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Static and Continuous flow-through pulsed light technology for pesticide abatement in water

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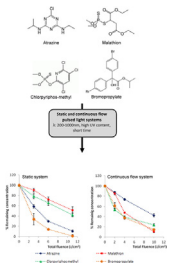
HIGHLIGHTS

- Water containing four pesticides was decontaminated by pulsed light technology.
- Efficacy of static and continuous flow-through pulsed light systems were compared.
- Pesticide degradation by pulsed light followed cero and first-order kinetics.
- Degradations up to 85 and 93% were observed after total fluences of 10–11 J/cm^2.
- The degradation pathways included several successive and competitive steps.

GRAPHICAL ABSTRACT



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ABSTRACT

Perspectives concerning pulsed light (PL) technology as a novel water decontamination treatment are overviewed in this work. Degradation of atrazine, malathion, chlorpyriphos-methyl and bromopropylate in aqueous solutions at different concentrations was performed employing static and continuous flow-through PL units. Results for both PL systems were compared in terms of efficacy of pesticides degradation and derived photoproducts formed. The pesticides degradation increased with the applied total fluence for both PL systems. In general, PL induced rapid degradation of all studied compounds although some differences were observed in the rate of degradation among them. The most intense treatment in the static unit (11 J/cm^2) yielded degradations between 60 and 85% whereas degradations between 74 and 93% were observed after exposure to slightly lower fluences (10 J/cm^2) in the dynamic unit. For both PL systems bromopropylate was the most affected compound. Photodegradation products arose from chemical reactions involving structural changes like dehalogenation, desulfuration, dealkylaytion and oxidation of the alkyl chains.

Although further research is needed before an efficient application in full-scale systems, PL appears as a promising technology in order to reduce the presence of pesticides residues in process or waste waters, allowing thereby recirculation of these waters with the subsequent economic and environmental advantages.

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Abbreviations: PL, pulsed light; IS, internal standard; Abs, absorbance; P, product; LC-DAD-MS, liquid chromatography-diode array- mass spectrometry; MSD, mass selective detector; APCI, atmospheric pressure chemical ionization; MRM, multiple reaction monitoring; EIC, extracted ion chromatogram; SIM, single ion monitoring; H, total fluence; C_o, initial concentration; C_H, concentration at a given fluence; k, degradation rate constant; $H_{1/2}$, half fluence; BPC, base peak chromatogram; t_R, retention time.

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1. Introduction

The use of pesticides in agriculture has increased during the last century due to the need of increasing the food production despite their potential hazardous impact on human and aquatic organisms. Different cleaning methods, such as boiling or washing, are carried out by the food processing industry in order to reduce concentrations or remove pesticide residues from agricultural products such as fruits and vegetables. Pesticides could be thereby transferred from these products into processing water, being the amount of subsequent contaminated wastewater a problem of concern once these waters are discharged in rivers, lakes and coastal areas due to the toxicity and low biodegradability of these pollutants. This fact together with the current need to minimize process water consumption and wastewater discharge rates has led to the development of alternative technologies for water decontamination and pollution prevention [1].

Pulsed light (PL) is an emerging non-thermal process that has a considerable potential to be implemented in the food industry for decontaminating food products, food packaging materials and water [2–5]. This technology consists of a successive repetition of short-time (50–350 μs), intense and high power light pulses of broadband emission spectrum (200–1100 nm) with a considerable amount of light in the short-wave UV range [6,7]. It is considered a safe and environmentally friendly technology due to the use of mercury free xenon flash lamps and the absence of applied chemicals disinfectants or preservatives and waste effluents.

PL has been successfully tested for the inactivation of a wide broad of waterborne microorganisms, being considered a faster and more effective decontamination method than conventional continuous UV light [5,8].

However, chemical pollutants that absorb UV light could also be considered as potential targets of PL technology due to its broadband emission light and high UV content. In fact, recent works using static PL prototypes have shown the efficacy of this technology to degrade diverse pesticides in water of different origin [3,9]. However, the effectiveness of PL for degradation of a broader range of pesticides must be demonstrated in order to evidence its high potential for water pesticide decontamination. In addition, to the authors' knowledge, there is not published research about pesticide degradation in continuous flow-through units. Therefore, the effectiveness of this technology under flow conditions must also be demonstrated prior to its use for the treatment of industrial waters.

With the aim of increasing the understanding of PL technology for decontamination of pesticides in water, four different pesticides (atrazine, malathion, chlorpyriphos-methyl and bromopropylate) were subjected to diverse PL treatments. These compounds are widely used and frequently detected in shallow ground water [10] causing environmental concern because of their low biodegradability and moderate or high toxicity. Among them, only atrazine has been previously shown to be effectively degraded by PL although only static PL systems were employed [3,9]. In the present work, we go further by assessing the ability of both static and dynamic flow-through PL systems to decontaminate the selected four pesticides in water. Results for both PL systems were compared in terms of degradation effectiveness as well as generation of derived photoproducts.

2. Material and methods

2.1. Chemicals

Atrazine and simazine deuterated, simazine-d_5, as internal standard (IS) were supplied from Dr. Ehrenstorfer (GmbH, Augsburg, Germany). Malathion was purchased by Riedel de Haën (Seelze, Germany), chlorpyriphos-methyl was obtained from Chem Service (West Chester, PA, USA) and bromopropylate was supplied by Fluka (Sigma, Boenem, Belgium). All the standards (Fig. 1) had a purity of at least 99%.

Acetonitrile HPLC (analytical-reagent grad) was purchased from Merck (LiChrosolv, Darmstadt, Germany), of. The high purity water was obtained from a Milli-Q plus water system from Millipore (Bardford, MA, USA).

2.2. Pulsed light treatments

2.2.1. Static PL unit

A SBS-XeMatic-2L-A device (SteriBeam Systems GmbH, Kehl, Germany) was used for pulsed light treatments. For the emission of light pulses, the electric power is stored in an energy storage capacitor and quickly released to a xenon lamp emiting high intensity light pulses of 325 μs duration. The emitted light spectrum includes wavelengths from 200 to 1000 nm with approximately 40% of the emitted light in the UV range (20% UV-C, 8% UV-B, and 12% UV-A).

For PL treatments, spiked water samples in sterile quartz cuvettes of 1 mm light path (Abs 600 nm: 0.1) were placed at a constant distance (5.5 cm) from the light strobe. The degradation of the pesticides was induced by exposure to 6, 12 and 22 light pulses of 0.5 J/cm^2 pulse fluence (H_0).

2.2.2. Dynamic PL unit

A dynamic flow-through pilot unit (Maria PUD system, Claranor, Manosque, France) consisting on an electronics bay with a control, a loading and a cooling unit, and a hydraulic system with reactors, a launch tank and a volumetric circulating pump, was used for PL treatments. The duration of each light pulse was 300 μs and the emitted light spectrum comprises wavelengths from 200 to 1000 nm, with approximately 20% of the emitted light in the UV range (10% UV-C and 10% UV-B and UV-A).

A volume of 1 L of spiked water was pumped through a reactor of 6.23 mm of thickness and exposed to different number of pulses at flow-rate of 5 L/min and at voltage input of 3000 V up to a total fluence of 10 J/cm^2. These conditions were chosen considering a previous work which demonstrated a high potential of this system for microbial inactivation [11]. Prior and following each treatment the unit was cleaned to avoid any possible cross-contamination, by circulating chorine solution (200 μg/mL) for 10 min, followed by sterile deionized water for the same time.

For both PL systems, each treatment was carried out at room temperature (23 °C) at least three times and untreated contaminated samples were used as controls.

2.2.3. Total fluence determination

Decontamination of water samples treated by both static and dynamic units was expressed as a function of total fluence (H), or amount of photons striking on the sample per area unit, which was calculated for each different treatment condition by multiplying pulse fluence (H_0) by the number of emitted light pulses (n) [$H = H_0 \times n$] [4].

2.3. Analytical methods

High performance liquid chromatography, diode array and mass spectrometry (LC-UV-MS) was used to monitor the concentration of each pesticide in solution, (Supplementary data). The percentage of degradation of each pesticide was determined by comparison of remaining concentration of both reference and PL treated samples.

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