



Potential effects of rainwater-borne hydrogen peroxide on pollutants in stagnant water environments



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HIGHLIGHTS

- Stagnant water environments are subject to chemical and microbial contamination.
- Microcosm experiments using canal and urban lake water as example stagnant water.
- Synthetic rainwater-borne H₂O₂ reacted with water-borne Fe²⁺ to trigger Fenton reaction.
- Coliforms, ammonia-oxidizing microbes and organic pollutant-degraders were inactivated.
- Nitrite and PAHs can be chemically decomposed by •OH generated from the reaction system.

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ABSTRACT

Microcosm experiments were conducted to examine the effects of rainwater-borne H₂O₂ on inactivation of water-borne coliforms, oxidation of ammonia and nitrite, and degradation of organic pollutants in canal and urban lake water. The results show that the soluble iron in the investigated water samples was sufficiently effective for reaction with H₂O₂ in the simulated rainwater-affected stagnant water to produce •OH (Fenton reaction), which inactivated coliform bacteria even at a H₂O₂ dose as low as 5 μM within just 1 min of contact time. Coliform inhibition could last for at least 1 h and repeated input of H₂O₂ at a 30 min interval allowed maintenance of microbial inhibition for at least 3 h. Nitrification was also impeded by the Fenton process. The resulting inhibition of ammonia-oxidizing microbes reduced the removal rate of NH₄⁺ and the emission of gaseous N species. In the presence of H₂O₂ at a dose of 20 μM, Fenton-driven chemical oxidation appeared to outplay the impediment of biodegradation caused by inhibited microbial activities in terms of removing total polycyclic aromatic hydrocarbons from the water column. The findings point to a potential research direction that may help to explain the dynamics of water-borne pollutants in ambient water environments.

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1. Introduction

Stagnant water environments such as lakes, ponds, canals act as sinks for organic matter and various inorganic, organic and microbial contaminants (Pal et al., 2010; DeLorenzo et al., 2012; Sales-Ortells et al., 2015). The organic-rich status could enhance the growth of microbial pathogens (Sato et al., 2013, 2016). This poses a health risk to people who are exposed to water-borne microbial pathogens e.g. during open water swimming (Wade et al., 2003;

Schets et al., 2011; McBride et al., 2013). Ammonia (NH₃) is a gaseous product of organic matter mineralization. In non-alkaline water environments, ammonia tends to be converted into ammonium ion (NH₄⁺) via a reversible reaction with water molecule (Karthikeyan et al., 2008). Therefore, ammonium acts as an immediate source of ammonia in the water environments. Stagnant water environments could receive ammonium from agricultural, urban and industrial discharges (Erisman et al., 2007; Reche et al., 2012; Wang et al., 2016). Ammonia can also be produced in-situ through in-water protein metabolism by aquatic biota (Walsh and Wright, 1995). Therefore, ammonia is commonly present in stagnant water environments. Due to its high toxicity to aquatic life, ammonia is viewed as one of the major inorganic pollutants in

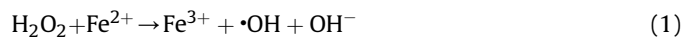
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aquatic ecosystems (Parker et al., 2012; Collos and Harrison, 2014). Potentially toxic organic pollutants such as polycyclic aromatic hydrocarbons (PAHs) and various pesticides from urban or agricultural runoffs can also enter into and accumulate in stagnant water environments (Kong et al., 2005; Kanzari et al., 2014; Hijosa-Valsero et al., 2016).

Ammonia/ammonium can be transformed into nitrite and eventually nitrate through microbially mediated nitrification (Bernhard et al., 2007; Stahl and de la Torre, 2012). It is believed that in natural environments, organic pollutants are mainly degraded through microbial metabolism (Glick, 2003; Liste and Prutz, 2006; Khan et al., 2013). The activity of water-borne microbes in a given water environment can be affected by many factors (Morens et al., 2004; Braks and de Roda Husman, 2013; Nguyen et al., 2016). While the effects of some environmental parameters such as pH, temperature, salinity etc. on activities of water-borne microbes have been well recognized (Wadowsky et al., 1985; Manas et al., 2003; Spinks et al., 2006), others are literally overlooked.

Hydrogen peroxide (H₂O₂) is commonly present in rainwater with a molar concentration frequently ranging from 5 to 70 μM (Willey et al., 1996; Kieber et al., 2001; Gonçalves et al., 2010). H₂O₂ is a strong oxidant, which can cause oxidative stress in cells (Schoonen et al., 2010; Matthijs et al., 2012; Ma and Lin, 2013). Due to relatively anoxic and organic-rich conditions, iron compounds in the benthic sediments under stagnant water layers are subject to reductive dissolution (España et al., 2007; Anna and Jacek, 2014; Shamsul Haque et al., 2016). This could lead to release of ferrous iron (Fe²⁺) into water column (Díez et al., 2007; Sanders et al., 2012; Willey et al., 2015). During rainfall events, the Fe²⁺ in stagnant water can react with rainwater-borne H₂O₂ to trigger Fenton reaction, as expressed in the following chemical equation:



Fenton reaction produces hydroxyl radical ($\cdot\text{OH}$), which is an even stronger oxidant, as compared to H₂O₂. Hydroxyl radical has the capacity to destroy cells and organic molecules within a short period of time (Winterbourn, 1995; Lee and Gunten, 2010). It is therefore likely that rainwater-borne H₂O₂ has a role to play in affecting the activity of microbial communities in stagnant water environments. Qin et al. (2013) showed that Fenton reaction triggered by H₂O₂ and Fe²⁺ (Fenton reagent) at a concentration range encountered in rainwater-affected surface water was capable of chemically degrading some herbicides (diuron, butachlor and glyphosate) in synthetic solutions. But there have not been further tests on other organic pollutants. Brito et al. (2010) demonstrated that hydroxyl radical generated from Fenton reaction could chemically oxidize ammonia in landfill leachate. However, the dose of H₂O₂ they used was much higher than what can be encountered in rainwater. It is likely that in natural water, Fenton reaction has a dual role to play in terms of oxidizing ammonia and degrading organic pollutants; the chemical reaction of hydroxyl radical with ammonia and organic pollutants may accelerate the oxidation of ammonia and decomposition of organic pollutants while the oxidative stress in ammonia-oxidizing and organic pollutant-degrading microbes caused by $\cdot\text{OH}$ is likely to impede ammonia oxidation and biodegradation of the organic pollutants. Consequently, the net oxidation rate of ammonia and degradation rate of organic pollutants by Fenton reagent is determined by the combined effects of these two counteractive processes.

Understanding the behaviour and fate of water-borne pollutants is important for evaluating aquatic ecosystem health and the risk to human exposure, which can inform development of appropriate

management strategies. To understand the potential role of rainwater-borne H₂O₂ in affecting the behaviour and fate of some key pollutants in stagnant water environments, a multi-stage research project has been set. This work reports on the results from the stage of microcosm experiments with a focus on coliform bacteria, ammonia and PAHs. The aim of conducting these microcosm experiments was to establish the dose-response relationships between the added H₂O₂ and the selected water-borne pollutants under controlled environmental conditions in order to inform the design of field-based investigations in the follow-up phase of the project. The specific objectives for this part of the project were to examine the effects of H₂O₂ at a concentration range that is likely to be encountered in rainwater on (a) inactivation of potential microbial pathogens, as indicated by coliform bacteria, (b) oxidation of ammonia and nitrite, and (c) degradation of PAHs in synthetic solutions and/or selected natural stagnant water samples.

2. Materials and methods

2.1. Natural water samples used in the experiment

Canals and urban lakes were used as example stagnant water environments in this study. The water samples were collected from the selected locations in the Bridgewater canal and Salford Quays in Manchester City (Latitude: 53°28'56.6"N; Longitude: 2°14'21.8"W) and the Clarence Dock in Leeds City (Latitude: 53°48'05.8"N; Longitude: 1°32'59.8"W). Both cities are located in England, The United Kingdom. The Bridgewater canal is arguably the first canal in England, which was built in mid-1700 to transport coal from the mine sites to the industrial areas in Manchester. Salford Quays is an urban lake area regenerated from the former ship docks, which form part of the Manchester Ship Canal that connected Manchester with Port of Liverpool. Part of the Salford Quays is used for regular water sport activities, including swimming. The open water in Leeds Clarence Dock is an urban lake surrounded by shops, office buildings and residential apartments with heavy population. These water areas were heavily contaminated during the period of Industrial Revolution (Douglas et al., 2002; Williams et al., 2010). The benthic sediments contained substantial amounts of organic and inorganic pollutants. Although pollutant loads from the upper catchment areas have been significantly reduced for the past decades as a result of de-industrialization in the region, water quality remains poor in the urban lakes and canal sections adjacent to urban areas due largely to pollutant inputs from urban runoff during rainfall events and re-entry of bed sediment-borne pollutants into water column (Rees et al., 1999; Quesada et al., 2014). Water quality monitoring data (our unpublished data) indicated that the canal water frequently contained elevated levels of coliform bacteria, nitrogen and certain potentially toxic organic pollutants.

Due to the practical difficulties in satisfactorily quantifying all the intended physical, chemical and microbial parameters in a single experiment, three sets of separate experiments were conducted to individually examine the effects of H₂O₂ in synthetic rainwater-affected stagnant water on the inhibition of coliform bacteria, oxidation of ammonia and nitrite, and degradation of organic pollutants. Some major physical and chemical characteristics of the five water samples used in the experiments are given in Table 1. The water samples WS-EXP1A, WS-EXP1B and WS-EXP1C used for Experiment 1A-1C (coliform experiments) were collected from the Bridgewater canal, Salford Quays and Leeds Clarence Dock, respectively. The sample WS-EXP2C used for Experiment 2C (ammonia experiment) was taken from Salford Quays. The sample WS-EXP3 used for Experiment 3 (organic pollutants) was collected from Bridgewater canal.

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