



# Effect of aluminum hydrolyte species on human enterovirus removal from water during the coagulation process



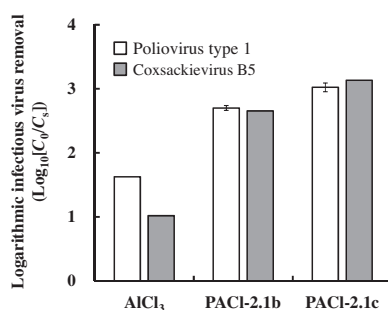
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## HIGHLIGHTS

- Differences in aluminum species in coagulant affected enterovirus removal.
- High  $Al_c$  content PACI with basicity 2.1 effectively removed enteroviruses.
- $Al_{30}$  species in PACI probably played a major role in enterovirus removal.
- PV and CV removal ratios were almost the same during coagulation.
- Viruses were removed mainly by coprecipitation into growing aluminum hydroxide.

## GRAPHICAL ABSTRACT



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## ABSTRACT

We prepared different types of aluminum-based coagulants, consisting of mainly monomeric aluminum species, polymeric aluminum species, or colloidal aluminum species, to investigate the effect of aluminum hydrolyte species on the removal of two types of human enteroviruses, poliovirus (PV) type 1 and the free-chlorine-resistant virus coxsackievirus (CV) B5, from lake and river water samples during the coagulation process. We found that differences in the distribution of the aluminum hydrolyte species in the coagulant affected the removal of these enteroviruses during coagulation: the removal ratios of PV and CV observed with polyaluminum chloride (PACI) with a high colloidal aluminum content and a basicity of 2.1 (i.e., PACI-2.1c) were larger than those observed with high monomeric aluminum content coagulant (i.e., AlCl<sub>3</sub> solution) and with high polymeric aluminum content coagulant PACI (PACI-2.1b). Unlike AlCl<sub>3</sub> or PACI-2.1b, PACI-2.1c contains Al<sub>30</sub> species, indicating that Al<sub>30</sub> species probably play a major role in the removal of enteroviruses. The PV and CV removal ratios were almost identical, regardless of the coagulant type or viral quantification method used (plaque-forming unit method or real-time polymerase chain reaction method), suggesting that PV and CV behaved similarly during the coagulation process. We also experimentally confirmed that the main mechanism for virus removal was coprecipitation into growing aluminum hydroxide during charge neutralization; virus adsorption onto formed aluminum hydroxide flocs also contributed to virus removal, but played a limited role.

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## 1. Introduction

Coagulation is an important process in water treatment to remove contaminants. Of the various coagulants used for the coagulation process, prepolymerized aluminum coagulants, such as polyaluminum chloride (PACI), have been widely used in drinking

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water treatment because of their superiority to traditional aluminum coagulants [e.g., aluminum chloride ( $\text{AlCl}_3$ ) and alum]; PACls are more efficient, are less dependent on temperature and pH, can be used at a lower dosage, and produce less sludge [1–3].

The hydrolysis reactions of aluminum species are very complicated, depending on water quality and coagulant type, which contribute to the formation of various aluminum hydrolyte species [4,5]. The ferron method has been employed extensively to categorize the aluminum hydrolyte species into the following three fractions on the basis of the kinetic differences between the reactions of the aluminum species and the ferron reagent: monomeric species, fast-reacting polymeric species, and slow-reacting colloidal species, denoted as  $\text{Al}_a$ ,  $\text{Al}_b$ , and  $\text{Al}_c$ , respectively [6].  $\text{Al}_a$  is composed mainly of monomeric species such as  $\text{Al}^{3+}$ ,  $\text{Al}(\text{OH})^{2+}$ , and  $\text{Al}(\text{OH})_2^+$ ; dimeric and trimeric species such as  $[\text{Al}_2(\text{OH})_2]^{4+}$  and  $[\text{Al}_3(\text{OH})_4]^{5+}$  are also classified as  $\text{Al}_a$  [1].  $\text{Al}_b$  is the intermediate polymeric species, and many researchers have suggested that  $\text{Al}_b$  could include the  $\text{Al}_{13}$  species  $[\text{AlO}_4\text{Al}_{12}(\text{OH})_{24}(\text{H}_2\text{O})_{12}]^{7+}$ , which was identified by using liquid  $^{27}\text{Al}$  nuclear magnetic resonance (NMR) spectroscopy [1,4,7].  $\text{Al}_c$  is the large polymer or colloidal species, and the  $\text{Al}_{30}$  species  $[\text{Al}_{30}\text{O}_8(\text{OH})_{56}(\text{H}_2\text{O})_{24}]^{18+}$ , which was also identified by using liquid  $^{27}\text{Al}$  NMR spectroscopy, is classified into  $\text{Al}_c$  [7].

The distribution of aluminum hydrolyte species in PACls and the formation of specific aluminum hydrolyte species such as  $\text{Al}_{13}$  and  $\text{Al}_{30}$  might be controlled by basicity ( $[\text{OH}^-]/[\text{Al}^{3+}]$ ), aluminum concentration, rate and mode of neutralization, and reaction temperature and time during PACl preparation [7–9]. In addition, the aluminum species originally contained in the PACls or formed *in situ* during the coagulation process play an important role in determining the behavior and efficiency of coagulants [4,5]. For these reasons, many researchers have paid particular attention to the effects of aluminum species on coagulation efficiency. For example, Yan *et al.* [1] reported that the removal efficiencies of turbidity and dissolved organic carbon (DOC) correlate well with the  $\text{Al}_c$  content and  $\text{Al}_b$  content in PACls, respectively, whereas both  $\text{Al}_b$  and  $\text{Al}_c$  content influence the removal of ultraviolet absorbance at 254 nm (UV254, an indicator of natural organic matter concentration). Our research group [10] and Duan *et al.* [11] reported that the residual aluminum concentration in treated water is closely related to the  $\text{Al}_a$  content in the PACl: low  $\text{Al}_a$  content in the PACl yields a low residual aluminum concentration. Moreover, among the identified aluminum hydrolyte species in PACls,  $\text{Al}_{13}$  and  $\text{Al}_{30}$  are believed to be effective coagulation species due to their strong charge neutralization capability and high structural stability [12]. In fact, Lin *et al.* [13] showed that DOC removal by PACl with a high  $\text{Al}_{13}$  content is superior to that by commercially available PACl with a low  $\text{Al}_{13}$  content at about pH 6, and that a lower dosage of high  $\text{Al}_{13}$  content PACl than of commercially available PACl is required for optimal DOC removal. Zhang *et al.* [2] reported that compared to  $\text{AlCl}_3$ , PACl with a high  $\text{Al}_{30}$  content exhibits greater UV254 removal efficiency and leads to a lower residual aluminum concentration across a broad pH range and a wide coagulant dosage range. In addition, Hu *et al.* [4] showed the value of PACl with a high  $\text{Al}_{13}$  content for the removal of arsenic (As) from As-spiked tap water over a broad pH range, and concluded that the removal efficiencies of As(V) correlated with the amount of  $\text{Al}_{13}$  species present. Moreover, Mertens *et al.* [14] reported that PACl with a high  $\text{Al}_{30}$  content contributed to the efficient removal of As(III) and As(V) from As-contaminated groundwater at pH 7–8.

Human enteric viruses, which are frequently present in contaminated drinking water sources and do not settle from suspension by gravity, can also be removed by the coagulation process with PACl [15]. However, the relationship between the removal efficiencies of human enteric viruses and the aluminum hydrolyte species in PACl remains unclear, and the role of  $\text{Al}_{13}$  and  $\text{Al}_{30}$  species in the

removal of human enteric viruses has not been investigated. We recently reported that laboratory-prepared PACl with a high  $\text{Al}_c$  content removed bacteriophages (i.e., viruses that infect bacteria) more efficiently than did laboratory-prepared PACl with a high  $\text{Al}_b$  content or commercially available PACls, suggesting that the  $\text{Al}_{30}$  species in high  $\text{Al}_c$  content PACl probably play a major role in bacteriophage removal during the coagulation process [16]. Because the removal ratios of human enteric viruses including poliovirus were different to and smaller than that of bacteriophage MS2 in the coagulation process with aluminum-based coagulant [17], the effect of aluminum hydrolyte species on virus removal, and specifically the role of  $\text{Al}_{13}$  and  $\text{Al}_{30}$ , may also differ between human enteric virus and bacteriophage removal. In addition, because human enteric viruses have high resistance to free-chlorine disinfection compared with human enteric bacteria [18], and the increase in free-chlorine dosage needed for sufficient disinfection of human enteric viruses sometimes results in the formation of high levels of toxic disinfection by-products [19], improvements in the coagulation efficiency of virus removal are highly desired. Therefore, the identification of aluminum hydrolyte species in aluminum-based coagulants that efficiently remove human enteric viruses from drinking water would improve coagulation efficiency and support the development of novel aluminum-based coagulants for the prevention and control of waterborne disease caused by exposure to such viruses through drinking water.

Here, we conducted batch coagulation experiments to investigate the effect of aluminum hydrolyte species on the removal of human enteric viruses, specifically human enteroviruses, by comparing five coagulants with different distributions of aluminum hydrolyte species. In addition, we investigated the mechanism of human enterovirus removal during the coagulation processes by examining the coprecipitation of enteroviruses into growing aluminum hydroxide during charge neutralization and the absorption of enteroviruses onto preformed aluminum floc particles. Because poliovirus (PV) type 1 is commonly used as representative of human enteric viruses [17,20], and coxsackievirus (CV) B5 has high resistance to free-chlorine disinfection compared with other types of CVs (e.g., CV B3 and CV B4) and other human enteric viruses including PVs (types 1, 2 and 3), echoviruses (EV types 1 and 11), and adenoviruses (AdV types 40 and 41) [21,22], we chose these two human enteroviruses for use in our study.

## 2. Materials and methods

### 2.1. Source water and coagulants

Lake water and river water were sampled from Lake Imbana in Chiba, Japan, and the Toyohira River in Sapporo, Japan, on 17 November 2014 (water quality data are shown in Table 1). The source water samples were stored at 4 °C until use, and the temperature was adjusted to 20 °C prior to use.

Five aluminum-based coagulants were used for the coagulation experiments (Table 2).  $\text{AlCl}_3$  solution was prepared by dilution of reagent-grade aluminum(III) chloride hexahydrate ( $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ , Wako Pure Chemical Industries, Osaka, Japan) dissolved in Milli-Q water (Milli-Q Advantage, Millipore Corp., Billerica, MA, USA).

**Table 1**  
Water quality data for the source water samples.

	Lake water	River water
pH	8.6	7.4
Turbidity (NTU)	40.5	1.5
DOC (mg/L)	2.2	0.9
UV260 ( $\text{cm}^{-1}$ )	0.052	0.024
Alkalinity (mg- $\text{CaCO}_3/\text{L}$ )	84.1	17.5

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