZ by LC/MS was 0.5 ng/l.