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Photochemical Behaviour of Carbamazepine in Natural Waters

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Abstract : Pharmaceuticals in the environment have become a very hot topic in the recent years. This interest is related to the large amounts dispensed and to their release in urine or faeces from treated patients, resulting in their ubiquitous presence in water resources and wastewater treatment plants (WWTP) effluents. Thereby, many studies focused on the prediction of pharmaceuticals' behaviour, to assess their fate and impacts in the environment. Carbamazepine is a widely consumed psychotropic pharmaceutical, thus being one of the most commonly detected drugs in the environment. This organic pollutant was proved to be persistent, especially with respect to its non-biodegradability, rendering it recalcitrant to usual biological treatment processes. Consequently, carbamazepine is very little removed in WWTP with a maximum abatement rate of 5 % and is then often released in natural surface waters. To better assess the environmental fate of carbamazepine in aqueous media, its photochemical transformation was undertaken in four natural waters (two French rivers, the Berre salt lagoon, Mediterranean Sea water) representative of coastal and inland water types. Kinetic experiments were performed in the presence of light using simulated solar irradiation (Xe lamp 300W). Formation of short-lifetime species was highlighted using chemical trap and laser flash photolysis (nanosecond). Identification of transformation by-products was assessed by LC-QToF-MS analyses. Carbamazepine degradation was observed after a four-day exposure and an abatement of 20% maximum was measured yielding to the formation of many by-products. Moreover, the formation of hydroxyl radicals (•OH) was evidenced in waters using terephthalic acid as a probe, considering the photochemical instability of its specific hydroxylated derivative. Correlations were implemented using carbamazepine degradation rate, estimated hydroxyl radical formation and chemical contents of waters. In addition, laser flash photolysis studies confirmed •OH formation and allowed to evidence other reactive species, such as chloride ($Cl_2^{\bullet-}$)/bromine ($Br_2^{\bullet-}$) and carbonate ($CO_3^{\bullet-}$) radicals in natural waters. Radicals mainly originate from dissolved phase and their occurrence and abundance depend on the type of water. Rate constants between reactive species and carbamazepine were determined by laser flash photolysis and competitive reactions experiments. Moreover, LC-QToF-MS analyses of by-products help us to propose mechanistic pathways. The results will bring insights to the fate of carbamazepine in various water types and could help to evaluate more precisely potential ecotoxicological effects.

Keywords : carbamazepine, kinetic and mechanistic approaches, natural waters, photodegradation
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