

## CHEMICAL AND MICROBIAL COMPOSITION OF LOOSE DEPOSITS IN DRINKING WATER DISTRIBUTION SYSTEMS

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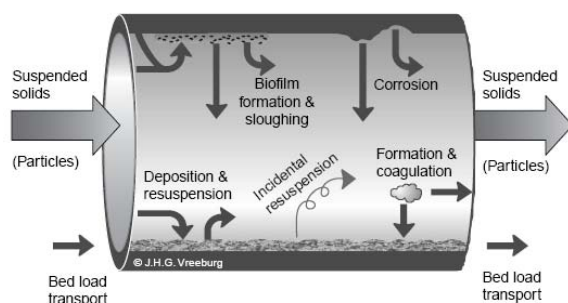
**Abstract.** Processes occurring in drinking water distribution system can form mineral and biological materials. These materials are present in the distribution systems as loose deposits. Origin of the mineral sediments in water networks is often linked to an insufficient treatment process or corrosion of distribution system pipes. The bacterial regrowth processes associated with biofilm formation may originate within these loose deposits. Sudden changes in hydraulic conditions may resettle the loose deposits leading to increase in turbidity or deterioration of microbiological water quality at the consumer tap, which in some circumstances can cause health risk.

This paper reviews current literature for composition of loose deposits and presents a simple operational method for removal of loose deposits from drinking water distribution system using unidirectional flushing (velocity 1.5 m/s, clear water front, 2 pipe turnovers). This method for effective loose deposit removal can be used to evaluate chemical and microbial status of a drinking water distribution system.

**Keywords:** loose deposits; drinking water distribution networks; unidirectional flushing.

### 1. Introduction

Within drinking water distribution systems (DS) a broad range of microbiological and chemical processes take place which form products as biological cover (biofilm) or sediments (particles, loose deposits) in the bulk water and on the inner surface of pipes (see Fig. 1). The particles with average size of 0.01 mm can be transported through the network due to turbulent forces [1].



**Fig. 1.** Overview of processes in the drinking water DS influencing the water quality

The sediments found in DS are mostly linked with deficient treatment processes or corrosion of pipes. There is an opinion that bacterial regrowth in DS is associated with biofilm formation although microorganisms may originate within the loose deposits as well. When the equilibrium of regrowth processes is disturbed or hindered, bacteria are released into the bulk water. The main reasons for this release are changes in nutrient concentration, fluctuating disinfection residuals and, assumedly, temperature changes. Increase of turbidity (e.g. “red water”, discoloration events) can take place as well as a result of sudden changes in hydraulic conditions (pump shut on and off, sudden valve closure). Loose deposits may be withdrawn from the networks leading to aesthetic complaints of consumers or deterioration of microbiological water quality at the taps and an increase in colony counts, which in some circumstances can cause a health risk. Thus, the distribution of drinking water has become one of the essential objectives in developed countries [2]. Therefore drinking water companies should pay more attention to loose deposit removal from DS.

The role of loose deposits on water quality, especially microbial parameters, is not fully understood. It is partly due to limitations and diversity of

methodology which was used in earlier studies for removal of loose deposits and for analyses of bacteria there.

This paper reviews literature of methods for collection of loose deposits and analyzing their microbiological and chemical composition. A simple method for removal of loose deposits from drinking water DS using unidirectional flushing (UDF) is presented in the section 10, Conclusions.

## 2. Countries and legislation.

The raw water quality and treatment in Latvia and in the Netherlands are quite different. In Latvia raw waters have concentrations of natural organic matter (NOM) measured as total organic carbon (TOC) of 15-20 mgC/L and 5 mgC/L in drinking water. Groundwater displays high concentration of iron in range 1-4 mg/L and manganese 0.1-0.5 mg/L. To control bacterial regrowth in drinking water treatment process chlorination as a disinfection method is used. In the Netherlands drinking water treatment from surface and groundwater is carried out without chlorination and for bacteriological safety combination of treatment methods as dune filtration, slow sand filtration, UV-disinfection, membrane filtration and ozonation are used (multiple barrier approach). For microbiological stability of drinking water a threshold value of assimilable organic carbon (AOC) of 50 µg/L is set in the Netherlands [3]. AOC is believed as a limiting factor for bacterial regrowth, while in Northern regions of Europe and Japan the limiting element is considered phosphorus [4, 5, and 6]. Although the treatment philosophy in both countries is different the quality goals and research topics for microbiologically stable and aesthetic drinking water are similar.

The microbial quality in the Drinking Water Directive 98/83/EC (DWD) of European Union (EU) countries is defined by setting maximal allowed limits of human gastroenteric bacteria *Escherichia coli* (*E. coli*) and Enterococci. Concentration of these microorganisms should not exceed 0 cells in 100 mL of water sample analyzed using cultivation methods [7].

Iron and manganese concentrations in drinking water cannot exceed 0.2 mg/L and 0.05 mg/L, respectively. To control drinking water turbidity in DWD there is not unitary methodology. There is a suggestion that the turbidity in drinking waters cannot exceed 1 NTU (nephelometric turbidity unit).

Spore forming bacteria *Clostridium perfringens* is a parameter which should be analyzed in some specific conditions, namely, after distribution main disinfection and in DS, which are supplied by surface waters.

In the DWD heterotrophic plate count (HPC) is used as a parameter to control bacteria number in bottled water. However, many EU countries have adapted HPC also for drinking water control. The maximal allowed value is 100 colony forming units (CFU)/mL after water cultivation on media for 3 days at 22°C.

## 3. Methods for analyzing biofilms, deposits, tubercles in distribution system

To analyse microbiological quality of accumulated bacteria on pipe surface research laboratories use several types of lab-scale systems: Rotortorque®/annular reactors [8], Propella® [9] and flow-cells. Bacterial concentration in bulk phase and biofilm can very effectively be analyzed using these systems as opposed to the behavior of loose deposits. The particle transport has been studied in pilot Torus [10] and other lab-scale pipe-loop systems [11, 12]. To get information about chemical and microbiological composition of loose deposits in DS it is necessary to use more practical approaches - cutting of a piece of the pipe section, particle counting and flushing of the distribution mains (Fig.2.).



Fig. 2. Flushing of a distribution main

## 4. Conventional flushing.

One of the cheapest and simplest methods for removal of loose deposits is a conventional flushing technique of DS through the hydrants and leaving them open until certain water quality objectives are met, mostly by a visual assessment of turbidity. Flushing velocities are not necessarily maximized and the water used to flush a particular pipe may not have originated from clean or pre-flushed pipes. The disadvantages of the technique are the quite large amount of water used and its unsuitability for large diameter pipes because it is usually not possible to achieve the desired flushing velocity. Often there can be observed increase of consumers complains as discoloration effect during and immediately after implementation of flushing do to low velocities that are the result of randomly opening fire hydrants [13].

The loose deposits were collected during conventional flushing with velocity 1.5 m/s of distribution main by attaching nylon netting (approx. pore size 300 µm) to the hydrant nozzle. The total amount of flushed water consisted of five pipe volumes (approx. 60 m<sup>3</sup>) [14]. The analyses of first flushed (30 sec) loose

deposits with inductively coupled plasma spectrometry showed that it contained 21.4% (by weight) of zinc, indicating that the loose deposits were predominantly accumulated zinc orthophosphate used for corrosion control. These loose deposits contained HPC in the range of  $2.4 \times 10^6$  CFU/mL and 2.2 coliform organisms per 100 mL. The HPC population was composed primarily of *Flavobacterium spp.* and *Pseudomonas vesicularis*. During the all flushings 25-30 g of sand were collected where HPC (R2A) level  $8.4 \times 10^6$  CFU/g was observed. These deposits predominantly consisted of *Arthobacter spp.* as well *Pseudomonas spp.*, *Flavobacterium spp.* and *Moraxella spp.* The *Enterobacter agglomerans* (coliforms) were detected at a concentration of 0.5 CFU/g of loose deposits.

The loose deposit analyses from hydrant flushing showed that volatile solids VS linearly correlated with the organic carbon fraction ( $VS(\%) = 3.2 C(\%) + 4.5$ ;  $R^2 = 0.81$ ,  $p > 99.9$ ) [15]. Infrared spectrometry indicated the differences in composition of the organic fraction of deposits and demonstrated that the organic nature of deposits was not uniform even in a DS supplied with only one type of treated water.

## 5. Pigging.

Pigging-swabbing technology involves driving of cylindrical foam sponge through pipes using water pressure. The pig has a diameter approximately 25% greater than the pipe and it is being forced through the pipe. The advantages of this method are the decreased water demand compared to flushing and the absence of diameter limitation. However pigging is more expensive because of necessary disinfection of the pig and required installations for insertion of them into the main. There is also a possibility that the pig may break or stick into the main.

Zacheus [16] and Torvinen with co-authors [17] showed that the repeated deposit collection a year after cleaning of the pipes using pigging had similar bacterial concentration than the loose deposits after the first cleaning. This can be explained by the fact that after reaching of the steady state within the first month microbiological quality didn't change further. There were found coliform bacteria in four old deposit samples out of studied eight samples [16]. The high number of coliform bacteria in two deposit samples could not allow the bacteria to be enumerated because the dilution of samples was not high enough (numbers were over  $10^3$  CFU/mL). In two samples the number of coliforms were 0.2 and 0.35 CFU/mL ( $2.6 \times 10^1$  and  $3.0 \times 10^2$  CFU/g dry weight). The mean number of HPC (R2A) in the loose deposits was  $5.4 \times 10^3$  CFU/mL. Actinomycetes and fungi were also found in the loose deposit samples. The deposits were mainly composed of iron (18%), manganese (3.7%), aluminium (4.5%) and calcium (3.0%). The highest concentration was measured in a deposits collected from the DS fed with groundwater. Concentration of manganese ( $r = -0.77$ ,  $p < 0.01$ ) and copper ( $r = -0.93$ ,  $p < 0.01$ ) correlated negatively with the HPC number. The

cultivable bacteria in loose deposits were not affected either by the origin of drinking water, pipe material, water temperature, pH, turbidity and concentration of chlorine, non-volatile organic carbon, iron and manganese.

There was found that the number of mycobacteria in deposits correlated positively with the number of HPC (R2A, 7days) ( $r = 0.64$ ,  $p < 0.051$ ) but negatively with manganese ( $r = -0.64$ ,  $p < 0.05$ ) and strontium ( $r = -0.71$ ,  $p < 0.01$ ) [17]. The numbers of mycobacteria ( $1.8 \times 10^5$  CFU/g) in deposits were higher than in the bulk water and higher in ozonated surface water than in chlorinated waters due to better degradation of NOM. The number of mycobacteria in bulk water correlated positively ( $r = 0.74$ ,  $p < 0.01$ ) with the AOC concentration at the effluent from water treatment plant (WTP), the same tendencies was observed earlier [18]. The three most ubiquitous species according to Torvinen and co-authors were *M. lentiflavum*, *M. tusciae* and *M. gordonae* [17]. It can be concluded that presence of mycobacteria was not observed with routine coliform bacteria measured with cultivation methods.

The pigging of a distribution main by LeChevallier and co-authors released iron tubercles (98.7% iron) containing high densities ( $> 160$  CFU/g) of coliform organism such as *E. coli* (fecal biotype), *Citrobacter freundii* and *E. agglomerans* [19]. They concluded that these tubercles contained the highest densities of coliform bacteria compared to loose deposits from flushing and pipe surface itself.

## 6. Air scouring.

Air scouring involves the controlled injection of filtered and compressed air into the pipes, usually via hydrant (Fig.3). Therefore given continuous supply of water and air in the right proportions, discrete „slugs” of water are formed in the main and driven along by the compressed air at high velocity. There is no need to turn the water or air on and off to achieve this effect. This method is effective in pipes with diameter of less than 200 mm, there is necessity for skilled staff and the precautions to prevent air contamination with compressor oil must be taken into account. Attention should be drawn to this procedure after stopping the air injection, because an additional usage of water is necessary in order to remove the air from the DS.

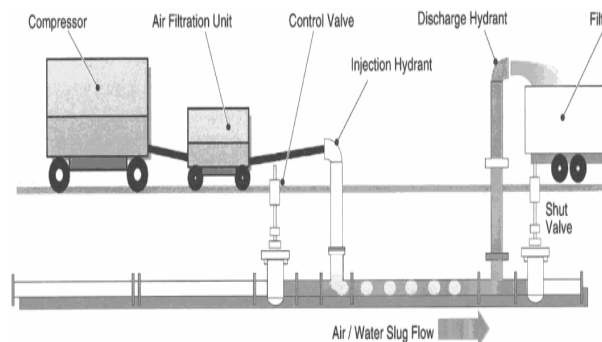


Fig. 3. Arrangement for air scouring operation [20]

Finnish researchers used this methodology with compressed air-water to analyze loose deposits [21]. Flushing velocity was 3-12 m/s. Loose deposit samples were collected at the beginning of the flushing when the thickest deposits were coming from the pipe. The sediments data was divided into the periods with high (0.4 mg/L and 10% of all samples) and low concentration of iron. For samples with high concentration, iron positively correlate with AOC ( $r=0.73$ ,  $p=0.007$ ), total bacteria number (TBN,  $r=0.69$ ,  $p=0.004$ ) and microbially available phosphorus (MAP,  $r=0.63$ ,  $p<0.027$ ). In these samples AOC, TBN, MAP and HPC were in average higher than in the samples with iron concentration less than 0.4 mg/L. Within loose deposits no coliform bacteria (analyzed with cultivation method) or Norwalk-like viruses were found.

## 7. Deposits from storage reservoirs.

Loose deposits can be collected from uninstalled water meters or emptied reservoirs. This approach has been used by French and American scientists. They reported that deposits from reservoirs contain only minor fraction of organic matter, on average 18.9% of dry weight, as expressed from the volatile solid (VS) measurements, and 5.0% of dry weigh, as expressed by the organic carbon analysis [22, 23]. It was found that loose deposits were mainly composed of minerals, including iron oxides (19%), insoluble sand material (18%), aluminium hydroxides (15%), calcium carbonates (10%) and manganese oxides (3%). A substantial amount of material (16%) also remained undetermined. X-ray diffraction analysis showed that quartz ( $\text{SiO}_2$ ) represented the main element in loose deposits. HPC (14 days) concentration in deposits was in average  $2.7 \pm 1.3 \times 10^8$  CFU/g but only 4% of the bacteria growing on the agar plates could be counted after 3 days of incubation.

## 8. TILVS.

Several studies are available on analysis of loose deposits after using online filtering set-up installed in DS mains [24, 25, 26, and 27]. This approach is called Time Integrated Large Volume Sampling (TILVS, see Fig.5), where 3-line filtration system containing different pore size is used for particle analysis.

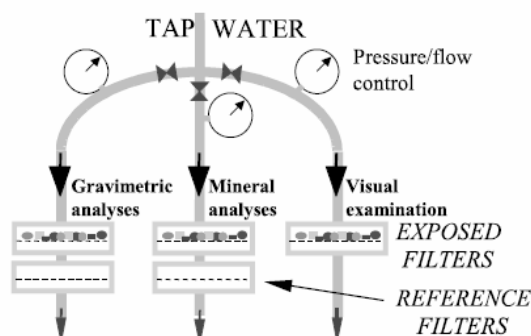


Fig. 5. Schematic picture of TILVS [24]

Gauthier with co-authors did mass balance analysis of the composition of suspended particles and concluded that throughout all DS the fraction of organic matter (as VS) ranged between 43-65%,  $\text{SiO}_2$ : 7-20%,  $\text{Al}(\text{OH})_3$ ,  $\text{CaCO}_3$  and  $\text{FeOOH}$  5% each with exception of increase of  $\text{FeOOH}$  with about 36% in the dead-end location which indicated the production of corrosion products [24, 25]. The comparison of loose deposit composition in different places of DS showed that WTP and storage tank outlets give similar quantities. In contrast, deposits from main flushing were mostly composed of iron oxides and hydroxides with very little Si compounds and negligible amount of Al and Ca (Fig. 4).

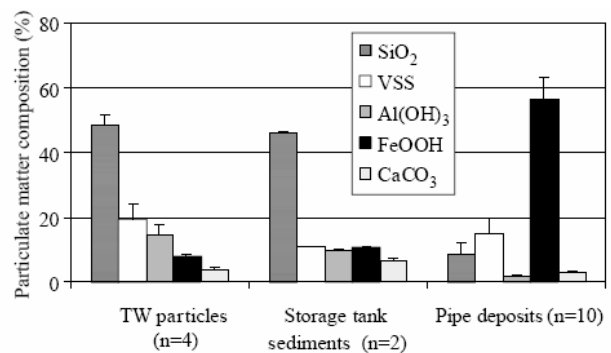


Fig. 4. Composition of treated water (TW) suspended particles during high turbidity ( $>0.5$  NTU) events, finished water storage tank sediments, and pipe loose deposits from UDF [25]

The research of particle composition in DS using TILVS showed that fraction of VS are in range of 15-55% of the total mass [25] which is lower than detected earlier [24]. Other major fractions such as  $\text{SiO}_2$ : 8%,  $\text{Al}(\text{OH})_3$ : 5% and  $\text{FeOOH}$  19% were in the range. Undetermined components accounted for less than 25% of deposits.

## 9. Unidirectional flushing.

Several studies of loose deposits have been made using UDF (velocity 0.6-2.3 m/s, turbidity  $< 1$  NTU is criterion for the "end of flushing") of DS. There were collected samples from four DS to analyze loose deposits over a 10 min period (1L/min- from faucet installed on a hydrant) [28]. This period was considered enough for effective deposit evacuation as observed in the turbidity profile. For each flushing run, total coliforms and *E. coli* were enumerated in flushing water collected at 2 and 4 min after opening the hydrant, which generally corresponds to the highest total solid concentration. Major constituent of the deposits was iron which accounted for 38-72% of deposits. The second important component found in deposits was organic matter (as VS) representing 14-24%. The percentage of calcium in deposits varied from 2-13% in different networks. Silicate was found in all of the networks studied in amount varying from 3.5-12.4%. Manganese was found in two networks in amount 0.16-5.9%. The TBN in loose deposits in one of the networks was in the range of  $10^{10}$

cells/g of dried deposits. In the other network it was not possible to analyze TBN, therefore HPC was measured instead ( $4 \times 10^7$  CFU/g of dried deposits). Only three samples were positive for total coliforms (although none for *E. coli*) out of 258 analyses, which represents 1.2% of the samples, but it should be mentioned that all flushed DS did not have a history of high total coliform occurrence. This explains the results obtained earlier [28] where 38.2% of the deposit samples collected by UDF (42 out of 110) were coliform-positive since coliforms were detected in 1.3% of the routine DS samples (252 out of 19'314).

Barbeau with co-authors studied loose deposits from the same DS using UDF in order to compare grey cast iron (CI) and cement-lined ductile iron (DI) pipes [30]. Collected loose deposits were mainly made of iron (27-71%), VS (19-22%), calcium (2.8-3.1%), manganese (0.35-6.2%) and an unknown material (5.7-45%). In addition DI pipe had higher amount of unknown material compared to CI pipe. No total coliforms were found however high numbers of atypical coliforms (up to 10 CFU/100 mL) were reported. The point made by the authors was that HPC is not a useful method for characterization of the microbiological quality of flushed waters because of the interference of residual chlorine and colonized particles. It was concluded that TBN significantly ( $p < 0.01$ ) correlated with log-transformed values of turbidities ( $r = 0.69$ ), total suspended solids ( $r = 0.78$ ) and total iron concentration ( $r = 0.55$ ). The flushing one year later give the same values of TBN ( $1.0\text{--}1.4 \times 10^{11}$  cells/g) within the loose deposits.

## 10. Conclusions.

The results from the different loose deposit collection methods (Table 1) cannot be directly compared due to difference in methodology. Some of methods were not effective thus only a part of deposits were removed and analyzed.

Two relative composition parameters of loose deposits have so far been reported, volatile suspended solids and iron in the range of 14–65% and 5–98.7%, respectively. There can be considerable fraction of calcium, silicate, aluminium, and manganese as well which depends on the pipe material, water source and the treatment technology.

The range of HPC and TBN within the loose deposits varied from  $8.4 \times 10^6$ – $5.8 \times 10^8$  CFU/g and  $10^{10}$ – $10^{11}$  cells/g. The adenosine 5-triphosphate (ATP) level within the loose deposits is found in the range 70-220 ng/L. The number of total coliforms and mycobacteria within loose deposits are in the range of  $2.6 \times 10^1$ – $3.0 \times 10^2$  CFU/g and  $2 \times 10^5$ – $4 \times 10^5$  CFU/g, respectively. More often coliforms are found after air scouring, pigging and tubercle analyses on surface of cutout piece of the main. Recent studies of pipe surfaces samples show that *E. coli* (analyzed with PNA fluorescence in situ hybridization (FISH)) is present in biofilms of European drinking water networks [31], but no *E. coli* was detected using culture or enzymatic methods. In this study cast iron and concrete

pipe sections cut out from intact water distribution main during the replacement works were used. This type of sampling is most representative but it is costly.

Some microbiological parameters within loose deposit studies correlate with inorganic parameters, such as copper, total suspended solids, manganese, strontium, iron, turbidity.

Ridgway and Olson reported that most of the chlorine-resistant microorganisms detected in a drinking water distribution system were associated with particles [32]. LeChevallier with co-authors suggested that high turbidity may play a role in coliform survival in the presence of chlorine residual because coliforms associated with particles may be protected from contact with the disinfectant [33]. However findings by McCoy and Olson show no predictable relationship between bacteriological quality and turbidity of particle counts [34].

A method for the removal of deposits introduced by Vreeburg and Boxall is based on assumption that velocity at some critical value re-suspends all sediments and disattaches the biological material which is then withdrawn out of the DS together with water flow [34]. The UDF (velocity 1.5 m/s, clear water front, 2 pipe turnovers) can be considered as effective and universal method for sampling of loose deposits during distribution network flushing. According to the pattern of turbidity obtained during flushing of mains at least three water samples of water (loose deposits) must be taken, one sample after 25% of time for the first turnover, the second after 75% of the first turnover and the last at the second turnover (Fig.6.).

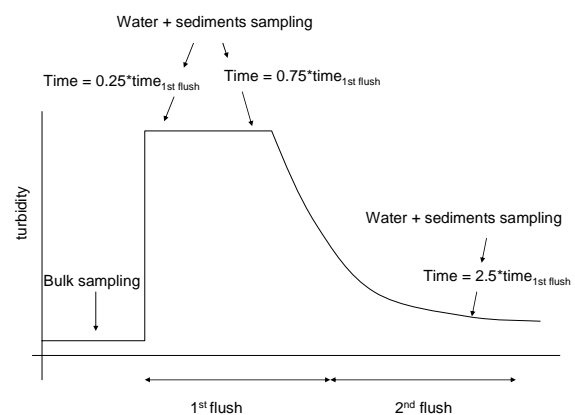


Fig. 6. The approach of sampling during UDF action

As biological material and particulates occur in low concentration in DS attention should be paid to concentration techniques, such as TILVS and Hemoflow [36] (Fig.7.). These tools can overcome the shortages of routine grab sampling by responsible utilities to fulfill DWD. These concentration techniques are necessary as studies by van Lieverloo and co-authors showed that monitoring of *E. coli* with grab sampling may not be sufficiently safe to indicate the potential health risk and does not give a representative overview of actual situation [37].



Fig. 7. Hemoflow pre-concentration unit [38]

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There hereby follows a disclaimer stating that the authors are solely responsible for the work. It does not represent the opinion of the Community and the Community is not responsible for any use that might be made of data appearing herein.

Table 1. Methods for loose deposit characterization

Method	Organics/inorganic	Microbiology	Reference
Conventional flushing with fixed nylon net on hydrant outlet	Zn 21.4%	HPC= $2.4 \times 10^6$ CFU/mL or $8.4 \times 10^6$ CFU/g, coliform= $2.2/100$ mL	[14]
		HPC= $5.8 \times 10^8$ CFU/g	[15]
Loose deposit sampling from reservoirs	FeOOH=19% SiO <sub>2</sub> =18% VS=18.9% undetermined components=16% Al(OH <sub>3</sub> )=15% CaCO <sub>3</sub> =10% MnO <sub>2</sub> =3% See [16]	HPC= $2.7 \pm 1.3 \times 10^8$ CFU/g	[15]
		HPC= $2.5 \times 10^4$ - $2 \times 10^5$ CFU/cm <sup>2</sup>	[23]
Pigging	FeOOH=98.7%	coliforms > 160 CFU/g	[14]
	FeOOH=18% Al(OH <sub>3</sub> )=4.5% MnO <sub>2</sub> =3.7% CaCO <sub>3</sub> =3.0% See [16]	HPC= $5.4 \times 10^3$ CFU/mL coliforms=0.2-0.35 CFU/mL or $2.6 \times 10^1$ - $3.0 \times 10^2$ CFU/g	[16]
		mycobacteria= $1.8 \times 10^5$ CFU/g	[17]
Air-water flushing		HPC=4545 CFU/mL TBN=110000 cells/mL coliforms=0 MAP=0.41 μg/L AOC=125 μg/L Norwalk-like viruses=0	[21]
Filtration system, TILVS	VS=43-65% FeOOH=5-36% SiO <sub>2</sub> =7-20% Al(OH <sub>3</sub> )= 5% CaCO <sub>3</sub> =5%		[24, 25]
	VS=15-55%, undetermined components < 25% FeOOH=19% SiO <sub>2</sub> =8%, Al(OH <sub>3</sub> )=5%		[26]
Unidirectional flushing	FeOOH=38-72% VS= 14-24% CaCO <sub>3</sub> =2-13% SiO <sub>2</sub> =3.5-12.4% MnO <sub>2</sub> =0.16-5.9%	HPC= $4 \times 10^7$ CFU/g TBN= $10^{10}$ cells/g total coliforms=1.2% of 258 samples (none E.coli)	[24, 28]
	FeOOH=27-71% VS=19-22% undetermined components=5.7-45% MnO <sub>2</sub> =0.35-6.2% CaCO <sub>3</sub> =2.8-3.1%	TBN= $1.0$ - $1.4 \times 10^{11}$ cells/g total coliforms=0 atypical coliforms=10 CFU/100 mL	[30]

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