



Coagulation of methylated arsenic from drinking water: Influence of methyl substitution



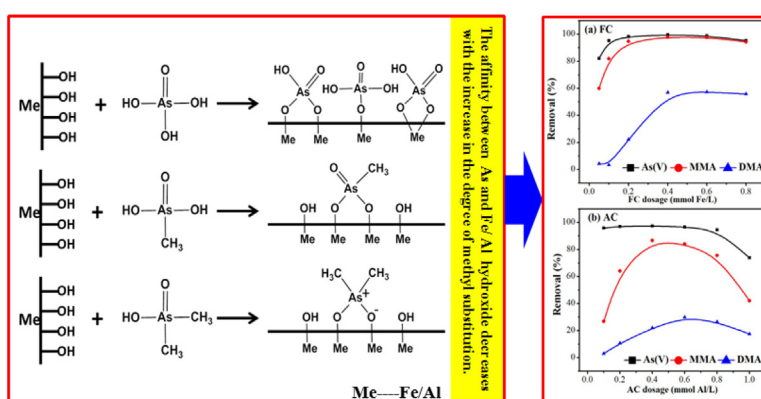
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HIGHLIGHTS

- FeCl_3 was more efficient than Al salts in methylated As removal by coagulation.
- As removal was negatively correlated with the degree of methyl substitution.
- Adsorption on Fe/Al hydroxide flocs was major coagulation mechanism of methylated As.
- As–O group of methylated As substituted O–H group of Fe/Al hydroxide to form complex.

GRAPHICAL ABSTRACT



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ABSTRACT

Methylated arsenic can be found in virtually all earth surface environments. So far, however, little information has been collected regarding their removal by coagulation. In this study, the removal of monomethylarsenate (MMA) and dimethylarsenate (DMA) from drinking water by coagulation was investigated from the viewpoint of methyl substitution. Results indicated that FeCl_3 was more efficient than AlCl_3 and polyaluminum chloride (PACl) in methylated As removal. For the initial arsenic concentration of $200 \mu\text{g/L}$, an FeCl_3 dosage of 0.2 mmol Fe/L was sufficient to attain about 95% removal of MMA, while a dosage of 0.6 mmol Fe/L achieved about 57% removal of DMA. Arsenic removal efficiency was negatively correlated with the degree of methyl substitution. With the increase in methyl group number, the quantity of negatively charged arsenic species decreased and molecular size increased, leading to the decrease of methylated As removal by coagulation. Adsorption on preformed hydroxide flocs was the major mechanism during coagulation. Both FTIR and XPS results indicated that the As–O group of As might substitute the O–H group of Fe/Al hydroxide to form a Fe/Al–O–As complex. Furthermore, the use of traditional oxidants and coagulation aids exhibited limited help for improving coagulation removal of DMA.

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1. Introduction

Arsenic (As) is of considerable environmental concern due to its toxicity [1], with drinking water, the main source of As exposure for

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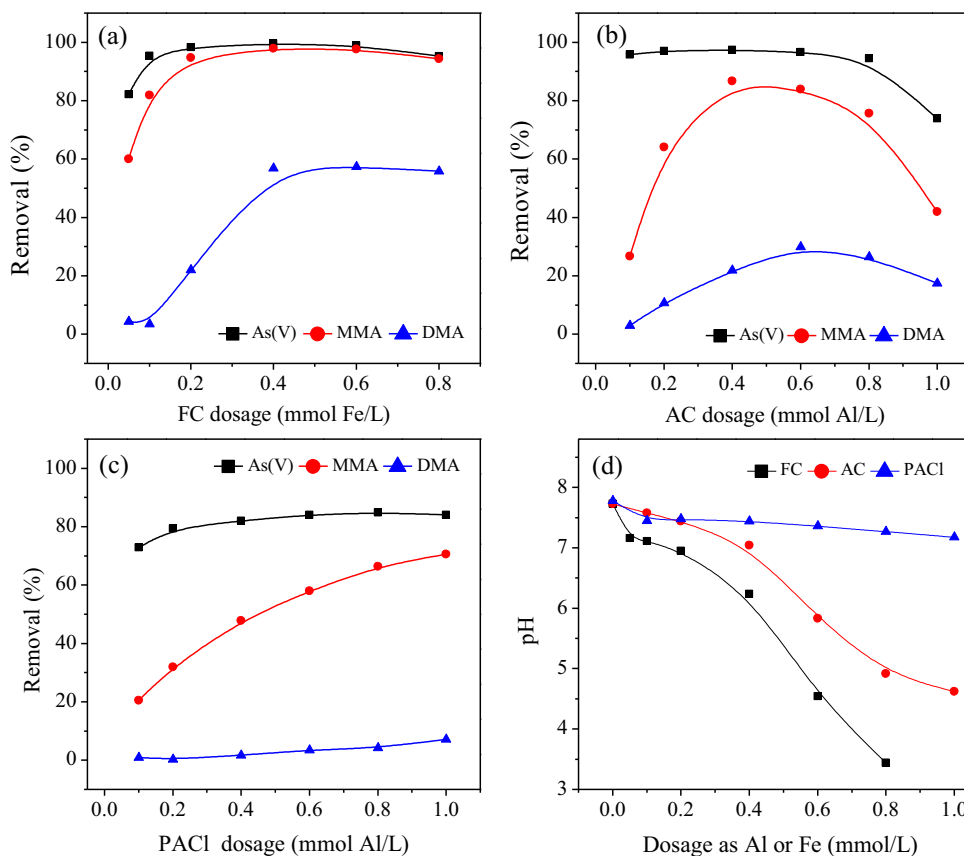


Fig. 1. Effect of coagulant dose on As(V), MMA, DMA removal and treated water pH (a) FC coagulation (b) AC coagulation (c) PACl coagulation (d) treated water pH. Initial As concentration was 200 $\mu\text{g As/L}$.

humans [2]. Although inorganic As(V) and As(III) are the most common arsenic species in the environment, organic forms of As can also be found in nature, typically occurring as monomethylarsenate (MMA) and dimethylarsenate (DMA) [3]. Previous research reported that 19.4% and 5.0% of total As occurred as DMA and MMA, respectively, in a Californian lake [4], and 24.1% of total As occurred as DMA (3.5 $\mu\text{g As/L}$) in a southwest Spanish river [5], where methylated As may threaten the safety of drinking water. In addition, MMA and DMA concentrations were found to be 35.4–233.9 $\mu\text{g As/L}$ and 3.5–13.1 $\mu\text{g As/L}$ from a source of drinking water in Inner Mongolian, respectively [6]. Some bacteria and fungi can produce MMA and DMA through the methylation of inorganic As [7]. Furthermore, MMA and DMA can be introduced into the environment through agricultural activities [8] with both used as herbicides and pesticides over the past several decades [7,9]. Although inorganic As has long been considered more toxic than organic As [10], some methylated As, such as MMA(III), is reportedly much more toxic than inorganic As [11]. Thus, the reduction of MMA and DMA can produce very toxic MMA(III) and DMA(III), respectively.

As a result of the widespread distribution and toxicity of methylated As in the environment, research on methylated As removal from water has grown in importance. The adsorption of MMA and DMA on several minerals has been studied previously [9,12–16]. Results found that adsorption of DMA on goethite and ferrihydrite was reportedly low compared with As(V) and MMA [13], adsorption of As(V) on aluminum oxide reached 100% within 5 min, while 78% and 15% of MMA and DMA were adsorbed, respectively [15], and adsorption of MMA on nanocrystalline TiO_2 achieved 100% at pH 7.5, with DMA removal reaching 65% at pH 5.5 [17].

The number of methyl groups of As(V), MMA and DMA is 0, 1 and 2, respectively. Variation in methyl substitution degree can lead to

remarkable differences in the adsorption behavior of As onto Fe/Al oxides. Coagulation by Fe- and Al-based salts have been used as the most practical choice for As removal from drinking water [18,19]. Many studies have investigated the coagulation removal of inorganic As [20–23]. However, only limited information is available on the removal of organic As by coagulation [24], and the removal of methylated As from drinking water by coagulation has not yet been studied. Furthermore, the influence of methyl substitution on As coagulation by Fe/Al-based salts is still unclear.

We investigated the coagulation efficiency and behavior of methylated As in drinking water from the viewpoint of methyl substitution. The removal of As(V), MMA and DMA was examined under various conditions including coagulant type, coagulant dose, and water pH. The mechanisms of methylated As removal by Al/Fe-based coagulants were also studied through particular coagulation/adsorption experiments. The interaction of flocs with As(V), MMA and DMA was characterized using Fourier transform infrared spectrometry (FTIR) and X-ray photoelectron spectrometry (XPS). We also evaluated the addition of common oxidants and coagulant aids to enhance methylated As removal by coagulation.

2. Material and methods

2.1. Materials and analysis methods

All chemical solutions used in this study were prepared using de-ionized water and analytical (or better) grade chemicals. FeCl_3 (FC), AlCl_3 (AC) and polyaluminum chloride (PACl) were used as coagulants. PACl was prepared by an electrochemical process [25]. The predominant Al species of PACl was Al_{13} polymer, which accounted for 86.3% of total Al

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