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Research article

Formation and transformation of chloroform during managed aquifer recharge (MAR)

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ABSTRACT

Chlorination is an effective method to protect the safety of groundwater systems during managed aquifer recharge. However, chlorination leads to the formation of disinfection by-products, whose behavior in aquifers remains unclear and has caused public concern. In this study, an in-site test was performed on an anoxic aquifer in Shouguang City, China, to investigate the formation and transformation of chloroform during managed aquifer recharge. The field tests showed that the formation of chloroform in groundwater caused by the recharge of chlorinated water, and that the fate of chloroform was affected by adsorption and biodegradation. The retardation factor was 1.27, and the half-life was 29 days. The formation and transformation of chloroform during continuous recharge under different hydrochemical conditions was further investigated by batch experiments. These experiments showed that the formation of chloroform increased with contact time, tended to be stable after 10 h, and was facilitated by high chloride/TOC ratios, high pH, and low ionic strength (IS) for a given contact time. The adsorption experiments showed that the process accords with the pseudo-second-order kinetic equations and the Freundlich model. The adsorption capacity was pH dependent (1.01–1.66 μg/g at pH 5 and 2.17–3.05 μg/g at pH 9). Increasing the IS promotes adsorption. The results from biodegradation experiments indicated that the biodegradation was well fitted by the Monod equation. The retardation factor in the batch experiments was close to that of the field test, but the half-life was less than the field test. This is mainly due to the difference in the concentration of dissolved oxygen.

1. Introduction

Managed aquifer recharge projects can increase the yields from groundwater resources and prevent the occurrence of environmental geological problems (Huang et al., 2012; Zhang et al., 2015). Stormwater (Wang, 2009), surface water (Fram, 2003), reclaimed wastewater (Pavelic et al., 2005) and drinking water (Wu et al., 2016a,b) can be used as recharge water. The treatment of water especially if the source was polluted with excessive nutrient, toxic chemical and harmful microorganisms (e.g. agricultural wastewater) is necessary to impede further environmental impacts (Hanifzadeh et al., 2017). To protect groundwater quality and to protect public health from risks associated with biofouling and potential microbial pathogens, the recharge water should be disinfected before recharging to the aquifer system. The difference in chemical composition between recharge water and groundwater as well as the process of recharge lead to disturbances in the hydrodynamic and hydrochemical fields and may pose a serious problem to the environment (Wood and Bassett, 1975). However, it is unclear how the managed recharge of chlorinated water negatively affects groundwater quality (Campillo et al., 2004).

Beginning with the pioneering work of Rook in 1974, it has been established that disinfectants in drinking water can react with dissolved organic matter (DOM) and generate disinfection by-products (DBPs), which are potentially harmful to human health and can be carcinogenic (Rook, 1973; Gopal et al., 2007; Hong et al., 2007). The commonly found DBPs include trihalomethanes (THMs),

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haloacetic acids, and haloacetonitrile (Richardson et al., 2007). Because of their toxicity, the maximum concentration level for total THM has been set at 80 μg/L in the United States, and 10 μg/L in Germany (Nicholson and Ying, 2005). The maximum concentration level of chloroform in drinking water as regulated by China is currently 60 μg/L. The artificial recharge process creates a secondary mechanism whereby DBPs can be generated and poses a significant threat to groundwater quality. However, the effects of managed aquifer recharge on DBPs have only been assessed in a few research studies (Thomas et al., 2010).

The formation and transformation of DBPs when chlorinated water is used for managed recharge has been studied in a few field and laboratory scale investigations. Mille et al. (1993) reported that concentration of DBPs in groundwater during initial recharge is significantly higher than that in recharge water in Las Vegas, and provided the seminal theory for the secondary formation of DBPs during managed aquifer recharge. Later studies by Singer et al. (1993) and Pavelic et al. (2005) corroborated this theory. Some studies (Reckhow et al., 1990; Yang, 2008) indicated that the formation of DBPs was related to the characteristics of the humic materials and the hydrochemical conditions. The adsorption of THMs to aquifer media which is related to the organic material of aquifers has been studied (And and Dural, 1998; Izbicki et al., 2010). Furthermore, some studies indicated that chloroform could undergo biodegradation under anaerobic conditions (Landmeyer et al., 2000; Pavelic et al., 2005; Izbicki et al., 2010). Other studies of the biodegradation of chloroform during managed aquifer recharge found that biodegradation under aerobic conditions is possible for specific THMs with suitable aerobic bacteria and substrate concentration conditions (Dillon et al., 2003; Thomas et al., 2010). Although some researches have described the environmental behavior of DBPs in simulated distribution systems (Sadiq and Rodriguez, 2004), systematic studies of this behavior during managed aquifer recharge are rare.

Chloroform is one of the common DBPs produced during managed aquifer recharge. The formation of chloroform is not instantaneous (Clark and Sivaganesan, 1998), and therefore knowledge of the formation and transformation behavior of chloroform during artificial managed recharge is needed to allay health concerns when the recharged water is used for human consumption. If chlorinated water was used for managed aquifer recharge, the concentration of chloroform will change during the recharge process. Therefore, we obtained field monitoring data and constructed laboratory experiments to evaluate the formation and transformation behavior of chloroform when chlorinated water was injected during managed aquifer recharge. The results from this study can provide a basis for the determination of the acceptable disinfectant concentrations in recharge water.

2. Materials and methods

2.1. Field site

Significant ground subsidence and saltwater intrusion has occurred in Shouguang City, Shandong Province, China. An artificial recharge test was conducted on the bank of an ancient river (Fig. S1). The recharge site has an area of about 400 m^2, and is a Quaternary alluvial aquifer. The target aquifer for managed recharge is a confined aquifer of about 8–10 m thick, which is primarily composed of fine sand and medium sand. The hydraulic conductivity is about 18.76 m/d and the hydraulic connections with the overlying and underlying aquifer are weak. The groundwater flow is from south-west to north-east, with a hydraulic gradient of 0.2‰ (Fig. S1).

All soil samples used in the laboratory experiments were collected by drilling at the recharge site. The samples were air-dried, and were sieved to three different sizes (0.50–0.25, 0.25–0.075, and <0.075 mm average diameter). The properties of the soil measured in the laboratory are shown in Table 1.

The groundwater in the target aquifer was found to be principally of the Cl-Na·Ca or Cl·SO_4-Na·Ca hydrochemical types. According to the water quality test results, the ionic strength (IS) ranged from 0.017 to 0.032 mol/L.

2.2. Artificial recharge field tests

The Mi River was selected as the recharge water source, and the water was chlorinated before injection because of the poor quality of the river water. The formation of chloroform may result in the groundwater quality not meeting drinking water criteria. Four monitoring wells (J1–J4) were installed around the recharge well (J0) (Fig. 1). The recharge water was injected continuously at a rate of 3 m^3/h. The hydrochemical characteristics (i.e., pH, electrical conductivity (EC), dissolved oxygen (DO), and total dissolved solid (TDS)) were measured in the field at the time groundwater samples were collected. Water levels were also monitored. The wells were monitored for 2.25 days. The monitoring intervals were 20 min for the first 160 min, 30 min for 161–340 min, and 60 min for the remainder of the test. The sampling density was about 32 samples for each well. Chloroform was only detected in well J2, and therefore only data from that well was used to study the behavior of chloroform during the artificial recharge test.

2.3. Laboratory experiments

The recharging tests in the field studied the integrative effect of each process in the recharge stage for fixed site conditions. Therefore, formation and batch experiments were conducted to confirm the formation and transformation process under different hydrochemical conditions. Chlorination was performed by adding sodium hypochlorite. Humic acid was used to simulate the organic matter in groundwater and TOC was used as the concentration index. The pH was adjusted by $HCl_{(aq)}$, and $NaOH_{(aq)}$, $NaCl_{(aq)}$, and $CaCl_{2(aq)}$ were used for IS adjustment. The excess chlorine present after experiments was quenched with sodium thiosulfate. All the reagents were analytical grade.

2.3.1. Laboratory experiments to study the formation of chloroform

Formation experiments were performed in 120 ml amber glass bottles, filled with a humic acid solution. The TOC = 5 or 10 mg/L, which is close to the TOC content of groundwater. Then the chlorine solution was added to the bottles at the required dosage, and a stock solution was injected to eliminate head space. The bottles were sealed and incubated at 15 °C to simulate subsurface conditions.

Managed aquifer recharge will cause the underground environment of the aquifer to change. To study the effect of different factors influencing chlorination, a single-variable experimental design was employed. The variation of the following variables from baseline conditions was investigated: contact time (0.25, 0.5, 0.75, 1, 2, 4, 6, 10, 24, 48, 60 h), ratio of chlorine and TOC (0.025, 0.05, 0.1, 0.2, 0.4, 0.6, 0.8, 1.2 1.6), TOC (5, 10 mg/L), pH (5, 6, 7, 8, 9), and IS (0, 0.02, 0.05, 0.1 M). The variables were considered not to be having an effect when the contact time was 24 h, the concentration of chlorine and TOC was 0.5 mg/L and 5 mg/L, respectively, pH was 7 and IS was 0 M when the indicators did not act as factors.

2.3.2. Laboratory batch experiments to study the transformation of chloroform

To improve the simulation of managed aquifer recharge with