

# Photooxidation and biotrickling filtration for controlling industrial emissions of trichloroethylene and perchloroethylene

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## Abstract

A two-stage process integrating ultraviolet oxidation and biotrickling filtration (UV–BTF) was developed for treatment of industrial air streams contaminated with trichloroethylene (TCE) and perchloroethylene (PCE). Laboratory-scale studies demonstrated that the UV–BTF system consistently achieved contaminant removal efficiencies of 99–100% under optimal conditions. The UV–BTF process employs an advanced oxidation process (AOP) such as UV photooxidation as pretreatment for alleviating biological toxicity attributed to high and/or fluctuating contaminant concentrations, and for transforming the contaminants to more easily biodegradable products. The UV–BTF process showed significantly better performance than the BTF process without UV photooxidation pretreatment. Free-radical reaction mechanisms, pathways, and product formations were proposed for the UV photolysis of TCE and PCE. The photooxidation was mediated by chlorine atom reactions, and the principal products for TCE were phosgene, dichloroacetyl chloride (DCAC), low levels of trichloroacetyl chloride (TCAC), carbon monoxide, and hydrochloric acid; while those for PCE were phosgene, TCAC, low levels of DCAC, carbon dioxide, and hydrochloric acid. These products were identified and quantified by a combination of techniques, namely, gas chromatography–mass spectrometry (GC–MS) and Fourier transform infrared (FTIR) spectroscopy. These photooxidation products were completely removed in the BTF stage of the UV–BTF process, as indicated by subsequent GC–MS analyses. The destruction of photolysis products in the BTF stage occurred by a combination of hydrolysis, adsorption and microbial degradation.

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## 1. Introduction

Chlorinated volatile organic compounds (VOCs) such as trichloroethylene (TCE) and perchloroethylene (PCE) are widely used in cleaning, degreasing and fumigating operations, and therefore their direct emissions are common in various industries including electronics, automobile, furniture, appliances, and textiles. Contamination of gas streams by TCE or PCE is a major issue because these compounds are toxic, carcinogenic and extremely persistent in the environment, and

are classified as hazardous air pollutants by the United States Environmental Protection Agency (Prager and Hartmann, 2001; Yamazaki et al., 2004). Therefore, there is considerable interest in vapor phase treatments of gas streams contaminated with these pollutants owing to their toxicity. Conventional technologies such as incineration and adsorption used in the purification of contaminated gas streams present the following problems: formation of undesirable byproducts such as dioxins and furans; consumption of high energy, especially at low contaminant concentrations; and, generation of hazardous wastes. Traditional purge-and-treat technologies just transfer these VOCs from aqueous phase to gas phase, and therefore require post-treatment strategies for their ultimate destruction. Development of technologies with greater efficiency, cost-effectiveness, and environmental safety would indeed be beneficial.

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The present research focuses on an integrated system consisting of ultraviolet (UV) photooxidation followed by biotrickling filtration (BTF) referred to as the UV–BTF process, for purification of gas streams contaminated with TCE or PCE. The UV photooxidation serves as a pretreatment to the BTF stage, and has distinct advantages such as protecting the microbial population from toxic shock, and breaking down the target contaminants into more easily biodegradable compounds, thereby increasing the biological treatment efficiency. BTF was preferred to conventional biofiltration because control of the media pH could be exercised by continuous recirculation of acid-neutralizing (alkaline) solution. Owing to the formation of UV photooxidation products, the pH control strategy was critically important to prevent acid accumulation and microbial inhibition. The UV–BTF technology is conceptually attractive because photochemical oxidation and BTF play complementary roles in achieving high destruction efficiencies at low costs. For example, in the UV photooxidation of chlorinated VOCs, formation and accumulation of toxins is a primary concern. While most UV sources provide longer oxidation retention times or higher energy outputs to potentially achieve complete mineralization of products, they also entail higher capital and operating costs owing to greater energy consumption and larger reactor volumes. Therefore, biological treatment serves as a polishing step in degrading or mineralizing residual contaminants and oxidation products. In the BTF system, the photooxidation products of chlorinated organics are removed by hydrolysis followed by activated carbon adsorption and microbial degradation. The success of the UV–BTF technology was demonstrated for the treatment of gas streams contaminated with TCE and PCE in the electronics industry specializing in nano-devices. The technology had been awarded process patents in Taiwan (Den et al., 2001), and the United States (Den et al., 2003).

In the present study, TCE and PCE were chosen as target compounds at gas stream concentrations typically encountered in industrial emissions. Performance comparisons of the integrated UV–BTF system and the BTF system without UV oxidation evaluated the advantage of employing UV oxidation prior to biological treatment. UV photooxidation studies were conducted in plug-flow reactors to evaluate the gas-phase decomposition kinetics of TCE and PCE, estimate their reaction rate constants, and determine the reaction products. Additionally, gas chromatography–mass spectroscopy (GC–MS) and Fourier transform infrared (FTIR) spectroscopy were used in combination to determine the UV photooxidation products. Furthermore, the GC–MS technique was used to evaluate the destruction and removal efficiencies of target contaminants after the UV photooxidation and the BTF stages. The products at each stage provided insight into reaction pathways for UV photooxidation, and ensured that treated effluent from the UV–BTF system was devoid of undesirable and toxic organics at trace levels. Microbial cell count measurements were conducted to evaluate the microbial cell viability in the BTF process, and to determine whether reduction in toxicity was achieved by UV oxidation pretreatment.

## 2. Background

### 2.1. Technologies for gas phase destruction of chlorinated organics

In recent years, advanced oxidation processes (AOPs) have attracted much attention because they are effective in the decomposition of organic contaminants in gas streams at convenient reaction rates and ambient temperatures (Shen and Ku, 2002). These processes include Fenton's process, photolysis by ultraviolet (UV) radiation, photolysis using photo-catalysts (including titanium dioxide and other semi-conductor oxides), UV photolysis with ozone, UV photolysis with hydrogen peroxide, electron beams, and non-thermal plasma and similar technologies producing hydroxyl and other free radicals (Haag et al., 1996; Shen and Ku, 2002). In most AOPs, chlorinated compounds are not completely transformed into carbon dioxide, water and hydrochloric acid, but are often converted to phosgene (carbonyl chloride,  $\text{COCl}_2$ ) and undesirable chlorinated organics depending upon process conditions. Haag et al. (1996) concluded that direct photolysis of several VOCs using pulsed xenon lamps could not generally compete with adsorption, catalytic oxidation or thermal oxidation owing to weak absorbances of most contaminants at the radiation wavelengths. Nevertheless, they experienced sufficiently high UV photo-oxidation rates for chlorinated aliphatics such as TCE and PCE. They attributed this to two factors, namely, high absorption of these compounds in the UV wavelength range, and fast free-radical chain reactions mediated by chlorine atoms. Haag et al. (1996) noted that several other researchers also confirmed high decomposition rates and quantum yields exceeding 1.0 for these compounds. Furthermore, Haag et al. (1996) observed that the advantage of using a photo-catalyst such as titanium dioxide for improving chemical kinetics was often offset by high catalyst costs. These factors provided the rationale for employing UV photooxidation with commercial viability for purifying gas streams contaminated with chloroethylenes at low concentrations.

The UV oxidation causes organic pollutants to enter excited states, and subsequently undergo series of free-radical chain reactions, whereby they are decomposed. Additionally, an oxidant such as ozone and hydrogen peroxide, or a catalyst such as titanium dioxide may be added to UV oxidation reactors to increase the oxidation efficiency. The general understanding is that under UV oxidation a molecule undergoes metastasis into the excited state after absorbing photons, per the Gottus–Draper and Einstein–Stark theories. During these excitation processes, electrons transit from orbitals of lower energies to those of higher energies. If the electrons in the bonding orbitals absorbed sufficient energy from the photons to move to anti-bonding orbitals, several chemical bonds could be broken, and fast free-radical reactions could occur, resulting in the decomposition of target contaminants.

A growing concern regarding AOPs in general and UV photooxidation in particular for the gas phase destruction of chloroethylenes exemplified by TCE and PCE is the generation of undesirable products such as phosgene and chlorinated

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