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 (O_3), and monochloramine (NH_2Cl) 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 O_3-As(III) reactions were extremely fast, with pH-dependent, apparent second-order rate constants, $k??_{app}$, of 2.6 (?0.1) ?? 10^5 M^{-1} s^{-1} and 1.5 (?0.1) ?? 10^6 M^{-1} s^{-1} at pH 7, whereas the NH_2Cl-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 ?g/L As(III) (6.7 ?? 10^{-7} M) showed that a 0.1 mg/L Cl_2 (1.4 ?? 10^{-6} M) dose as FAC was sufficient to achieve depletion of As(III) to <1 ?g/L As(III) within 10 s of oxidant addition to waters containing negligible NH_3 concentrations and DOC concentrations <2 mg-C/L. Even in a water containing 1 mg-N/L (7.1 ?? 10^{-5} M) as NH_3, >75% As(III) oxidation could be achieved within 10 s of dosing 1-2 mg/L Cl_2 (1.4-2.8 ?? 10^{-5} M) as FAC. As(III) residuals remaining in NH_3-containing waters 10 s after dosing FAC were slowly oxidized ($t_{1/2}$? 4 h) in the presence of NH_2Cl formed by the FAC-NH_3 reaction. Ozonation was sufficient to yield >99% depletion of 50 ?g/L As(III) within 10 s of dosing 0.25 mg/L O_3 (5.2 ?? 10^{-6} M) to real waters containing <2 mg-C/L of DOC, while 0.8 mg/L O_3 (1.7 ?? 10^{-5} M) was sufficient for a water containing 5.4 mg-C/L of DOC. NH_3 had negligible effect on the efficiency of As(III) oxidation by O_3, due to the slow kinetics of the O_3-NH_3 reaction at circumneutral pH. Time-resolved measurements of As(III) loss during chlorination. ?? 2006 American Chemical Society.

Index Keywords: Concentration (process); Oxidation; Ozone; pH effects; Reaction kinetics; Stoichiometry; Water treatment; Buffered reagent solutions; DOC concentrations; Time resolved measurements; Arsenic; ammonia; arsenic; chloramine derivative; chlorine; drinking water; ozone; Arsenic; Concentration (process); Oxidation; Ozone; pH effects; Reaction kinetics; Stoichiometry; Water treatment; arsenic; chlorine; drinking water; oxidation; ozone; water treatment; aqueous solution; article; chlorination; concentration (parameters); kinetics; molecular model; oxidation; ozonation; pH measurement; stoichiometry; water treatment; Ammonia; Arsenic; Cations; Chloramines; Chlorine; Hydrogen-Ion Concentration; Kinetics; Oxidation-Reduction; Ozone; Water Purification; Water Supply

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