Water Research 101 (2016) 402–410

Contents lists available at ScienceDirect

Water Research

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

Depth investigation of rapid sand filters for drinking water production reveals strong stratification in nitrification biokinetic behavior

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ARTICLE INFO

Article history:
Received 3 December 2015
Received in revised form
8 April 2016
Accepted 27 April 2016
Available online 9 May 2016

Keyword:
Groundwater treatment
Ammonium oxidizing bacteria
Nitrification capacity
Nitrification kinetics
Loading

ABSTRACT

The biokinetic behavior of NH_4^+ removal was investigated at different depths of a rapid sand filter treating groundwater for drinking water preparation. Filter materials from the top, middle and bottom layers of a full-scale filter were exposed to various controlled NH_4^+ loadings in a continuous-flow lab-scale assay. NH_4^+ removal capacity, estimated from short term loading up-shifts, was at least 10 times higher in the top than in the middle and bottom filter layers, consistent with the stratification of Ammonium Oxidizing Bacteria (AOB). AOB density increased consistently with the NH_4^+ removal rate, indicating their primarily role in nitrification under the imposed experimental conditions. The maximum AOB cell specific NH_4^+ removal rate observed at the bottom was at least 3 times lower compared to the top and middle layers. Additionally, a significant up-shift capacity (4.6 and 3.5 times) was displayed from the top and middle layers, but not from the bottom layer at increased loading conditions. Hence, AOB with different physiological responses were active at the different depths. The biokinetic analysis predicted that despite the low NH_4^+ removal capacity at the bottom layer, the entire filter is able to cope with a 4-fold instantaneous loading increase without compromising the effluent NH_4^+. Ultimately, this filter up-shift capacity was limited by the density of AOB and their biokinetic behavior, both of which were strongly stratified.

1. Introduction

Rapid sand filtration is widely used in drinking water production to remove compounds such as NH_4^+, Fe^{2+} and Mn^{2+}, which are typical for anoxic groundwaters. Among these compounds, Fe^{2+} and Mn^{2+} are chemically and biologically oxidized to low-solubility oxyhydroxides and are removed by precipitation in the filter, while NH_4^+ is biologically oxidized to NO_3^-. EU member states must comply with the guideline limit of 0.5 mg/L NH_4^+ in the water leaving the waterworks (Council Directive 98/93/EC, 1998), while stricter limits can be applied by the individual member states e.g. 0.05 mg/L NH_4^+ in Denmark (Danish Ministry of Environment, (2014). Guideline limits for NH_4^+ are set to ensure biological stability in the down-stream distribution network in non-chlorinated systems (Chu et al., 2005), and to control the disinfection residual in chlorinated systems (Zhang et al., 2009).

Single media filters are typically assumed to be homogeneous due to their frequent backwashing (Uhl and Gimbel, 2000). However, recent studies have shown that some filters have heterogeneous flow patterns (Lopato et al., 2013) and stratified biomass distributions even with regular backwashing (Albers et al., 2015; Bai et al., 2013; Feng et al., 2013). Stratification can be the result of changes in the filter material density if mineral precipitates form a persistent coating on the sand grains (Gülay et al., 2014). In this case, the lighter mineral-coated grains accumulate preferentially at the top of the filter after each backwashing. There, they are exposed to higher NH_4^+ loadings compared to the heavier less coated grains, which reside deeper in the filter. Stratification of both nitrifying bacterial density and nitrification activity has, indeed, been observed in samples collected from full-scale filters (de Vet et al., 2009; Kihn et al., 2000; Madoni et al., 2001). The assays used in these works involved strong physical mixing that is likely to detach the microbial activity from the filter material, poorly reflecting the *in situ* biokinetics. Proper estimation of the *in situ* biokinetics would maintain the spatial attachment of the biomass, and the associated supposed mass transfer limitations.

Stratification of nitrification activity was also observed in pilot-scale filters (Lee et al., 2014; van den Akker et al., 2008). Specifically, Lee and collaborators investigated filter response to short-term NH_4^+ loading increases, created by increasing either the influent

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http://dx.doi.org/10.1016/j.watres.2016.04.073
0043-1354/

NH_4^+ concentration or the flowrate. No difference was observed between the two increased loading scenarios, and the overall filter capacity was estimated to be 5 times higher than the normal operating loading (Lee et al., 2014). Most of this additional capacity was concentrated in the top 0.15 m, while deeper filter layers exhibited very small additional capacities (Lee et al., 2014). This stratification in activity was solely attributed to the change of Ammonium Oxidizing Bacteria (AOB) density with depth, assuming that AOB have the same biokinetic behavior everywhere in the filter (Lee et al., 2014). The biokinetic behavior itself, however, was not directly examined for stratification with depth.

In the present study, we investigate in detail the previously observed limited nitrification capacity at the bottom layer of a rapid sand filter. We aim to examine whether this stratification in activity is exclusively the result of a decrease in AOB density. Hence, the NH_4^+ removal capacity at different depths of a full-scale filter was investigated by exposing the filter material to a series of controlled loading conditions in an offline continuous-flow assay. Molecular analysis before and after the experiments was used to identify the predominant nitrifiers, and to calculate cell specific NH_4^+ removal rates at each depth. The activity observations were augmented by a 1-D biofilm model analysis to estimate the kinetic parameters at each depth. Ultimately this analysis aimed to reveal if the NH_4^+ removal biokinetics is also stratified in the filter.

2. Materials & methods

2.1. Filter core sampling and depth profiling

Islevbro (Copenhagen, Denmark) waterworks was selected due to the strong activity stratification observed in previous pilot-scale investigations (Lee et al., 2014). The raw water is abstracted from a deep chalk aquifer and the treatment train consists of an aeration step, followed by a retention tank providing about 20 min contact time for oxidation of Fe^{2+}, and a double filtration step consisting of pre- and after-filters. Pre-filters have a bed of coarse stones (3–6 cm diameter) to retain the formed Fe-hydroxides, while after-filters are designed to biologically remove NH_4^+. After-filters have an average influent flowrate of 1.73×10^6 L/d, an influent concentration of 0.13 (±0.04) mg/L NH_4^+-N, a cross sectional area of 18 m^2 and are 0.70 m deep. The hydraulic loading is 4 m/h and the filters have been in operation for approx. 30 years without filter material replacement. Nominal grain diameter is 1 mm and after-filters are backwashed every 14 days by air scouring (3 min at 90 m/h) and high water flowrates (10 min at 25 m/h).

Filter material from one after-filter was core-sampled by inserting and gently removing a plexiglass cylinder (1 m height and 5 cm inner diameter), closed at the top with a rubber stopper. From the collected 0.50 m filter material core three depth layers were separated: 0–0.10 m (top), 0.20–0.30 m (middle) and 0.35–0.50 m (bottom) that were used for the lab-scale NH_4^+ removal investigations. The full-scale filter was core-sampled at 3 randomly selected locations and composite samples were created by mixing filter material from the same depth. Sampling took place approximately midway between two backwash events (day 9 after last backwashing). Additionally, 6 filter material sub-samples were segregated every 0.05–0.10 m from the collected core for molecular quantification of AOB and Ammonium Oxidizing Archaea (AOA). Filter material samples for the NH_4^+ removal investigations were stored wet at 10 °C for 4 days, while sub-samples for molecular quantification were drained and stored at −20 °C.

The *in situ* NH_4^+ profile in the filter was obtained by sampling water at 0.05, 0.1, 0.15, 0.2, 0.3 and 0.4 m depth during 6 sampling campaigns conducted over 2 filter-run cycles. Stainless steel pipes were fixed in the middle of the filter and were connected to a multi-channel peristaltic pump (110 ACR, Ole Dich) sampling water at a 0.6 L/h. Water samples were immediately filtered (Sartorius Minisart 0.20 μm), frozen and analyzed for NH_4^+ (Merck Spectroquant test kit 1.14752) within 7 days. The *in situ* volumetric NH_4^+ loading at each filter depth was calculated as the product of the measured NH_4^+ concentration and the flowrate, normalized for the packed filter material volume of the respective depth section.

2.2. Quantification of depth specific NH_4^+ removal rates

Filter material from the three investigated depth layers was exposed to a series of NH_4^+ loading levels in a lab-scale column assay to observe the removal rate at each imposed condition. The assay is described in details elsewhere (Tatari et al., 2013). In brief, it consisted of small (5 cm bed height, 2.6 cm inner diameter) columns packed with the collected filter material that were operated under continuous flow conditions. Filter material from each depth layer was packed in separate columns and the three columns were operated in parallel. The effluent from each column was recirculated at a high recirculation ratio (50) in order to approximate completely-mixed bulk hydrodynamic conditions (Tatari et al., 2013). Each column was supplied with effluent water from Islevbro waterworks, supplemented with 1 mg/L NH_4^+-N (as NH_4Cl, Merck chemicals) at an influent flowrate of 0.94 L/d. Recirculation for each column was at 46.8 L/d and the hydraulic loading (influent plus recirculation) was 3.8 m/h, matching full-scale conditions (Tatari et al., 2013). The resulting volumetric (expressed per volume of packed filter material) loading was 35 g NH_4^+-N/m^3 filter material/d, termed the reference loading. The reference loading was chosen as an equivalent to the full-scale filter loading, based on the initial assumption that complete NH_4^+ removal occurs in the upper 0.40 m of the filter. The *in situ* loading at each depth was calculated based on the depth profiles and was compared to the reference loading. The experimental set-up is schematically illustrated in the SI (Fig. S1).

The columns were operated at reference loading for 9 days, with intermittent short-term (3–5 h) loading up-shifts (Fig. S2 in SI). The loading up-shifts were performed within the first 4 days of operation, and the columns were operated for an additional 5 days to identify growth of nitrifiers. The loading was up-shifted to 88, 175 and 350 g NH_4^+-N/m^3 filter material/d by increasing the influent flowrate to 2.34, 4.68 and 9.36 L/d at day 3, 4 and 2 of operation, respectively, while maintaining the influent concentration at 1 mg NH_4^+-N/L. Each loading up-shift lasted 3–5 h to prevent substantial growth of nitrifiers during these short-term perturbations. External mass transfer conditions were maintained by keeping a nearly steady total (influent and recirculation stream) hydraulic loading to the columns (Tatari et al., 2013). Experiments were carried out at 10 °C to mimic the *in situ* water temperatures. Column influents and effluents were manually sampled and analyzed for NH_4^+ and NO_2^- (Merck Spectroquant test kits 1.14752 and 1.14776 with detection limits of 0.01 and 0.002 mg N/L respectively). The volumetric removal rate at each loading was calculated from concentration differences multiplied by the influent flowrate and normalized for the volume of packed filter material in the column. The maximum observed NH_4^+ removal rate from each column was termed the NH_4^+ removal capacity and was used to predict the full-scale filter performance as described in Section 2.5. At the end of the experiments, filter material from each column was collected and stored at −20 °C for molecular quantification.

2.3. Molecular quantification of total bacteria and nitrifiers

Total *Eubacteria*, *Nitrospira*, AOB and AOA were quantified by real-time quantitative PCR (qPCR). Genomic DNA was extracted in