Science of the Total Environment 631–632 (2018) 298–307

Contents lists available at ScienceDirect

Science of the Total Environment

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

Assessment of the disinfection capacity and eco-toxicological impact of atmospheric cold plasma for treatment of food industry effluents

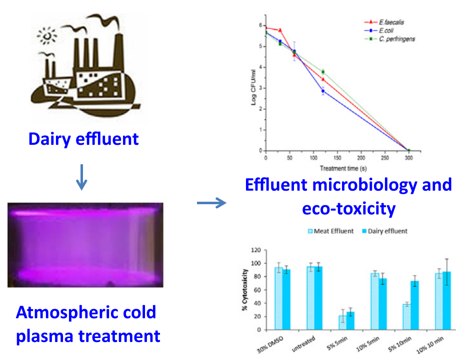
Apurva Patange [a], Daniela Boehm [a], Michelle Giltrap [a,b], Peng Lu [a], P.J. Cullen [c], Paula Bourke [a,*]

[a] School of Food Science and Environmental Health, Dublin Institute of Technology, UK
[b] Nanolab, FOCAS Research Institute, Dublin Institute of Technology, Kevin Street, Dublin 8, UK
[c] Department of Chemical and Environmental Engineering, University of Nottingham, UK

HIGHLIGHTS

- ACP is an effective alternative solution for wastewater treatment.
- Effective to inactivate key mono/mixed indicator bacteria from model effluents
- ACP shows useful efficacy within short periods of both treatment and retention times.
- System and treatment parameters affect the bacterial inactivation efficiency.
- Treated samples displayed limited effect on test species up to 24 h exposure, a prolonged contact of up to 48 h was toxic.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 6 November 2017
Received in revised form 20 February 2018
Accepted 22 February 2018
Available online xxxx

Editor: Daniel Wunderlin

Keywords:
Dairy and meat effluent
Wastewater treatment
Atmospheric cold plasma
Bacterial inactivation
Aquatic toxicity

ABSTRACT

Generation of wastewater is one of the main environmental sustainability issues across food sector industries. The constituents of food process effluents are often complex and require high energy and processing for regulatory compliance. Wastewater streams are the subject of microbiological and chemical criteria, and can have a significant eco-toxicological impact on the aquatic life. Thus, innovative treatment approaches are required to mitigate environmental impact in an energy efficient manner. Here, dielectric barrier discharge atmospheric cold plasma (ACP) was evaluated for control of key microbial indicators encountered in food industry effluent. This study also investigated the eco-toxicological impact of cold plasma treatment of the effluents using a range of aquatic bioassays. Continuous ACP treatment was applied to synthetic dairy and meat effluents. Microbial inactivation showed treatment time dependence with significant reduction in microbial populations within 120 s, and to undetectable levels after 300 s. Post treatment retention time emerged as critical control parameter which promoted ACP bacterial inactivation efficiency. Moreover, ACP treatment for 20 min achieved significant reduction ($\geq$2 Log_{10}) in *Bacillus megaterium* endospores in wastewater effluent. Acute aquatic toxicity was assessed using two fish cell lines (PLHC-1 and RTG-2) and a crustacean model (*Daphnia magna*). Untreated effluents were toxic to the aquatic models, however, plasma treatment limited the toxic effects. Differing sensitivities were observed to ACP treated effluents across the different test bio-assays in the following order: PLHC-1 > RTG-2 ≥ *D. magna*; with greater sensitivity retained to plasma treated meat effluent than dairy effluent. The toxic effects were dependent on concentration and treatment time of the ACP treated effluent; with 30% cytotoxicity in *D. magna* and fish cells observed after 24 h of exposure to ACP treated effluent for concentrations up to 5%. The findings suggest the need to employ wider variety of aquatic organisms for better understanding and complete toxicity evaluation of long-term effects.

* Corresponding author at: Dublin Institute of Technology, 31 Marlborough Street, Dublin 1, UK.
E-mail address: paula.bourke@dit.ie (P. Bourke).

https://doi.org/10.1016/j.scitotenv.2018.02.269

The study demonstrates the potential to tailor ACP system parameters to control pertinent microbial targets (mono/poly-microbial, vegetative or spore form) found in complex and nutritious wastewater effluents whilst maintaining a safe eco-toxicity profile for aquatic species.

1. Introduction

Food processing industries are one of the largest sources of wastewaters, with a trend of increasing volumes being produced. Water plays a vital role in the food industry with large volumes of wastewater derived from various processing units including washing, cleaning, sanitization and various steps in the manufacturing of food products themselves. The physical and chemical properties of the effluents derived from the food sector vary in line with the product type and quantity. Therefore, the wastewater streams can hold a multiplicity of microbiological and chemical contaminants within an environment characterized by high amounts of organic content, nutrients like proteins, carbohydrates, fats, minerals and higher concentrations of suspended solids; biological oxygen demand (BOD) and chemical oxygen demand (COD), with large variation in pH (Gough et al., 2000; Perle et al., 1995).

Treatment methods currently applied to wastewaters include electrochemical treatment, anaerobic processing, ultrafiltration, chlorination, heat treatment, radiation treatment and different combinations of these (Demirel et al., 2005; Yavuz et al., 2011). The increasing costs involved for treatment methods, residual chemical by products, high energy consumption along with the stringent standards for wastewater treatment, comprise some of the major limitations of these technologies. Public health authorities and environment management systems explore innovative methods to mitigate the environmental impact of the wastewaters discharged from these organic intensive industries. Atmospheric cold plasma is under intensive investigation as a novel decontamination technology. Plasma comprises of charged particles (positive and negative electrons), free radicals, UV photons and wide range of reactive species such as hydrogen peroxides, nitric oxide derived species. Non-thermal plasma or cold plasma generated from atmospheric or near atmospheric pressure at room temperature is called atmospheric cold plasma. Cold plasma is characterized by non-equilibrium, where cooling of the ions and the uncharged molecules is significantly more effective than that of energy transfer from electrons resulting in the gas remaining at a low temperature (Bourke et al., 2017). The introduction of an electric discharge into the liquid surface causes a number of chemical reactions (Fridman, 2008; Locke et al., 2012). The oxidative degradation of the organic matter and biological content is effected through the ensuing reactive species and the effects of the secondary reactive species are longer lived and mediated through liquids. The basis of the treatment is transfer of the charged species at the gas-liquid interface (Pivovarov and Tischenko, 2005). Non-thermal plasma technology has been studied to degrade phenol pollutant dyes (Sarangapani et al., 2017a) and for reducing COD and TOC (Reddy et al., 2013; Tomizawa and Tezuka, 2007) in effluents. Also, ACP has proven effective for the wide range of pathogenic and spoilage microorganisms predominantly found in food and biomedical sectors. Studies from Han et al. (2014) demonstrated effective bacterial inactivation using in-package DBD-ACP system within short period time and post treatment storage time of 24 h. Also work by Pavlovich et al. (2013) demonstrated effective decontamination of bacteria with 120 s of treatment present in phosphate buffer using indirect dielectric barrier discharge treatment ACP system. Although ACP studies have been focused on aqueous treatment, there are limited studies pertaining to its application in microbial contamination in complex media compositions like in wastewater effluents with much shorter contact time.

Several studies report significant microbial concentrations and the presence of pathogens in the food industrial environment (McGarvey et al., 2004; Parkar et al., 2015). A wide range of microbial profiles have been reported in food effluents including *Cryptosporidium parvum, Giardia, Escherichia coli, Clostridium perfringens, Enterococcus faecalis*, *Salmonella* etc. (Chapman, 2000; Dungan and Leytem, 2013; Ibekwe et al., 2003). Since food industrial wastes contain high levels of organic content, they act as an ideal medium for the growth and survival of both pathogenic and spoilage bacteria within the industrial environment, causing serious challenges for the efficient removal of organics and the safety profile of the wastewater effluent. The effluents released from the food industries are characterized by high biological oxygen demand (BOD) and chemical oxygen demand (COD). The bacteria decompose the organic materials present in the wastewater, depleting the oxygen levels in rivers proving lethal for most aquatic life (Enderlein et al., 1997).

This study focussed on two sectors and employed meat and dairy model effluents to address several outstanding research objectives. These were to ascertain the impact of ACP on bacterial communities prevalent in wastewaters and to devise parameters for their efficient control using custom built DBD-ACP system. Target bacteria were selected based on common indicator organisms encountered in the food industries and as recommended by EPA (USEPA, 2010). Considering the most predominant and resistant of pathogens, the principle indicator microorganisms *Escherichia coli, Clostridium perfringens Enterococcus faecalis* and *Bacillus megaterium* endospores were selected. A better understanding of microbial inactivation mechanism by ACP and the behaviour of multi-species communities will promote the definition of treatments that either alone or in combination with other control agents will assure the effective decontamination of food derived wastewater streams.

Additionally, considering that the longer term effects of ACP on biological systems are mediated through liquid, it was investigated if this technology can be safely deployed and what the impact of ACP treated effluent on eco-toxicity markers may be. The aquatic markers selected for eco-toxicity testing were fish cell lines (PLHC-1 and RTG-2 cells) and crustaceans (*Daphnia magna*) representative of a range of trophic levels. These are simple test organisms which are known to be sensitive to a wide range of pollutants and have a standard reproducible response to facilitate comparison.

2. Material and methods

2.1. Preparation of model effluent

Model dairy effluent (DE) of pH 6.0 was prepared as reported by Daverey and Pakshirajan (2011) which contained 2 g/l semi-skimmed milk powder (TESCO, Ireland), 0.2% (w/v) milk fat (East end Foods, UK), 0.01% (w/v) sodium hydroxide (Sigma-Aldrich, Ireland) and sterile distilled water. The milk powder was composed of 35% proteins, 50.3% carbohydrates, 0.6% fat, 1.05% calcium, 0.8% phosphorus, 0.095% magnesium. The organic content in dairy effluent varies with the manufactured product; therefore, the relative impact of organic milk fat content on bacterial inactivation efficiency was examined. The milk fat content as an experimental variable ranged between 0.2 g/l to 4 g/l.

Model meat effluent was prepared using procedure of Barrera et al., with few modifications. The synthetic meat effluent consisted of commercial meat extract powder (Scharlau Chemie, Barcelona, Spain) 1950 mg/l, glycerol ($C_3H_8O_3$) (Sigma-Aldrich, Ireland) 200 mg/l, ammonium chloride (NH_4Cl) (Sigma-Aldrich, Ireland) 360 mg/l, sodium