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The fate of H_2O_2 during managed aquifer recharge: A residual from advanced oxidation processes for drinking water production

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HIGHLIGHTS

- Different decomposition factors of H_2O_2 as a residual of AOP in MAR systems were studied.
- Inorganic substances in sand and microbial biomass are main factors to H_2O_2 decomposition.
- Low concentration H_2O_2 may decompose in the first several centimeters of MAR systems.

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ABSTRACT

The fate of H_2O_2 residual from advanced oxidation process (AOP) preceding managed aquifer recharge (MAR) is of concern because H_2O_2 could lead to undesired effects on organisms in the MAR aquatic and soil ecosystem. The objective of this study was to distinguish between factors affecting H_2O_2 decomposition in MAR systems, simulated in batch reactors with synthetic MAR water and slow sand filter sand. The results showed that pure sand and soil organic matter had no considerable effect on H_2O_2 decomposition, whereas naturally occurring inorganic substances on the surface of sand grains and microbial biomass are the two main factors accelerating H_2O_2 decomposition in MAR systems. Additionally, the results showed that the H_2O_2 decompositions with different initial concentrations fitted first-order kinetics in 2–6 h in a mixture of slow sand filter sand (as a substitute for sand from a MAR system) and synthetic MAR water with high bacterial population. An estimation indicated that low concentrations of H_2O_2 (<3 mg/L) could decompose to the provisional standard of 0.25 mg/L in the first centimeters of MAR systems with the influent water containing high microbial biomass 38 ng ATP/mL.

1. Introduction

Managed aquifer recharge (MAR), such as river bank filtration, dune infiltration and artificial recharge, is a natural water treatment process that induces surface water to flow through soil/sediment and into a vertical or horizontal well (Maeng et al., 2011; Tufenkji et al., 2002). This treatment process is robust and cost-effective and is frequently applied in Europe (Van der Hoek et al., 2014). For example, in the Netherlands and Germany, water utilities using MAR as a water treatment process supply drinking water without chlorination as disinfection process (Lekkerkerker, 2012; Maeng, 2010). Previous research demonstrated that the combination of advanced oxidation process (AOP) and subsequent MAR is a potential treatment system to remove various organic micropollutants (OMPs) during drinking water production (Lekkerkerker-Teunissen et al., 2012; Lekkerkerker et al., 2009; Oller et al., 2011). A disadvantage of applying AOP with O_3 is the formation of bromate during oxidation of bromide containing waters. In order to reduce the formation of bromate which has been designated as carcinogenic to humans (Kurokawa et al., 1990), H_2O_2 should be dosed excessively (Knol, 2012; Von Gunten and Oliveras, 1998; Wert et al., 2007). Consequently, the MAR infiltration water may contain residual concentrations of H_2O_2.

A number of studies about H_2O_2 decomposition in aquatic ecosystems and soil ecosystems have focused on biotic factors, such as bacteria (Richard et al., 2007; Zappi et al., 2000) and other microorganisms (Cooper and Lean, 1989; Richard et al., 2007) and

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abiotic factors, such as iron (Moffett and Zafiriou, 1993; Wilson et al., 2000), manganese (Do et al., 2009; Häkkinen et al., 2004; Russo et al., 2013), transition metals (Lousada and Jonsson, 2010; Moreno et al., 2011), lanthanide oxides (Lousada et al., 2013) and iodide (Wong and Zhang, 2008). H_2O_2 decomposition in water also has been reported (Cooper and Lean, 1989; Moffett and Zafiriou, 1993; Richard et al., 2007; Wilson et al., 2000). The results of Schumb (1949) showed that manganese and iron were extremely reactive with concentrated H_2O_2 solutions. Also, H_2O_2 decomposition studies have been conducted in metal- or DOC-rich waters (Chiriţă, 2009; Wilson et al., 2000). Previous research found that a large fraction of H_2O_2 loss in both a fresh water system and soil was attributable to biotic mechanisms. Richard et al. (2007) found that biologically based reactions (i.e., catalase) were the primary mechanism for H_2O_2 decomposition in a shallow fresh water system in New Zealand. It was observed from the literature of Zappi et al. (2000) that the first-order rate constant of biotic reactions was always much higher than that of abiotic reactions for H_2O_2 decomposition in various soils with different calcium, iron, manganese, TOC and phosphorus contents. It is clear that the fate of H_2O_2 in aquatic systems has been investigated comprehensively, and a few studies focused on the reactions of H_2O_2 with natural-occurring constituents in soil (Bissey et al., 2006; Miller and Valentine, 1999). These publications investigated the stability of H_2O_2 as the oxygen source for bioremediation activities in soil, because of several potential interactions of H_2O_2 with various soil constituents and its potentially fast decomposition. Studies of Morgan and Watkinson (1992) and Schumb (1949) reported reaction of H_2O_2 with naturally occurring stabilizers, such as tripolyphosphate, MnO_4^- and Cu^{2+} within soils. Bissey et al. (2006) investigated the interactions between catalyzed H_2O_2 propagations and soil organic matter (SOM) within surface soil and reported that the H_2O_2 decomposition rate decreased with the increase of SOM at neutral pH. Miller and Valentine (1999) examined mechanisms and kinetics of abiotic H_2O_2 decomposition in the presence of sand collected from an aquifer and a riverbed. However, more understanding is needed to determine the fate of H_2O_2 in MAR systems specifically. High concentrations of H_2O_2 can cause damage to cell membranes and have deleterious effects on biological systems (Ananthaswamy and Eisenstark, 1976; Collén and Pedersén, 1996; Wong et al., 2003). Schmidt et al. (2006) concluded that H_2O_2 minimum inhibitory concentration (MIC) to the most sensitive bacteria species *Psedomonas aeruginosa* was 5.1 mg/L. The study of Urfer (1998) demonstrated that the continuous presence of around 1 mg/L H_2O_2 did not lead to a major inhibition of the biological removal of acetate and formate in a lab-scale sand drinking water biofilter. Knol (2012) stated that even very low concentrations of H_2O_2 could lead to undesired destruction of organisms in MAR infiltration ponds and he mentioned a provisional standard of 0.25 mg/L H_2O_2 for MAR infiltration water. Consequently, an improved understanding of the fate of H_2O_2 in MAR systems would be essential to see whether this provisional standard or lower concentrations can be reached.

The objective of this study was to distinguish between different factors affecting H_2O_2 decomposition in MAR systems. The general approach in this study was to divide the aquifer environment into two separate physical compartments (water and sand) that contain naturally existing biological and chemical species that might react with H_2O_2. Batch reactor experiments were conducted to determine the reactions of H_2O_2 with biotic (microbial community in water) and abiotic constituents (pure sand particles, inorganic ions in infiltration water, SOM in MAR sand and naturally occurring inorganic substances coating on sand).

2. Materials and methods

2.1. Materials

The top 0.5–2.0 cm (schmutzdecke) of a slow sand filter (SSF) has diverse microbial communities and greatly contributes to the removal of organic matter by biodegradation processes, so this layer is considered to represent aerobic microbial activity of sand filtration systems (Chekol, 2009; Dizer et al., 2004). The SSF sand in the facilities of drinking water utility Dunea (The Hague, the Netherlands) originated from the dune infiltration area. Consequently, schmutzdecke sand (top of SSF) with natural microbial communities was used in batch reactors as a substitute for the sand in the dune infiltration ponds. As a reference, pure sand (silicon dioxide without any impurities; 1.07711.1000, VWR company) was used. The water for batch reactors was prepared with demineralized water (demi-water) and additive chemicals (33 mg Na_2HPO_4/L, 7.5 mg NaH_2PO_4/L, 22 mg K_2HPO_4/L, 140 mg $CaCl_2$/L, 0.031 mg $FeCl_3$/L, 0.032 mg NH_4Cl/L, 40.75 mg $MgSO_4$/L, 17.823 mg $NaNO_3$/L, 0.00114 mg $MnCl_2$/L, 82 mg CH_3COONa/L) to simulate the water quality at the MAR site of Dunea. The characteristics are presented in Table 1. Based on preliminary experiments, it was found that CH_3COONa (Merck, Germany) was rapidly consumed as the source of DOC in the batch reactors, so 24 mg/L DOC was added in order to have residual DOC in the reactors and avoid bacterial starving conditions. Dosing carbon source to levels exceeding natural MAR systems may lead to higher microbial biomass concentration in batch reactors than in natural MAR systems (Pharand et al., 2014) and enhance the endurance ability to decompose H_2O_2. Therefore, a short inventory was performed based on observed adenosine triphosphate (ATP) concentrations in different waters to estimate the effect of carbon dosage on H_2O_2 decomposition (§ 3.4). The H_2O_2 solutions were prepared from a 30% standard solution (Merck, Germany). All the solutions used in this study were prepared using water from a Millipore Milli-Q system. All chemicals were of AR grade.

2.2. Batch experimental setup

Batch experiments were performed with 39 glass batch reactors with a volume of 1 L for around 3 months. Batch reactors were filled with 100 g SSF sand and 500 mL synthetic MAR water to simulate MAR systems (Lekkerkerker, 2012; Maeng, 2010). In addition, reference batch reactors were prepared with 100 g pure sand silicon dioxide and 500 mL synthetic MAR water. All batch reactors were placed in a dark room, either temperature controlled (12 ± 0.5 °C) or ambient temperature (23–27 °C), depending on the experiment. Batch reactors were uncovered so that air could enter batch reactors to maintain oxic conditions. To avoid anaerobic conditions, the batch reactors were slightly shaken daily without disturbing the biofilm that had developed on the sand.

2.3. Experiments

To divide the aquifer environment into two separate physical compartments (water and sand) that contain naturally existing biological and chemical species that might react with H_2O_2, this study used an experimental set-up as shown in Fig. 1, providing an overview of batch reactors' conditions used in the experiments. All batch reactors were prepared and sampled in triplicate. The performed experiments were divided into:

a) Abiotic: H_2O_2 decomposition under autoclaved conditions (with/without sand)