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Assessment of the efficacy of membrane filtration processes to remove human enteric viruses and the suitability of bacteriophages and a plant virus as surrogates for those viruses

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ABSTRACT

Here, we evaluated the efficacy of direct microfiltration (MF) and ultrafiltration (UF) to remove three representative human enteric viruses (i.e., adenovirus [AdV] type 40, coxsackievirus [CV] B5, and hepatitis A virus [HAV] IB), and one surrogate of human caliciviruses (i.e., murine norovirus [MNV] type 1). Eight different MF membranes and three different UF membranes were used. We also examined the ability of coagulation pretreatment with high-basicity polyaluminum chloride (PACl) to enhance virus removal by MF. The removal ratios of two bacteriophages (MS2 and φX174) and a plant virus (pepper mild mottle virus; PMMoV) were compared with the removal ratios of the human enteric viruses to assess the suitability of these viruses to be used as surrogates for human enteric viruses. The virus removal ratios obtained with direct MF with membranes with nominal pore sizes of 0.1–0.22 μm differed, depending on the membrane used; adsorptive interactions, particularly hydrophobic interactions between virus particles and the membrane surface, were dominant factors for virus removal. In contrast, direct UF with membranes with nominal molecular weight cutoffs of 1–100 kDa effectively removed viruses through size exclusion, and >4-$\log_{10}$ removal was achieved when a membrane with a nominal molecular weight cutoff of 1 kDa was used. At pH 7 and 8, in-line coagulation–MF with nonsulfated high-basicity PACls containing Al_{30} species had generally a better virus removal (i.e., >4-$\log_{10}$ virus removal) than the other aluminum-based coagulants, except for φX174. For all of the filtration processes, the removal ratios of AdV, CV, HAV, and MNV were comparable and strongly correlated with each other. The removal ratios of MS2 and PMMoV were comparable or smaller than those of the three human enteric viruses and MNV, and were strongly correlated with those of the three human enteric viruses and MNV. The removal ratios obtained with coagulation–MF for φX174 were markedly smaller than those obtained for the three human enteric viruses and MNV. However, because MS2 was inactivated after contact with PACl during coagulation pretreatment, unlike AdV, CV, MNV, and PMMoV, the removal ratios of infectious MS2 were probably an overestimation of the ability of coagulation–MF to remove infectious AdV, CV, and caliciviruses. Thus, PMMoV appears to be a suitable surrogate for human enteric viruses, whereas MS2 and φX174 do not, for the assessment of the efficacy of membrane filtration processes to remove viruses.

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1. Introduction

The provision of safe drinking water is essential for ensuring public health. However, increases in the global populations of humans and domestic animals have resulted in increased demand for safe drinking water, which has led to the use of alternative water sources, sometimes of compromised quality, in many parts of the world (Bosch, 2007; Ferrer et al., 2015). These population increases have also led to increased fecal contamination of water sources, and because large numbers of pathogenic microorganisms are shed in the feces of infected people and animals, water sources receiving sewage discharge are often contaminated with those microorganisms (Rose et al., 1991; Albinana-Gimenez et al., 2006; Bosch, 2007). Therefore, water safety plans built on the principles of multiple barriers, hazard analyses, and critical control points are crucial not only for preventing the contamination of source water via human

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and animal waste but also for providing adequately treated and disinfected drinking water (WHO, 2011).

Membrane filtration, particularly low-pressure membrane filtration, is commonly used as an absolute barrier to microorganism contamination in the production of drinking water. However, although complete removal of bacteria and protozoa has been reported for the low-pressure membrane filtration processes of direct microfiltration (MF) and ultrafiltration (UF) (Jacangelo et al., 1995; Howe, 2006), varying virus removal ratios have been reported for these processes (Jacangelo et al., 1995; Madaeni et al., 1995; Urase et al., 1996; van Voorthuizen et al., 2001; Langlet et al., 2009; Boudaud et al., 2012; ElHadidy et al., 2013; Matsushita et al., 2013; Ferrer et al., 2015). The mechanisms underlying the removal of viruses by low-pressure membrane filtration include size exclusion and adsorptive interactions (i.e., hydrophobic and electrostatic interactions) between the virus particles and the membrane surface. The relative contribution of these mechanisms to the removal of viruses depends on the characteristics of the virus particles and membrane such as the relative size of the virus particles to the size of the membrane pores and the hydrophobicity and surface charge of the virus particles and membrane surface (Urase et al., 1996; van Voorthuizen et al., 2001; ElHadidy et al., 2013). Indeed, since the pore sizes of membrane filters are generally larger than the size of virus particles, there is almost no removal of viruses via direct MF when the relative contribution of absorptive interactions to virus removal is negligible (van Voorthuizen et al., 2001; Matsushita et al., 2013).

Coagulation pretreatment prior to membrane filtration, particularly in MF processes, is widely used in the water industry to improve the quality of drinking water, and the combination of coagulation pretreatment and MF processes (coagulation–MF) also shows promise as an effective means of removing virus particles. Indeed, under appropriate coagulation conditions, the virus removal performances of coagulation–MF with membrane with a nominal pore size larger than the size of the virus particles are similar or greater than the performance of direct UF with membrane with a nominal molecular weight cutoff (MWCO) smaller than the size of the virus particles (Matsui et al., 2003; Matsushita et al., 2013). Aluminum-based coagulants such as polyaluminum chloride (PACl) and alum are common coagulants in coagulation–MF processes, and coagulation conditions such as coagulation pH, coagulant dosage, and coagulation time have been shown to affect virus removal performance (Matsushita et al., 2005; Shirasaki et al., 2009; Tanneru et al., 2013). In addition, coagulant properties, such as the distribution of the aluminum hydrolyte species in the coagulant, which might be partly controlled by basicity ($[OH^-]/[Al^{3+}]$), have also been shown to affect membrane permeability and the quality of water processed by means of coagulation–MF (Zhao et al., 2010; Kimura et al., 2015). Kimura et al. (2015) reported that increasing the basicity of PACl coagulants from the typical basicity of 1.5 to a high basicity of 2.1 not only increased the removal of dissolved organic carbon and reduced the concentration of residual aluminum in the filtrate but also mitigated irreversible membrane fouling, which is a major cause of reduced membrane permeability that increases operating costs, in coagulation–MF processes. Thus, the integration of pretreatment with high-basicity PACls into MF processes may increase the removal of virus particles.

The virus removal performances of direct MF, direct UF, and coagulation–MF processes are often evaluated by using bacteriophages such as MS2, Qβ, and φX174 as surrogates of human enteric viruses because these viruses do not infect humans and are easier to cultivate than human enteric viruses (Jacangelo et al., 1995; Urase et al., 1996; van Voorthuizen et al., 2001; Matsui et al., 2003; Matsushita et al., 2005; Langlet et al., 2009; Shirasaki et al., 2009; Boudaud et al., 2012; Tanneru and Chellam, 2012; ElHadidy et al., 2013; Matsushita et al., 2013; Tanneru et al., 2013; Ferrer et al., 2015). However, data on the removal of human enteric viruses via membrane filtration is limited, particularly regarding fundamental principles such as the contributions of the size exclusion and adsorptive interaction mechanisms to virus removal via direct MF or UF and the effects of coagulation conditions and coagulant properties on virus removal via coagulation–MF, partly due to the need for bio-containment facilities for the safe handling of viruses and the fact that virus cultivation is labor intensive and time consuming (Ryu et al., 2010). Thus, whether these bacteriophages are adequate surrogates for human enteric viruses in membrane filtration processes remains unknown.

Recently, a metagenomic analysis revealed that pepper mild mottle virus (PMMoV; genus *Tobamovirus*, family Virgaviridae), which infects pepper species, was the most abundant viral RNA in human feces (concentrations up to 10^9 virus particles/g of feces; Zhang et al., 2006). In addition, because PMMoV is more frequently detected at higher concentrations in environmental waters, including drinking water sources, than are human enteric viruses (Hamza et al., 2011; Haramoto et al., 2013), PMMoV may be a potential indicator of fecal contamination of surface water. Thus, if the removal efficiencies of PMMoV and human enteric viruses are comparable, PMMoV could be a useful surrogate for evaluating the efficacy of membrane filtration processes to remove human enteric viruses.

In the present study, we used eight types of MF membranes and three types of UF membranes to investigate the efficacy of direct membrane filtration processes to remove the representative human enteric viruses (i.e., adenovirus [AdV], coxsackievirus [CV], and hepatitis A virus [HAV]) included in the Fourth Drinking Water Contaminant Candidate List (CCL4) published by the US Environmental Protection Agency (2016), and murine norovirus (MNV) as a surrogate of human caliciviruses. In addition, we examined the effects of membrane pore size and membrane material, which are related to the hydrophobic and electrostatic interactions between virus particles and the membrane surface, on virus removal. Next, we examined the potential of pretreatment with a high-basicity PACl to enhance virus removal via membrane filtration by comparing virus removal efficiencies obtained with several PACls and alum under various coagulation conditions. Finally, we examined removal efficiencies of the bacteriophages MS2 and φX174, and of the plant virus PMMoV, to assess the suitability of these viruses as surrogates for human enteric viruses.

2. Materials and methods

2.1. Source water, coagulants, and membranes

On 30 September 2015, river water was sampled from the Edo River (Tokyo, Japan; see Table 1 for water quality), which is the source water for the Kanamachi Water Purification Plant (Tokyo, Japan). The source water samples were stored at 4 °C until use and brought to 20 °C immediately prior to use.

To investigate the effect of coagulant type (i.e., effects of coagulant basicity and sulfate content) on virus removal via in-line

Table 1
Edo River water quality.

pH	7.5
Turbidity (NTU)	2.8
DOC (mg/L)	1.0
UV260 (cm^{-1})	0.020
Alkalinity (mg-$CaCO_3$/L)	36.0

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