Kinetics and mechanistic aspects of As(III) oxidation by aqueous chlorine, chloramines, and ozone: Relevance to drinking water treatment

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Abstract: Kinetics and mechanisms of As(III) oxidation by free available chlorine (FAC—the sum of HOCl and OCl⁻), ozone (O₃), and monochloramine (NH₂Cl) were investigated in buffered reagent solutions. Each reaction was found to be first order in oxidant and in As(III), with 1:1 stoichiometry. FAC-As(III) and O₃-As(III) reactions were extremely fast, with pH-dependent, apparent second-order rate constants, k_{app} of 2.6 (±0.1) × 10⁵ M⁻¹ s⁻¹ and 1.5 (±0.1) × 10⁶ M⁻¹ s⁻¹ at pH 7, whereas the NH₂Cl-As(III) reaction was relatively slow (k_{app} = 4.3 (±1.7) × 10⁻¹ M⁻¹ s⁻¹ at pH 7). Experiments conducted in real water samples spiked with 50 µg/L As(III) (6.7 × 10⁻⁷ M) showed that a 0.1 mg/L Cl₂ (1.4 × 10⁻⁶ M) dose as FAC was sufficient to achieve depletion of As(III) to <1 µg/L As(III) within 10 s of oxidant addition to waters containing negligible NH₃ concentrations and DOC concentrations <2 mg-C/L. Even in a water containing 1 mg-N/L (7.1 × 10⁻⁵ M) as NH₃, >75% As(III) oxidation could be achieved within 10 s of dosing 1-2 mg/L Cl₂ (1.4-2.8 × 10⁻⁵ M) as FAC. As(III) residuals remaining in NH₃-containing waters 10 s after dosing FAC were slowly oxidized (t½ = 4 h) in the presence of NH₂Cl formed by the FAC-NH₃ reaction. Ozonation was sufficient to yield >99% depletion of 50 µg/L As(III) within 10 s of dosing 0.25 mg/L O₃ (5.2 × 10⁻⁶ M) to real waters containing <2 mg-C/L of DOC, while 0.8 mg/L O₃ (1.7 × 10⁻⁵ M) was sufficient for a water containing 5.4 mg-C/L of DOC. NH₃ had negligible effect on the efficiency of As(III) oxidation by O₃, due to the slow kinetics of the O₃-NH₃ reaction at circumneutral pH. Time-resolved measurements of As(III) loss during chlorination. ?? 2006 American Chemical Society.

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