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Variations in dissolved organic nitrogen concentration in biofilters with different media during drinking water treatment

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HIGHLIGHTS

- Dissolved organic nitrogen (DON) concentration varies in water biofiltering.
- Along the depth of biofilters, DON concentration first decreased and then increased.
- The greatest variation in DON occurred in the activated carbon biofilter.
- The physical properties of the biofilter media affected the microbial population.
- The heterotrophic bacterial proportion may be responsible for DON variation.

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ABSTRACT

Dissolved organic nitrogen (DON) is potential precursor of disinfection byproducts (DBPs), especially nitrogenous DBPs. In this study, we investigated the impact of biofilters on DON concentration changes in a drinking water plant. A small pilot plant was constructed next to a sedimentation tank in a drinking water plant and included activated carbon, quartz sand, anthracite, and ceramsite biofilters. As the biofilter layer depth increased, the DON concentration first decreased and then increased, and the variation in DON concentration differed among the biofilters. In the activated carbon biofilter, the DON concentration was reduced by the largest amount in the first part of the column and increased by the largest amount in the second part of the column. The biomass in the activated carbon filter was less than that in the quartz sand filter in the upper column. The heterotrophic bacterial proportion among bacterial flora in the activated carbon biofilter was the largest, which might be due to the significant reduction in DON in the first part of the column. Overall, the results indicate that the DON concentration in biofiltered water can be controlled via the selection of appropriate biofilter media. We propose that a two-layer biofilter with activated carbon in the upper layer and another media type in the lower layer could best reduce the DON concentration.

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1. Introduction

Dissolved organic nitrogen (DON) is composed of a series of compounds with functional nitrogen groups. In the drinking water chlorination process, organic matter reacts with chlorine to generate a variety of routine disinfection byproducts (DBPs), and DON in particular generates nitrogenous (N)-DBPs such as nitrosamines, haloacetonitrile (HAN), cyanogen halides, halonitromethanes (HNMs), and haloacetamides (HacAm) (Choi and Valentine, 2002; Plewa et al., 2004; Chen and Valentine, 2006; Krasner et al., 2006). Compared with non-nitrogenous DBPs, N-DBPs are more toxic and more dangerous with strong carcinogenic and mutagenic characteristics (Gopal et al., 2007; Lee et al., 2007; Muellner et al., 2007). N-Nitrosamines are known carcinogens, and their presence, particularly N-nitrosodimethylamine (NDMA), in water has been of great concern. The State of California Department of Public Health established a notification level of 10 ng L^{-1} for NDMA, N-nitrosodiethylamine (NDEA), and N-nitrosodipropylamine (NDPA) (CDPH, 2010). In addition, the World Health Organization proposed a level of 100 ng L^{-1} for NDMA as the guideline value for drinking water (WHO, 2008). Therefore, the removal/control of DON in the drinking water treatment process has great importance for reducing the generation of N-DBPs.

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It is well documented that the DON concentration is elevated while the dissolved organic carbon (DOC) concentration is reduced during filtering of source water through sand filters in the conventional water treatment process (Gu et al., 2010). Liu et al. found that along the biofilter depth, the DON concentration is reduced first (to the lowest level within 10–20 cm) and then increases gradually (Liu et al., 2012). Fan et al. found that the DON concentration increases with increasing depth in the two-stage O_3-BAC filter when nitrate is the sole inorganic nitrogen source. They also demonstrated that typical soluble microbial products (SMPs), such as tryptophan, tyrosine and protein, are generated, and the levels of SMPs are correlated with the DON concentration (Fan et al., 2012). During filter operation, a layer of biofilm is formed, and the filter becomes a biofilter. DON removal and generation occurs in the biofilter filtration process. Microorganisms decompose amino acids and other susceptible nitrogen-containing organic matter, while releasing SMPs. SMPs are considered an important component of newly generated DON, and this inevitably results in the more facile generation of DBPs, especially N-DBPs, in the chlorine disinfection process. Therefore, research on the impact of DON concentration changes in biofilters for the purpose of controlling N-DBP production is needed.

Microorganisms play an important role in DON concentration changes. Such changes are related to the microbial distribution within the filter, and microbial properties within the filter directly affect water quality and the nature of the media. Differences of the nature of the media, particle size, porosity, specific surface area, and other surface properties affect the capacity of a filter to adsorb and trap pollutants and lead to different microbial growth environments in the biofilter. Such properties can also cause biofilm organisms to behave differently, leading to differences in DON concentration within the biofilter. To compare these differences and explore potential underlying mechanisms, an experimental investigation was carried out using four commonly employed media in the present study.

2. Materials and methods

2.1. Experimental site and installations

The experiments were conducted at the Zhejiang Pinghu Drinking Water Plant. The source of raw water was the Haiyantang River in Pinghu City, which is contaminated by organic matter. The average concentrations of NH_4^+–N, NO_2^-–N, NO_3^-–N, and DON were 3.3 mg L^{-1}, 0.23 mg L^{-1}, 2.74 mg L^{-1}, and 1.54 mg L^{-1}, respectively. The source water was coagulated using poly aluminum chloride (PAC) before sedimentation. Sedimentation was achieved in horizontal sedimentation tanks with a treatment capacity of 25 000 $m^3\,d^{-1}$, an effective depth of 3 m, and a hydraulic retention time of 2.1 h. The small pilot plant was located next to the sedimentation tanks. The raw water was the effluent of the sedimentation tanks. Quartz sand, activated carbon, ceramsite, and anthracite were selected as the filter media.

Four parallel Plexiglas filtration columns with the same specifications of 10 cm in diameter and 100 cm in height of the filter layer were filled with different media and equipped with a flow meter to control the water flow. Sampling ports were arranged along the filter layers in order to determine water and biofilm characteristics at different heights within each biofilter. The biofilters were designed to down flow, i.e., the inlet was at the top and the outlet was at the bottom. When the pilot plant was running, as influent, sedimentation tank effluent was piped into the four biofilters by siphons. The liquid flow of 60 L h^{-1} was controlled by flow meters. The hydraulic retention time was set at 8 min to ensure that the filtration rate was 7–9 m h^{-1}. Each biofilter column was backwashed from the bottom up with the overflow pipe. The backwash intensity was 12–15 L s^{-1} m^2, and the backwash time was 5–7 min according to the water supply design specifications. A backwash cycle was performed once per day.

2.2. Raw water

The raw water quality remained stable within the parameters listed in Table 1. Samples were collected before the backwash at 9:00 every day.

2.3. Analytical methods

A Shimadzu TOC-VCHS analyzer was used to measure DOC. UVA_{254} was determined by a spectrometer. Ammonia was measured using the salicylate–hypochlorite method, nitrite using the N-(1-naphthyl)-ethylenediamine photometric method, nitrate using the UV spectrophotometry method, and total dissolved nitrogen (TDN) using the alkaline potassium persulfate digestion-UV spectrophotometric method. These determinations were carried out according to the Chinese National Standard Methods (SEPA, 2002). DON was quantified as TDN minus ammonia, nitrite, and nitrate. A turbidity meter (AQ4500, Thermo Orion) and a pH meter (HQ40d, HACH) were calibrated prior to use. The physical properties of the media were measured using a Surface Area and Porosity Analyzer (Micromeritics, ASAP 2020 + C).

The lipid phosphorus method was used to determine media biomass (Yu et al., 2002). The biomass was represented by the amount of phosphorus in the phospholipid of microbial membranes, and the unit was nmol (P) cm^{-3} (filter), where 1 nmol P is equal to 10^8 *Escherichia coli* cells. The experimental steps were as follows. (1) First, approximately 5 g wet weight of the experimental media was placed in a 100-mL stoppered flask, (2) 5 mL chloroform, 10 mL methanol, and 4 mL pure water (volume ratio of 1:2:0.8) were added, (3) the mixture was shocked for 10 min on a shaker and allowed to stand for 12 h, (4) 5 mL chloroform and 5 mL pure water were added, making the final chloroform:methanol:water composition 1:1:0.9, and the mixture allowed to stand for another 12 h, (5) the lower layer of the chloroform phase containing the lipid component was transferred to a round bottom flask and the liquid was evaporated in a water bath, and (6) potassium persulfate solution was added to the round bottom flask, which was then sealed, to allow for digestion for 30 min in a high-pressure steam sterilization pot at 121 °C. The total phosphorus content was determined using an ammonium molybdate spectrophotometric method (SEPA, 2002). The media were dried for 2 h at 105 °C in an oven and weighed after cooling to calculate the biomass per unit volume of media.

Biological activity was determined using the method of microbial specific oxygen uptake rate (SOUR). SOUR was defined as the unit of oxygen consumed by a unit of biofilter media in the unit of time, i.e., mg (O_2) cm^{-3} (filter) · h (Urfer and Huck, 2001). The specific experimental steps were as follows: (1) a media sample (wet weight about 5 g) from was taken from the biofilter; (2) the sample was transferred to a 250-mL narrow mouth bottle with a magnetic stirrer in it; and (3) the bottle was filled with raw water containing a sufficient amount of dissolved oxygen (DO) (>5 mg L^{-1}). If the amount of DO in the water was insufficient, pre-aeration was performed; (4) a rubber stopper including a DO probe was lightly placed in the mouth of the bottle to ensure that no air bubbles or biomass were lost, and the bottle was sealed with a polytetrafluoroethylene film seal; (5) the bottle was placed a magnetic stir plate and the DO meter turned on. The DO reading was recorded after stabilization for 2 min and then DO was continuously measured for 20 min; (6) the SOUR of the raw water sample blank was determined according to steps (3) and (4) without the

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