



Hotspots for selected metal elements and microbes accumulation and the corresponding water quality deterioration potential in an unchlorinated drinking water distribution system



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ABSTRACT

Biofilm formation, loose deposit accumulation and water quality deterioration in drinking water distribution systems have been widely reported. However, the accumulation and distribution of harbored elements and microbes in the different niches (loose deposits, PVC-U biofilm, and HDPE biofilm) and their corresponding potential contribution to water quality deterioration remain unknown. This precludes an in-depth understanding of water quality deterioration and the development of proactive management strategies. The present study quantitatively evaluated the distribution of elements, ATP, *Aeromonas* spp., and bacterial communities in distribution pipes (PVC-U, D = 110 mm, loose deposit and biofilm niches) and household connection pipes (HDPE, D = 32 mm, HDPE biofilm niches) at ten locations in an unchlorinated distribution system. The results show that loose deposits in PVC-U pipes, acting as sinks, constitute a hotspot (highest total amount per meter pipe) for elements, ATP, and target bacteria groups (e.g., *Aeromonas* spp., *Mycobacterium* spp., and *Legionella* spp.). When drinking water distribution system niches with harbored elements and microbes become sources in the event of disturbances, the highest quality deterioration potential (QDP) is that of HDPE biofilm; this can be attributed to its high surface-to-volume ratio. 16s rRNA analysis demonstrates that, at the genus level, the bacterial communities in the water, loose deposits, PVC-U biofilm, and HDPE biofilm were dominated, respectively, by *Polaromonas* spp. (2–23%), *Nitrosipira* spp. (1–47%), *Flavobacterium* spp. (1–36%), and *Flavobacterium* spp. (5–67%). The combined results of elemental composition and bacterial community analyses indicate that different dominant bio-chemical processes might occur within the different niches—for example, iron-arsenic oxidizing in loose deposits, bio-calumniation in PVC-U biofilm, and methane oxidizing in HDPE biofilm. The release of 20% loose deposits, 20% PVC-U biofilm and 10% HDPE biofilm will cause significant changes of water bacterial community.

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1. Introduction

The deterioration of the quality of water during its transport through a drinking water distribution system (DWDS) has been

widely observed in the form of increased particle load (Liu et al., 2016), heterotrophic plate counts (HPC), and *Aeromonas* plate counts; these are the traditional microbial indicators for regrowth (van der Wielen et al., 2016) at customers' taps. In extreme cases,

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aesthetic problems such as discolored water (dirty water, red water) may occur, or the water may contain heavy metals and health-problem associated (opportunistic) pathogens; these risks might increase in the event of disturbances (Sly et al., 1990; Vreeburg and Boxall, 2007; Li et al., 2010; Schwake et al., 2016; Liu et al., 2017).

A DWDS is a pressurized pipe network which delivers treated drinking water from a centralized treatment plant to the water meters at the consumers' buildings (Snoeyink et al., 2006). It is a complex system typically consisting of different kinds of pipe, including transportation pipes (which connect the treatment plant and reservoir with the supply areas, with a typical diameter of >200 mm); distribution pipes (the main pipes under the street, which distribute water within the supply area, with a typical diameter of 63–110 mm); and household connection pipes (which connect the distribution pipe networks to the water meters at the consumers' building, with a typical diameter of 25–32 mm). Water quality deterioration has been widely observed during transport in both distribution pipes and household pipes; the phenomenon has been attributed to the water's long retention time in these pipes and to the pipes' high surface-to-volume ratio (Tsvetanova and Hoekstra, 2010; Liu et al., 2013a,b,c). Moreover, there are different niches present within a pipe section, e.g., pipe surfaces and loose deposits (Liu et al., 2014; Proctor and Hammes, 2015; Prest et al., 2016; van der Wielen and Lut, 2016).

Although water quality has been significantly improved over the last decades as a result of new and/or improved treatment processes at the plants, the treated drinking water that enters a DWDS still contains particles, microorganisms, and nutrients (Liu et al., 2013a,b,c; Proctor and Hammes, 2015; Prest et al., 2016). During drinking water distribution, the niches present within a DWDS become sinks for particle accumulation and microbial growth, which gradually develop and stabilize over lengthy periods of time as the water flows through the system (Boe-Hansen et al., 2002; Liu et al., 2013a,b,c; Makris et al., 2014). The established sinks/niches have been studied and sub-divided into pipe scales (Sarin et al., 2001; Renner, 2008; Makris et al., 2014), biofilm matrices (Flemming and Wingender, 2010; Fish et al., 2016), and loose deposits (Smith et al., 1997; Gauthier et al., 1999), all of which can constitute reservoirs for organic compounds, heavy metals, and microbes (including pathogens) (Liu et al., 2017).

It is noteworthy that, in the event of a disturbance which destabilizes the established physiochemical and microbiological equilibriums, these different components can become resuspended in the drinking water and thereby result in a deterioration of water quality (Makris et al., 2014). Such destabilization can be caused by hydraulic turbulence of a peak velocity in the pipe (e.g., during a morning water-demand peak or a firefighting event) (Matsui et al., 2007; Vreeburg and Boxall, 2007); and/or by changes on the physiochemical and microbiological water characteristics (e.g., as a consequence of implementing disinfection strategies, switching source water, and changing treatment processes) (Li et al., 2010; Schwake et al., 2016; Liu et al., 2017).

To resolve these water quality and the related public health concerns associated with drinking water distribution, an understanding of the accumulation and distribution of material (e.g., cells, particles, and metals) across different niches in different DWDS pipes is critical. Although there is a consensus about the major part played by the DWDS in water-quality deterioration, the function of the DWDS as a sink (contaminants accumulation) and source (contaminants release), and the specific contribution of each niche remain unknown. This is especially true of full-scale distribution systems because of their low accessibility (Berry et al., 2006). The objectives of this study are (i) to identify hotspots for the accumulation of different microbial parameters and selected metals, and (ii) to determine the corresponding water quality

deterioration potential (QDP) that predicts the possible contribution of each DWDS niche to drinking water deterioration in a full-scale DWDS.

2. Material and methods

2.1. Sampling

The sampling was conducted between February and March 2014 in the unchlorinated DWDS of the Oasen drinking water company in the central area of the Netherlands. At the drinking water treatment plant (DWTP) the abstracted groundwater is submitted to aeration, rapid sand filtration, softening, activated carbon filtration, and UV disinfection before the treated water is pumped into the distribution system. In the treated water, Fe, Mn, As, Al and *Aeromonas* spp. are below detection limit, with 23 ± 1.2 mg/l Ca and 8.0 ± 2.3 ng/l ATP.

As illustrated in Fig. 1 and Table S1, ten locations were selected for this study in the DWTP's supply areas. An integral sampling from PVC-U distribution pipes ($D = 110$ mm) was performed at each location, as previously described (Liu et al., 2014). In summary, water samples were collected from customers' taps connected directly to the main supply, and close to the hydrants for flushing loose deposits. Water samples were taken from each tap after the taps were left to run until the water temperature is constant. Loose deposits were collected at fire hydrants by flushing the distribution pipe with a velocity of 1.5 m/s (Vreeburg et al., 2008). Subsequently, two sections of the flushed pipe (length = 30 cm) were cut out to sample the biofilm in duplicate. The pipe sections were closed using pre-disinfected caps and filled with 1 l DNA-free water (Millipore, H20MB1006) to keep the inner surface wet during transportation. The HDPE household connection pipes ($D = 32$ mm) were taken in duplicate at each sampling location ($l = 30$ cm), closed using pre-disinfected caps, and filled with DNA-free water.

The sampling procedure involved the following steps: obtaining the water samples, removing the household connection pipes, flushing the distribution pipe in the street for loose deposit sampling, and cutting out parts of the distribution pipe. During flushing, the turbidity was recorded online, the timing of loose deposit sampling at hydrant is calculated according to distance between flushed hydrant and cut pipe specimen as detail described in Fig. S1. The online recorded turbidity and measured ATP of flushed loose deposits were included in Fig. S2. All samples were kept at 0 °C as soon as they were taken and subsequently transported at 0 °C to the lab. To detach the bacteria from the surface of the loose deposits and pipe material, the samples were pre-treated three times using 2-min ultrasonication at 42 KHz (Magic-Knezev and van der Kooij, 2004). The obtained suspensions were used for further physiochemical and microbiological analysis. All samples were processed within 24 h of being taken.

2.2. Physiochemical analysis

Concentrations of Iron (Fe), Manganese (Mn), Calcium (Ca), Aluminum (Al), and Arsenic (As) were determined by inductively coupled plasma-mass spectroscopy (ICP-MS, PerkinElmer ELAN DRC-e), as previously described (Lytle et al., 2004; Peng et al., 2010; Liu et al., 2014). Quality control samples, including laboratory-fortified blanks and laboratory-fortified samples, were performed for every ten samples analyzed.

2.3. Microbiological analysis

2.3.1. Adenosine triphosphate (ATP)

The ATP concentrations, as a measure for active biomass, were

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