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Oxidation of diatrizoate in aqueous phase by advanced oxidation processes based on solar radiation



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ABSTRACT

The aim of this study was to investigate the use of solar radiation to treat recalcitrant pollutants, using sodium diatrizoate (DTZ), an ionic X-ray contrast media, as reference compound. The effectiveness in DTZ removal was compared among solar radiation, H₂O₂, solar radiation/H₂O₂, K₂S₂O₈, solar radiation/K₂S₂O₈, Fenton, solar radiation/Fenton, Fenton-Like, and solar radiation/Fenton-Like systems. Using direct DTZ photolysis, the percentage DTZ degradation is low and independent of the medium pH, but not of the irradiance level. However, in the presence of radical-promoting species, 100% DTZ degradation is achieved. Results obtained with solar radiation/Fenton and solar radiation/Fenton-like systems suggest that the formation of peroxy and iron hydroperoxy complexes susceptible of absorbing radiation in the solar radiation area represents a new source of OH[•] and/or OH₂[•] radicals in the medium, favoring DTZ degradation. The byproducts formed in each process and the post-treatment medium toxicity were studied, observing an increased toxicity after all oxidation treatments, indicating that the degradation products generated are more toxic than DTZ. Moreover, a direct relationship was found between percentage DTZ degradation and aqueous medium toxicity.

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1. Introduction

Over the past few years, considerable attention has been paid to the treatment of organic compounds known as emerging contaminants (ECs) present in waters, including personal care products, detergents, and pharmaceutical products, among others. Most ECs are partially degraded by wastewater treatment in municipal wastewater treatment plants (MWTPs), while others remain unaltered and are detected in the plant effluents [1–4]. ECs concentrations detected range from ng L⁻¹ to µg L⁻¹, but their wide diversity and continuous release into the environment mean that they pose a grave threat to the quality of waters [5–10].

ECs include iodinated contrast media, including sodium diatrizoate (DTZ). In common with all contrast media, DTZ is highly polar, stable, and chemically inert, and is eliminated from the organism unmetabolized. Conventional MWTP treatments are

not effective to remove this contrast medium [11–16], which is therefore released into the environment. DTZ has been detected in surface waters [12,13,17,18], groundwaters [19,20], and even in waters intended for human consumption [21,22]. DTZ has also been detected in the effluents of MWTPs [15,16]. Advanced oxidation processes (AOPs) have been proposed as an option in the tertiary treatment of MWTP effluents, because they achieve the transformation/elimination of a wide large variety of ECs and frequently increase their biodegradability and/or reduce their toxicity [23–27]. The operative costs of AOPs remain elevated but can be markedly reduced by combining these processes with solar radiation [28,29].

Various AOPs have been used for DTZ removal, obtaining different results, as follows: (i) ozone treatment showed a low effectiveness, reaching a percentage DTZ degradation of only 14% [30,31]; (ii) Fenton's reagent and UV/Fenton achieved 41.5% and 64.7% DTZ removal, respectively [32]; (iii) UV-A radiation, using titanium oxide as catalyst, obtained 65% removal after one hour of treatment [33]; (iv) non-thermal plasma reached 100% degradation after six hours of treatment [34]; and (v) advanced oxidation reduction processes (AORPs) based on gamma radiation achieved 100% removal at doses of only 1000 Gy [4,35].

With this background, the objective of this study was to investigate the removal of sodium DTZ using different AOPs in

Abbreviations: ANOVA, Analysis of variance; AOPs, Advance oxidation processes; AORPs, Advanced oxidation reduction processes; DTZ, Sodium Diatrizoate; ECs, Emerging contaminants; HPLC, High performance liquid chromatography; MWTP, Municipal wastewater treatment plant; P, P-value; SR, Solar radiation; UPLC, Ultra performance liquid chromatography.

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Table 1
Influence of irradiance and solution pH on DTZ degradation. $[DTZ]_0 = 25 \text{ mg L}^{-1}$.

Irradiance (W m^{-2})	pH	$k \times 10^3$ ($\text{M}^{-1} \text{s}^{-1}$)	% Degraded
300	2.0	0.88 ± 0.07	8.27
300	6.5	0.95 ± 0.07	8.27
300	9.0	0.96 ± 0.07	9.27
300	12.0	0.89 ± 0.07	7.26
450	6.5	2.10 ± 0.20	16.44
610	6.5	2.30 ± 0.20	17.69

combination with solar radiation, analyzing: (i) DTZ degradation kinetics by direct solar photolysis; (ii) DTZ photodegradation with solar radiation in the presence of hydrogen peroxide, potassium peroxodisulfate, or Fenton's reagent; (iii) the mechanism of formation of the different degradation products generated by each process and (iv) the time course of toxicity with each treatment system studied.

2. Materials and methods

2.1. Materials

All chemical reagents used (sodium diatrizoate, phosphoric acid, potassium peroxodisulfate, sodium sulfite, ferrous sulfate, ferric chloride, potassium ferrate hydrochloric acid, sodium nitrite) were of high purity analytic grade and supplied by Sigma–Aldrich. All solutions were prepared with ultrapure water obtained using Milli-Q[®] equipment (Millipore).

2.2. Experimental

2.2.1. Solar simulator device

The solar simulator used was a model 1500 Solarbox, equipped with a Xenon lamp (irradiance range from 250 to 1100 W m^{-2}) that supplies radiant energy in a spectral range from 290 to 800 nm. The photodegradation experiments were conducted in an irradiance range of $300\text{--}610 \text{ W m}^{-2}$ without UV filter.

2.2.2. DTZ degradation by solar radiation in the presence of radical promoter species

DTZ solar photodegradation experiments were conducted using a solution of 25 mg L^{-1} DTZ with the appropriate concentration of H_2O_2 , $\text{K}_2\text{S}_2\text{O}_8$, $\text{Fe(II)/H}_2\text{O}_2$, $\text{Fe(III)/H}_2\text{O}_2$; 100 mL of each solution were treated in the solar simulator, drawing samples at different exposure times to determine the corresponding DTZ degradation kinetics. The reaction tubes were made of glass and had an inner diameter of 7.5 cm and height of 4 cm.

2.3. Analytical procedures

2.3.1. DTZ determination in aqueous solution

DTZ concentration in aqueous solution was determined by high performance liquid chromatography (HPLC) in reverse phase, using a liquid chromatograph (Thermo-Fisher) equipped with a visible/UV detector and autosampler with capacity for 120 vials. A PHENOMENEX Kinetex C-18 chromatographic column was used ($2.6 \mu\text{m}$ particle size; $4.6 \times 150 \text{ mm}$). The mobile phase delivered in isocratic mode was 70% 0.1% formic acid solution (v/v) and 30% 0.1% acetonitrile solution at a flowrate of 0.6 mL min^{-1} . Detector wavelength was 254 nm and injection volume was $100 \mu\text{L}$.

2.3.2. Identification of DTZ degradation byproducts

The different DTZ degradation byproducts were determined using an ultra-high pressure liquid chromatograph (UPLC) (Waters, ACQUITY H CLASS) equipped with mass spectrometer

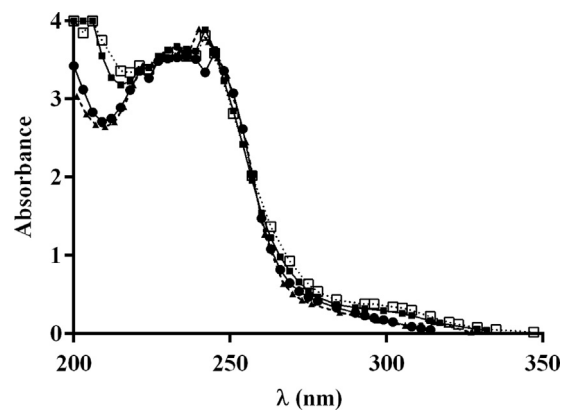


Fig. 1. DTZ absorption spectrum as a function of medium pH. (●), pH=2.0; (▲), pH=6.5; (■), pH=9.0; (□), pH=12.0.

and time-of-flight (TOF) analyzer (Waters, LCT Premier Xe), and an Acquity BEH C18 column with a diameter of 2.1 mm, length of 50 mm, and particle size of $1.7 \mu\text{m}$. Water with 0.1% formic acid was used for mobile phase A and acetonitrile with 0.1% formic acid for mobile phase B. The mobile phase gradient elution was programmed as follows: $t_0 = 95.0\%$ A and 5.0% B; $t_8 = 0.0\%$ A and 100.0% B; $t_{10} = 0.0\%$ A and 100.0% B; $t_{12} = 95.0\%$ A and 5.0% B. The flow rate was set at 0.30 mL min^{-1} throughout the gradient.

2.3.3. Determination of byproduct toxicity

Degradation byproduct toxicity was determined by normalized biotest (DIN/EN/ISO 11348-2) of the inhibition of *Vibrio fischeri* bacteria (NRRL B-11177) [36]. The decrease in bioluminescence was measured in LUMISTOX 300 equipment [37], and results were expressed as percentage inhibition at 15 min of exposure with reference to stock saline solution (control).

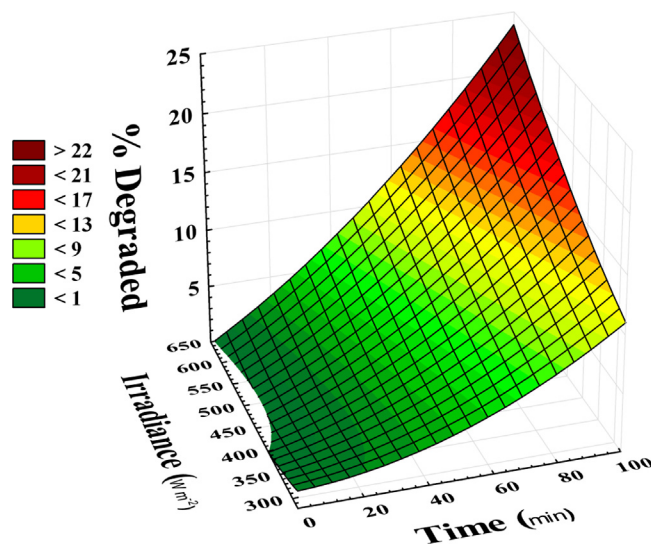


Fig. 2. Response surface relating percentage DTZ degradation to treatment time and irradiance. $[DTZ]_0 = 25 \text{ mg L}^{-1}$, pH=6.5.

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