Chemosphere 115 (2014) 47–53

Contents lists available at ScienceDirect

Chemosphere

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

Occurrence of carbamazepine and five metabolites in an urban aquifer

Anna Jurado [a,b,*], Rebeca López-Serna [c], Enric Vázquez-Suñé [a], Jesus Carrera [a], Estanislao Pujades [a,b], Mira Petrovic [d], Damià Barceló [c,d]

[a] GHS, Department of Geosciences, Institute of Environmental Assessment & Water Research (IDAEA-CSIC), Jordi Girona 18-26, 08034 Barcelona, Spain
[b] GHS, Dept Geotechnical Engineering and Geosciences, Universitat Politècnica de Catalunya, UPC-Barcelona Tech, Jordi Girona 1-3, 08034 Barcelona, Spain
[c] Department of Environmental Chemistry, Institute of Environmental Assessment & Water Research (IDAEA-CSIC), Jordi Girona 18-26, 08034 Barcelona, Spain
[d] Catalan Institute for Water Research (ICRA), Emili Grahit 101, Edifici H2O, Parc Científic i Tecnològic de la Universitat de Girona, 17003 Girona, Spain

HIGHLIGHTS

- Urban groundwater (GW) contaminated by carbamazepine (CBZ) and 5 metabolites (MET).
- High concentrations are found when the aquifer is infiltrated by a polluted river.
- The fate of CBZ and METs appeared to be dependent on the GW redox conditions.
- Residence time could also play a major role in controlling their fate in aquifers.

ARTICLE INFO

Article history:
Received 25 July 2013
Received in revised form 11 December 2013
Accepted 6 January 2014
Available online 21 February 2014

Handling Editor: J. de Boer

Keywords:
Urban aquifer
Carbamazepine
Human metabolites
Redox conditions
Field study

ABSTRACT

This paper deals with urban groundwater contaminated with carbamazepine (CBZ) and five of its human metabolites in Barcelona. Groundwater samples were accordingly collected in the aquifers of Poble Sec and Besòs River Delta. Higher concentrations and more compounds were found in the Besòs River Delta aquifer, which is recharged by a river contaminated with treated effluent from numerous treatment plants. By contrast, the urban area of Poble Sec presented lower concentrations and fewer compounds. The results showed that CBZ could be attenuated in the Poble Sec aquifer since concentrations in groundwater were lower than those evaluated from mixing of the recharge sources. Conversely, CBZ and its human metabolites were not removed under the reducing conditions of the Besòs River Delta aquifer probably because of the short residence time in this aquifer.

1. Introduction

Pharmaceutically active compounds (PhACs) including their human metabolites have become a matter of concern because they probably have a deleterious effect on ecosystems and human health. They may enter surface and groundwater via a number of pathways, especially from raw sewage and waste water treatment plant (WWTP) effluents. As a result, a wide range of PhACs exert an adverse influence on groundwater quality.

Of the PhACs, carbamazepine (CBZ) is one the most important with respect to the environment. CBZ shows no or low removal during wastewater treatment. Effluent concentrations are often reduced by no more than 10% in most of WWTPs (Zhang et al., 2008). Moreover, some metabolites of CBZ have been detected at concentrations higher than the parent compound in WWTP influents (Gros et al., 2012) and also in finished drinking water (Huerta-Fontela et al., 2011).

Most research into CBZ and its metabolites has been focused on wastewater. Methods such as UV irradiation and ozonation have proved to be effective in removing these compounds from wastewater and/or from raw waters intended for drinking water (Kosjek et al., 2009; Lekkerkerker-Teunissen et al., 2012).

In groundwater, CBZ has been studied at bank filtration sites (Heberer et al., 2004; Massmann et al., 2008), artificial aquifer recharge (Drewes et al., 2003); and irrigated farms (Ternes et al., 2007; Chávez et al., 2011). CBZ is one of the most frequently detected pharmaceuticals in groundwater (Rabiet et al., 2006; Fenet et al., 2012; Kuroda et al., 2012; Reh et al., 2013). It has been detected at concentrations up to 2100 ng L^{-1} in an aquifer seriously

* Corresponding author at: Jordi Girona 18, 08034 Barcelona, Spain. Tel.: +34 934006100; fax: +34 932045904.
E-mail address: annajuradoelices@gmail.com (A. Jurado).

http://dx.doi.org/10.1016/j.chemosphere.2014.01.014
0045-6535/

affected by sewage water discharge (Müller et al., 2012). In fact, CBZ has been considered to be a wastewater indicator in aquatic environments because of its persistence (Strauch et al., 2008; Wolf et al., 2012).

The widespread occurrence of CBZ in groundwater highlights the need to better understand its fate in aquifers, which depends on its physicochemical properties and other environmental characteristics such as residence time and aquifer redox conditions (Lapworth et al., 2012). CBZ degradability has been studied in artificial aquifer recharge (Patterson et al., 2011), in water reuse (Tiehm et al., 2011) through laboratory column experiments and in batch experiments (Barbieri, 2011). The aforementioned studies have pointed out that CBZ is not degraded under aerobic or anaerobic conditions. Conversely, it is more efficiently biodegraded by the fungus *Trametes versicolor* (Jelic et al., 2012). Little research has been undertaken on the occurrence of CBZ metabolites in groundwater (Kahle et al., 2009; Fenet et al., 2012) and no works have addressed the contamination of CBZ metabolites in urban groundwater.

The present study is concerned with the occurrence of CBZ and its metabolites in urban groundwater, taking into consideration common contaminants and the redox conditions of the aquifers of Barcelona.

2. Materials and methods

2.1. Site description

The study area is located in NE Spain and includes Barcelona and part of its metropolitan area. The area is bounded by the Serra de Collserola (Catalan coastal ranges) and the Mediterranean Sea (Fig. 1), both boundaries running approximately NNE–SSW. Other boundaries are constituted by the Llobregat (SW) and the Besòs (NE) rivers. The climate is Mediterranean and the rainfall averages about 600 mm/year. Fig. 1 shows the different aquifers that crop out in Barcelona.

The aquifers of Barcelona are recharged by a number of sources (Vázquez-Suñé et al., 2010). Direct rainfall recharge occurs in the non-urbanised areas in the Collserola Range (REC). Seawater intrusion (SEA) and water from the heavily polluted river Besòs (RIV) are potential recharge sources in low-lying areas. Other sources of groundwater recharge include loss from the water supply network from the Ter and Llobregat rivers (TER and LLOB). The city is therefore divided into two zones with a different water quality with the result that two chemical compositions are found in the waste water (SW T and SW LL). In paved areas, runoff water recharges the aquifers through direct infiltration or sewer loss (RUNOFF).

The aforementioned water quality division can be confirmed by comparing major and minor ion concentrations and some redox indicators in the Poble Sec and Besòs River Delta aquifers (Table S1 of Supplementary data). In general, the groundwater of the Besòs River Delta is less mineralised than that of Poble Sec since the average concentrations of chloride, sulphate, calcium, magnesium, nitrate and dissolved oxygen are much lower (Table S1). Both zones have neutral pHs. Some redox indicators such as the high concentrations of dissolved oxygen and nitrate (on average 4 mg L^{-1} and 95.7 mg L^{-1}, respectively) and the low or null concentrations of ammonium (on average 0.03 mg L^{-1}) have confirmed the oxidising conditions of the Poble Sec aquifer.

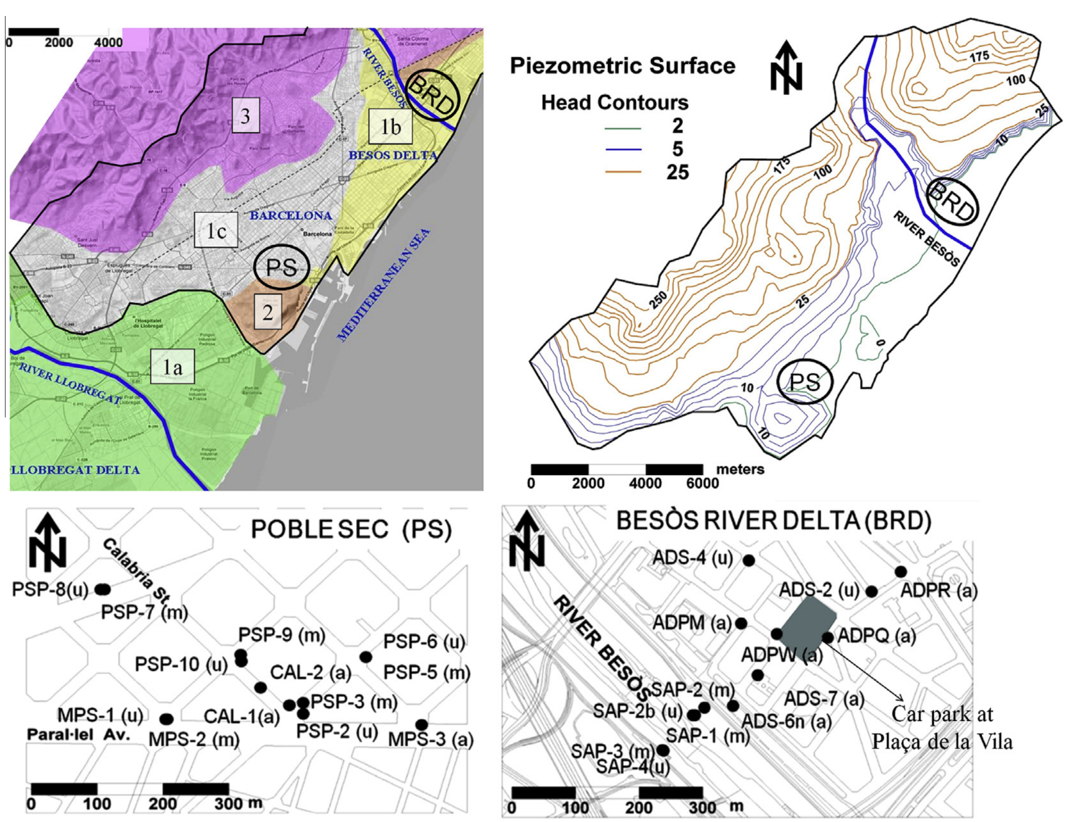

Fig. 1. On the right, a piezometric map of the study area, which is divided into two zones: Poble Sec and Besòs River Delta. On the left, schematic description of the hydrogeology of Barcelona: (1a) Llobregat Delta made up of gravels, sands, silts and clays (Holocene, Quaternary), (1b) Besòs Delta composed of gravels, sands, silts and clays (Holocene, Quaternary), (1c) Barcelona Plain consisting of carbonated clays (Pleistocene, Quaternary), (2) Barcelona Plain made up of marls, sandstones and sands (Tertiary) and (3) Collserola Range consisting of shale and granites (Palaeozoic). At the bottom, observation points on each zone, including the depth of the screen: (u) upper, (m) middle, (l) lower and (a) totally screened (modified from Jurado et al., 2012).