Aluminum speciation in drinking water distribution system: A case study in northeastern China

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Abstract−This study aimed to investigate the variation of aluminum species in a drinking water distribution system in a city in northeastern China. The aluminum species were determined by fluorometric methods. Results showed that suspended aluminum (Sus-Al) was the major species in the drinking water supplied by plant B and accounted for about 42% of the total aluminum (Tol-Al). The concentrations of Sus-Al and Tol-Al could be controlled effectively by introducing reservoir water. In the water source switching process, the water quality variation led to the suddenly release of Sus-Al, especially in a cast iron pipeline that had been in service for more than 30 years, but the soluble aluminum varied little. In the plant A service areas, the average concentrations of the inorganic monomeric aluminum (IM-Al), monomeric aluminum (Mon-Al), and soluble aluminum (Sol-Al) were 0.008 mg L^{-1}, 0.03 mg L^{-1}, and 0.04 mg L^{-1}, respectively, and their concentrations in the plant B service areas were higher. The pH and fluoride were the major parameters affecting the soluble aluminum speciation. With a solution pH of 6.5-7.5 and fluoride below 0.3 mg L^{-1}, the Sol-Al could be controlled within 0.1 mg L^{-1}. Water quality regulation and terminal filtration were suggested for residual aluminum control.

Key words: Aluminum Speciation, Drinking Water, Fluorometric Method, Water Quality

INTRODUCTION

With modern industrial development, acid precipitation is becoming more and more common. This has led to a large amount of aluminum dissolving from soil into natural waters [1,2]. Meanwhile, aluminum salts are widely used as coagulants in the drinking water treatment process. Although they are effective for removing turbidity, Al-based coagulants may result in a high Al concentration in the treated water. Driscoll and Letterman [3] reported that approximately 11% of the aluminum input remained in the finished water [3]. All of the surveys on residual aluminum made in China, the United States, and Europe also showed that its concentration is usually higher in drinking water coagulated with aluminum salts than that with other coagulants [4-7].

A high aluminum concentration in drinking water can cause high turbidity [8,9], pipe deposition [10-13], and disinfection inhibition [6]. Health problems such as neurological disorders are associated with the presence of aluminum in drinking water. Al^{3+} is known to be severely neurotoxic by promoting the formation of neurofibrillary tangles and senile plaques in the brain. When it reacts with other matter, its toxicity can be reduced significantly. The reaction between Al^{3+} and silicic acid could reduce its bioavailability effectively by inhibiting the gastro-intestinal adsorption process [14]. Most organic compounded aluminum also shows weak biotoxicity. Therefore, any evaluation on the effects of aluminum on drinking water distribution and human health should consider its speciation.

For dilute solutions above a pH of 3.0, the main aluminum species

is Al^{3+}, and in the pH range of 5.0-7.0, it coexists with Al(OH)$_2^+$, Al(OH)$_3^0$(aq), and Al(OH)$_4^-$ in varying proportions [15]. For solutions above a pH of 7.0, aluminum mainly exists as Al(OH)$_4^-$. Van Benschoten and Edzwald [16] found that the concentration of Sol-Al would increase notably in a solution with a high content of fluoride [16]. The existence of organic acids could restrain aluminum hydrolysis by complexation [17]. Exley et al. [18] also found that silicic acids could also restrain its hydrolysis process [18]. Also, cations such as K$^+$, Na$^+$, Fe^{3+}, Ca^{2+}, Mn^{2+}, and Mg^{2+} could affect its solubility by competition and co-precipitation [19].

City M is in northeastern China. The annual mean water temperature is about 8.0 °C. Survey results showed that residual aluminum is high in drinking water, and 90% of the samples with total aluminum concentrations are above 0.2 mg L^{-1} [4]. The Songhua River was the major water source before 2007. To improve the drinking water quality, the Mopan Reservoir began to serve as its secondary water source after 2007. Meanwhile, a new water purification plant was also built. The use of the Mopan Reservoir led to a significant change in the water quality. Contaminants that had accumulated in the pipe sediments might have been released to the drinking water. The purpose of the present study was to investigate (a) the variation law of aluminum speciation in water source switching process, and (b) the effects of water quality, flow conditions, and pipe material on the aluminum species concentrations.

MATERIAL AND METHODS

1. Basic Information on the Drinking Water Supply System of the City

The major water plants in city M were fed by the Songhua River

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Table 1. Drinking water plants used by the city

Plant	Coagulant type	Capacity ($m^3\ d^{-1}$)	Source	Purpose
Plant 1	Alum	100000	Songhua river	Industry water
Plant 4	PACl	77000	Songhua river	Drinking water
Plant 5	-	10000	Underground water	Drinking water
Plant 6	-	20000	Underground water	Industry water
Plant 7	-	70000	Underground water	Drinking water
Plant A	PAFC	450000	Reservoir water	Drinking water
Plant B	PACl	650000	Songhua river	Drinking water

and the Mopan Reservoir (Table 1). Before 2007, the total water supply capacity was 1,380,000 $m^3\ d^{-1}$. Plant 1, plant B, and plant 4 (fed by the Songhua River) supplied 827,000 $m^3\ d^{-1}$. Plant 5, plant 6, and plant 7 (fed by underground water) supplied 100,000 $m^3\ d^{-1}$. In the first stage of the water source switching, from June 2007 to February 2008, the drinking water supplied by plant A was 225,000 $m^3\ d^{-1}$. In the second stage, since March 2008, the Mopan Reservoir has supplied 450,000 $m^3\ d^{-1}$. In this process, part of the areas supplied by plant B would be replaced by plant A, and most of the underground water plants would be shut down gradually.

Plant A, which was located on the south side of the city, treated drinking water by a traditional process with polymeric aluminum ferric chloride (PAFC) coagulation. Plant B also treated drinking water by a traditional process, but poly aluminum chloride (PACl) was used as the coagulant agent, instead of PAFC (Table 1). As the reservoir water quality was better, the drinking water supplied by plant A was much better than that supplied by plant B (Table 2). To remove the organic matter in the raw water, powder activated carbon (PAC) adsorption was applied in plant B before coagulation.
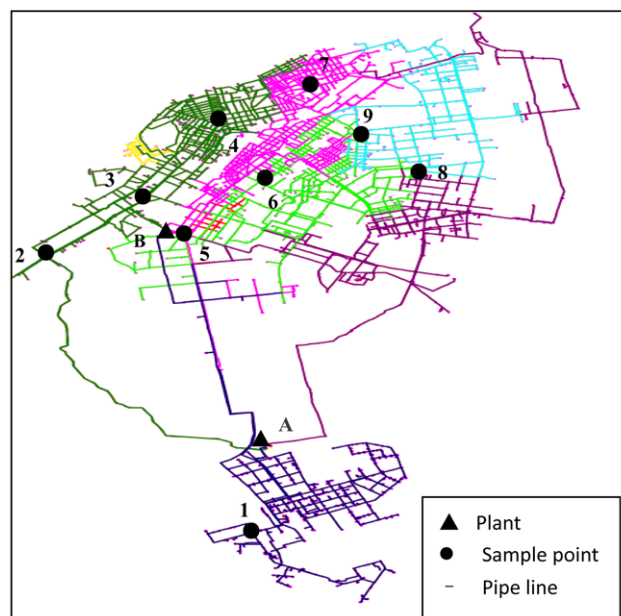
The average service time of the water distribution system was not long. Pipes laid in 1930-1969, 1970-1989, and 1990-2008 occupied 12%, 19.0%, and 68.9% of the system, respectively. Most of the pipe material was cast iron, which accounted for 95% of the total length of the system. Steel pipe, stainless steel pipe, glass fiber reinforced plastic pipe, and polyethylene pipe were also used in the system.

2. Sample Points Information

The drinking water distribution system in city M is shown in Fig. 1.

Fig. 1. The drinking water distribution system and the selected sample points in city M.

Before 2007, most of the areas were supplied by plant B. In the first stage of the water source switching, areas with the bottle green, navy blue, and purple pipelines were supplied by plant A, and the areas with the red, pink, reseda, and nattierblue colors were supplied by plant B. Since March 2008, the nattier blue area was also supplied by plant A in its second stage of operation.

Considering the service area variation during the water source switching process, nine sample points were selected. Points 1, 2, 3, and 4 were located in the plant A service area. Points 5, 6, and 7 were in the plant B service area. Before March 2008, points 8 and 9 were in the plant B service area, and after that, they were supplied by plant A. The diameter, material, and service time of the pipeline at each sample point are listed in Table 3.

To investigate the effects of the water source switching, flow condition, and water quality on aluminum speciation, samples were collected before and after the second stage of the water source switching, on 11th-13th November 2007, 13th-15th March 2008, and 11th-

Table 2. Plant A and plant B raw water and treated water quality

Item	Raw water		Treated water	
	Plant A	Plant B	Plant A	Plant B
Aluminum ($mg\ L^{-1}$)	0.08	0.12	0.09	0.191
pH	7.0	7.5	6.81	7.32
Calcium ($mg\ L^{-1}$)	23	35	13.52	23.67
Magnesium ($mg\ L^{-1}$)	5.7	11	1.53	5.81
Soluble silicic acid ($mg\ L^{-1}$)	5.1	5.3	2.15	1.93
Fluoride ($mg\ L^{-1}$)	0.12	4.5	0.08	2.10
Alkalinity ($mg\ L^{-1}$)	26	80	28.18	62.2
NPOC ($mg\ L^{-1}$)	3.0	5.0	2.33	2.56
Conductivity ($\mu s\ cm^{-1}$)	103	265	85.2	186.7

Table 3. The pipe material, diameter, and service time at each sample point

Sample	Pipe material	Pipe diameter (mm)	Service time (year)
1	Cast iron	600	10
2	Stainless steel	1800	2
3	Cast iron	500	15
4	Cast iron	700	20
5	Cast iron	400	51
6	Cast iron	900	60
7	Cast iron	500	11
8	Cast iron	900	19
9	Cast iron	1000	15

12th July 2008. Based on the drinking water demand variation of the city, samples were collected twice during the day, at 8:00-9:00 a.m. and 5:00-6:00 p.m.

3. Analysis Methods

To determine the concentration of the aluminum species, an approximately 30 mL water sample was needed. One subsample was digested at pH 1.0 with reagent grade HNO_3 for 24 h to analyze the concentration of Tol-Al. The other subsample was filtered through 0.22 μm polycarbonate filters. Then, one part of the filtrate was digested at pH 1.0 with HNO_3 for 24 h to analyze the concentration of Sol-Al. The other part of the filtrate was used to determine the concentrations of IM-Al and Mon-Al. The concentrations of OM-Al, Pol-Al, and Sus-Al were obtained by the following equations: [OM-Al]=[Mon-Al]−[IM-Al], [Pol-Al]=[Sol-Al]−[Mon-Al], and [Sus-Al]=[Tol-Al]−[Sol-Al].

The aluminum concentration was measured by applying the fluorometric method [20]. The minimum detection limit of the method ranged from 0.006 to 0.015 mg L^{-1}, where accuracy (% recovery) was 91.7-106.1%, the precision (relative standard deviation, n=6) was 0.68%, and the linearity range was 0.027-0.27 mg L^{-1}. Aluminum species were determined by a fluorometer (Model F-2500, Hitachi High-Technologies Corporation).

The concentrations of calcium, magnesium, iron, and manganese were determined by applying an IRIS intrepid-II ICP (Thermo Elemental, USA). Parameters, such as temperature, pH and chlorine, were determined in situ. Parameters including turbidity, conductivity, alkalinity, fluoride, phosphate, and silicic acid were measured according to the standard methods of water and wastewater analysis of China [21]. The residual chlorine was measured applying a chlorine meter (ORION AQ2070, Thermo Electron).

RESULTS AND DISCUSSION

1. Aluminum Speciation in the Drinking Water Distribution System

Before the water source switching, all of the aluminum species were less than 0.2 mg L^{-1}. The Tol-Al average concentrations were 0.047 mg L^{-1} and 0.127 mg L^{-1} at sample points fed by Plant A and plant B, respectively (Fig. 2(a)). The Sus-Al was the major aluminum species in the drinking water supplied by plant B and accounted for about 42% of the Tol-Al, but in areas supplied by plant A, the value was less (25%). Conversely, the Sol-Al concentration (including IM-Al, OM-Al, and Pol-Al) varied little at the different sample points and was about 0.03 mg L^{-1}. The IM-Al was considered to be the most toxic species in drinking water. Its concentrations were 0.008 mg L^{-1} and 0.02 mg L^{-1} in the drinking water fed by plant A and plant B, respectively, and accounted for about 17% of the aluminum at both locations. The concentrations of all the aluminum species matched the national standards of China, and the aluminum water toxicity was not high.

After the water source switching, water samples were collected in July 2008. The experimental results are shown in Fig. 2(b). Compared with Fig. 2(a), the Sus-Al decreased notably at most of the sample points. However, the concentrations of IM-Al, OM-Al, Mon-Al, and Sol-Al all varied little. In the plant A service areas, the Mon-Al and Sol-Al were 0.03 and 0.04 mg L^{-1}, respectively. In the plant B service areas, their concentrations were 0.05 and 0.06 mg L^{-1},
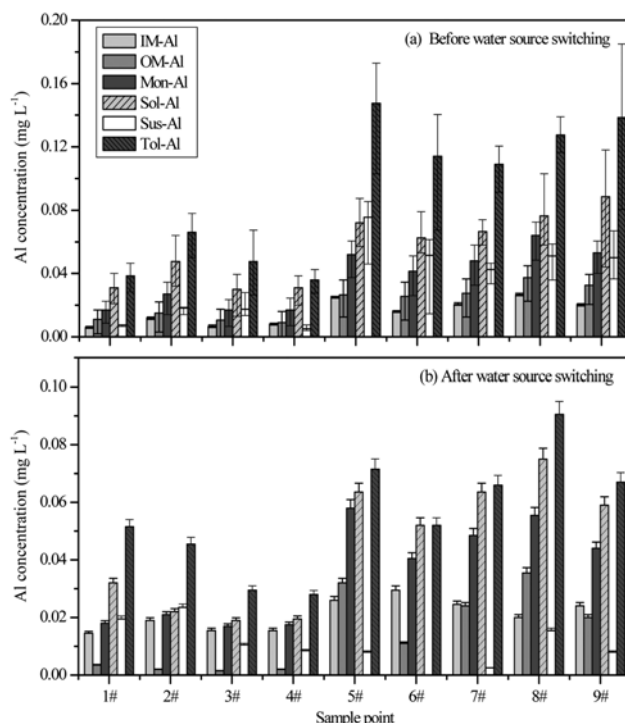

Fig. 2. Aluminum speciation in the drinking water before and after the water source switching.

respectively. The aluminum concentrations in the plant A service areas were still lower than those in the plant B service areas, which were connected with its water quality after the water source switching.

Survey results also showed that the Sus-Al accounted for a high proportion of the Tol-Al, especially in the drinking water before the water source switching. It was mainly because of the hydraulic conditions in the pipeline. However, as the drinking water distribution system was complicated, the water flow in it was generally difficult to control. The Sus-Al was filterable by microfiltration membrane (see the Sus-Al analysis methods). Early study results also showed that nearly all of the Sus-Al could be removed by a microfiltration system [22]. Terminal microfiltration treatment was suggested, which would be an efficient way to control the Sus-Al. Also, other suspended pollutants such as $Fe(OH)_3(s)$, $CaCO_3(s)$, and others would also be removed.

2. The Effects of the Water Source Switching on Aluminum Speciation

The IM-Al, OM-Al, Mon-Al, and Sol-Al concentrations varied little with the sample time, but the Sus-Al varied in a large range, especially in the water source switching process. Thus, the variation in the Tol-Al was mainly attributable to Sus-Al. The stability of the aluminum-containing sediments would be the key factor affecting the Tol-Al. All of the factors (water flow, water quality, pipe material, and pipe service time) affecting the pipe sediment stability could lead to the flocculation of the Sus-Al and Tol-Al. The pipeline at Point 2, the main pipe of plant A, was laid in 2007. Both its material and the water flow in it were in a steady state, and thus led to the Sus-Al and Tol-Al concentrations in it also varying little in the water source switching process (Fig. 3(a)).

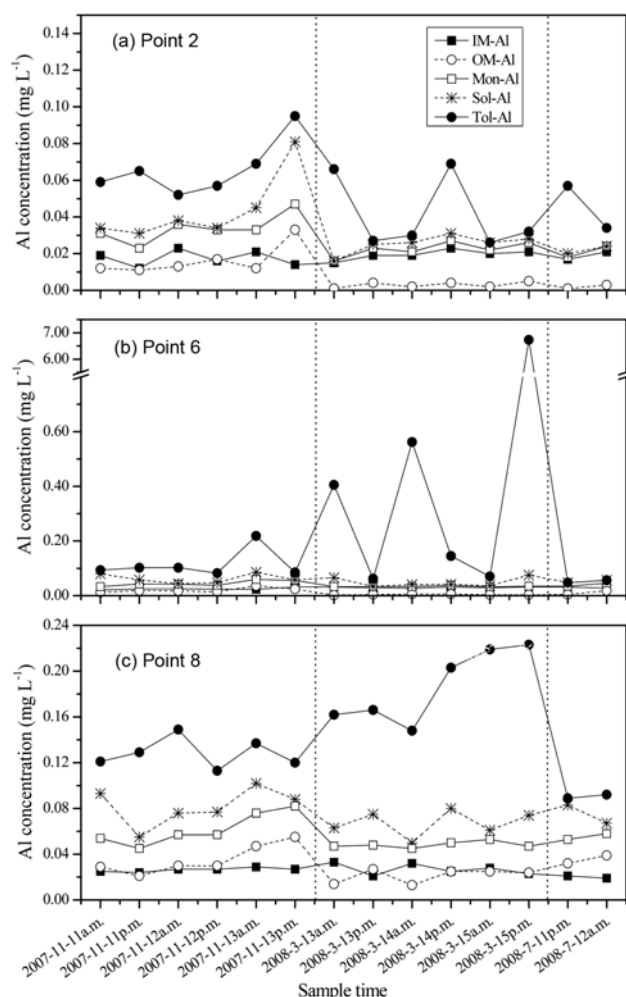As opposed to point 2, the pipeline at point 6 had been in service

Fig. 3. Aluminum species variation with the sample time at Points 2, 6, and 8.

for 60 years, and the pipe material was cast iron. Corrosion and sediment accumulation were serious. Sudden changes in the flow and water quality would affect the sediment stability and lead to the Sus-
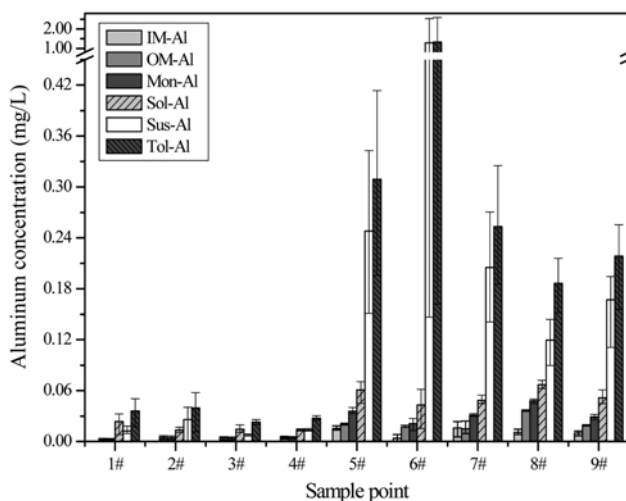

Fig. 4. Aluminum speciation in water source switching process.

Al and Tol-Al concentrations to increase notably. The Sus-Al ranged between 0.02 mg L^{-1} and 0.10 mg L^{-1} before and after the water source switching. However, the value was much higher, ranging from 0.03 mg L^{-1} to 6.6 mg L^{-1} during the switching process (Fig. 4). The Sus-Al in the morning peak periods was usually higher than that in the evening peak period, which might have a relationship with the difference in water flow conditions.

In the early morning hours, the water flow was low, leading to pipe erosion and solids precipitation. However, the water velocity and direction would both change suddenly in the morning peak periods, leading to the Sus-Al concentration increase on the effect of flushing. After about 10 hr of operation, the sediments in the pipe reached a relative steady state in the evening peak periods. The Sus-Al concentrations also varied little in a day, at about 0.029 mg L^{-1} and 0.046 mg L^{-1} in areas supplied by plant A and plant B, respectively.

Point 8 was on the boundary of the plant A and plant B service area. The pipeline was made of cast iron and had been in service for about 20 years. The water source switching also had notable effects on the Sus-Al and Tol-Al in it. However, the flocculation range was much smaller than that at point 6, indicating that the pipe corrosion and sedimentation were not serious. Meanwhile, the water source switching notably affected its water quality. However, the soluble aluminum species also varied little at different sample points.

3. The Effects of the Water Quality on Aluminum Speciation

The water quality parameters concerned in the experiment included temperature, pH, chlorine, turbidity, alkalinity, TOC, Ca^{2+}, Mg^{2+}, Fe^{3+}, Mn^{2+}, silicic acid, orthophosphate, and fluoride. Mn^{2+} and phosphate were not detected in the drinking water. Statistical results showed that besides the pH and fluoride, the chlorine and TOC could also affect the Mon-Al concentration. The IM-Al concentration correlated with the pH, fluoride, chlorine, and water temperature positively, and it correlated with TOC negatively (Table 4). However, turbidity, alkalinity, and cations had little effect on it. The existence of TOC could promote the conversion of the IM-Al to OM-Al, but chlorine would resist this process by destroying the structure of the organic matter. Compared with IM-Al, the Pol-Al concentration positively correlated with the TOC, indicating that the existence of organic matter might promote the production of Pol-Al [23]. Further research was still needed to verify the conclusion.

Based on the Pearson correlation coefficient, the effects of the water quality on aluminum speciation were also evaluated (Table 4). For the IM-Al, OM-Al, Mon-Al, and Sol-Al, pH and fluoride were the most important parameters. TOC and turbidity were the parameters that affected the Pol-Al and Sus-Al concentrations the most. In areas supplied by plant A, the average pH and fluoride concentration was 6.81 and 0.11 mg L^{-1}, respectively, and the values in the plant B service areas were 7.35 and 0.20 mg L^{-1}, respectively. The content difference in the pH and fluoride led to the Sol-Al concentration in the drinking water fed by plant A lower than that fed by plant B. Meanwhile, As the water quality in plant A was good for Sol-Al control, its concentration varied little in the water source switching process at point 8, which was consistent with the conclusion made in section 2.2.

Comparing the major parameters (including pH, fluoride, turbidity, and TOC) with the corresponding aluminum species, it can be concluded that when the solution pH varied between 6.5 and 7.0

Table 4. Relation analysis between water quality parameters and aluminum species (the total sample number N=135)

		Temperature	pH	Chlorine	Turbidity	Alkaline	TOC	Ca^{2+}	Mg^{2+}	Fe^{3+}	Silicate	F^-
IM-Al	R	0.309**	0.695**	0.353**	0.003	−0.028	−0.253**	−0.058	0.112	−0.290*	−0.180	0.358**
	P	0.001	0.000	0.000	0.975	0.766	0.006	0.536	0.227	0.020	0.058	0.000
OM-Al	R	0.239**	0.715**	−0.003	0.034	0.510**	0.142	0.482	0.606	0.069	0.423**	0.680**
	P	0.009	0.000	0.972	0.719	0.000	0.127	0.060	0.051	0.457	0.000	0.000
Mon-Al	R	0.307**	0.811**	0.164	0.024	0.333**	−0.215*	0.300*	0.464*	−0.088	0.204*	0.629**
	P	0.001	0.000	0.076	0.795	0.000	0.020	0.050	0.046	0.343	0.027	0.000
Pol-Al	R	−0.045	0.206*	0.148	0.114	−0.030	0.258**	−0.093	−0.033	−0.034	−0.073	0.157
	P	0.633	0.026	0.111	0.223	0.751	0.005	0.318	0.721	0.720	0.433	0.091
Sol-Al	R	0.191*	0.707**	0.211*	0.088	0.217*	0.316**	0.153	0.308*	−0.085	0.097	0.546**
	P	0.039	0.000	0.022	0.343	0.019	0.001	0.099	0.040	0.364	0.296	0.000
Sus-Al	R	−0.162	0.063	0.071	0.946**	0.000	0.414**	−0.034	0.001	0.114	−0.029	0.182*
	P	0.082	0.499	0.445	0.000	0.996	0.000	0.713	0.992	0.220	0.756	0.050
Tol-Al	R	−0.152	0.094	0.080	0.944**	0.009	0.425**	−0.027	0.015	0.110	−0.025	0.205*
	P	0.102	0.312	0.390	0.000	0.922	0.000	0.770	0.875	0.239	0.793	0.026

R: Pearson correlation index; P significance; * Correlation is significant at 0.05 level (2-tailed); ** Correlation is significant at 0.01 level (2-tailed)
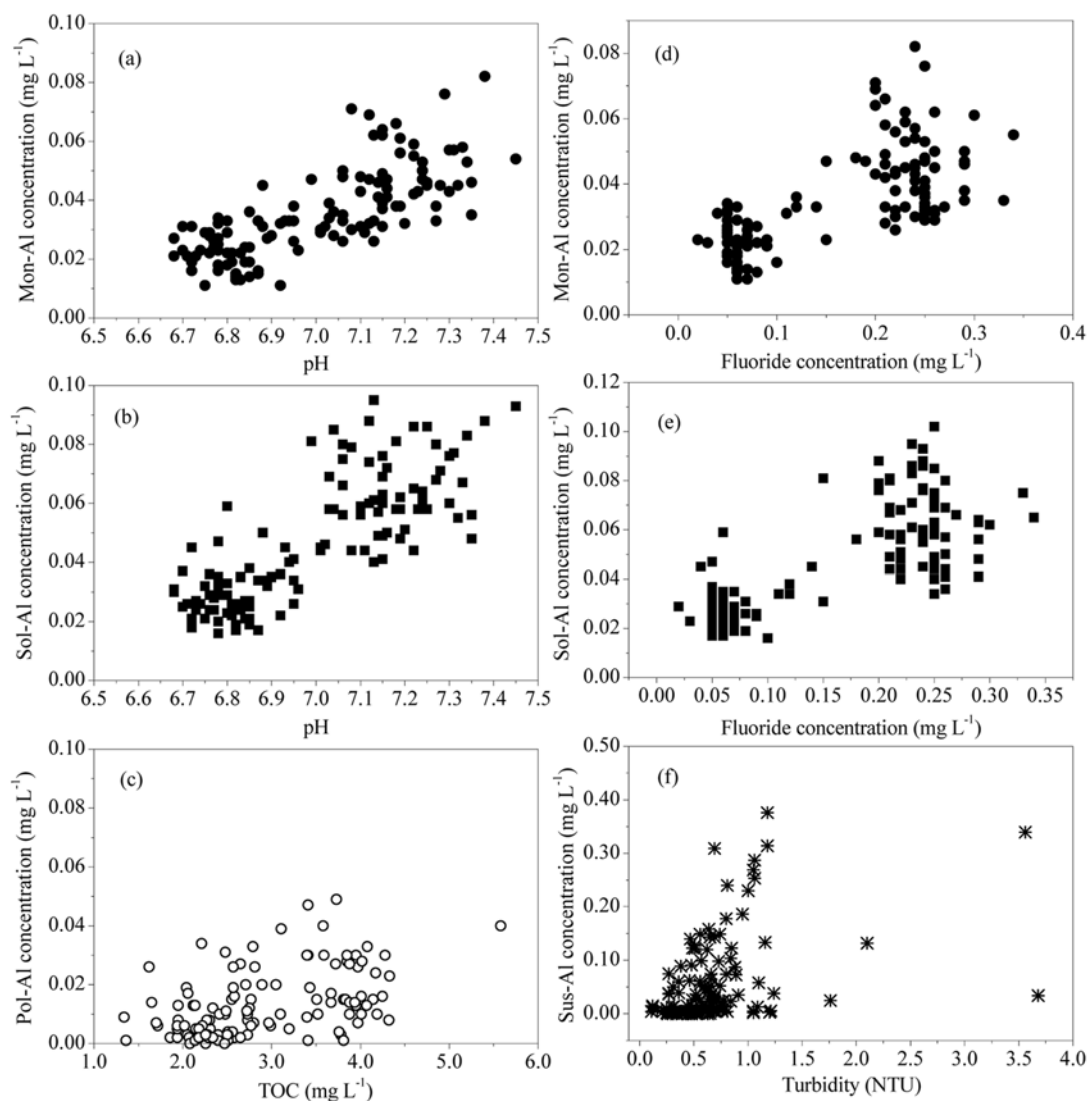
Fig. 5. Effects of the pH, fluoride, TOC, and turbidity on aluminum speciation in the drinking water.

and the fluoride was below 0.1 mg L^{-1}, the Sol-Al concentration could be controlled within 0.05 mg L^{-1} (European standard for aluminum control), as shown in Fig. 5(b) and 5(e). In a solution with a pH of 7.0-7.5 and fluoride of 0.1-0.3 mg L^{-1}, the Sol-Al could be controlled within 0.10 mg L^{-1}. The Mon-Al presented similar variation with the pH and fluoride concentration (Fig. 5(a) and 5(d)). When the soluble organic matter concentration was below 2.5 mg L^{-1} (calculated as TOC), the Pol-Al could be controlled within 0.04 mg L^{-1}. Accordingly, water quality adjusting that is usually conducted in the drinking water treatment process would be an efficient method for Sol-Al control.

CONCLUSION

Before the water source switching, all of the aluminum species were less than 0.2 mg L^{-1}. The Sus-Al was the major species in the drinking water supplied by plant B, accounting for about 42% of the Tol-Al, but in areas supplied by plant A, the value was 25%. The IM-Al concentrations were 0.008 mg L^{-1} and 0.02 mg L^{-1} in the drinking water fed by plant A and plant B, respectively. By introducing reservoir water, the Sus-Al and Tol-Al were controlled effectively in city M. However, during the water source switching process, the water quality variation led to the sudden release of Sus-Al. Meanwhile, the hydraulic conditions also affected the Sus-Al concentration, especially in pipelines with long service times. Compared with Sus-Al, the concentrations of both Mon-Al and Sol-Al were low and varied little.

The stability of the aluminum-containing sediments was the major factor affecting the Sus-Al concentration. Factors such as the water flow, water quality, pipe material, and pipe service time, which affected pipe sediment stability, could lead to the flocculation of the Sus-Al and Tol-Al. The correlation analysis results also showed that the Sus-Al concentration was positively related to the solution turbidity.

The soluble aluminum species were mainly affected by the water quality. The statistical analysis results showed that the pH and fluoride were the major factors affecting the IM-Al, OM-Al, Mon-Al, and Sol-Al concentrations. Besides the pH and fluoride, chlorine and TOC could also affect the aluminum speciation. The existence of TOC would promote the conversion of the IM-Al to OM-Al, but the chlorine would resist this process by destroying the structure of the organic matter. The existence of soluble organic matter might promote the production of Pol-Al, but the reaction mechanism is still not clear.

By maintaining the solution pH at 6.5-7.5 and the fluoride below 0.3 mg L^{-1}, the Sol-Al concentration could be controlled within 0.10 mg L^{-1}. Because the hydraulic conditions are generally difficult to control, water quality regulations in the water treatment process and terminal filtration were suggested to remove the Sol-Al and Sus-Al from the drinking water.

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