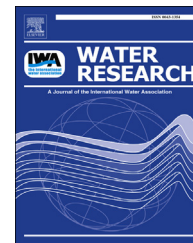




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Removal of iodide from water by chlorination and subsequent adsorption on powdered activated carbon

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ABSTRACT

Chlorine oxidation followed by treatment with activated carbon was studied as a possible method for removing radioactive iodine from water. Chlorination time, chlorine dose, the presence of natural organic matter (NOM), the presence of bromide ion (Br^-), and carbon particle size strongly affected iodine removal. Treatment with superfine powdered activated carbon (SPAC) after 10-min oxidation with chlorine (1 mg- Cl_2/L) removed 90% of the iodine in NOM-containing water (dissolved organic carbon concentration, 1.5 mg-C/L). Iodine removal in NOM-containing water increased with increasing chlorine dose up to 0.1 mg- Cl_2/L but decreased at chlorine doses of >1.0 mg- Cl_2/L . At a low chlorine dose, nonadsorbable iodide ion (I^-) was oxidized to adsorbable hypiodous acid (HOI). When the chlorine dose was increased, some of the HOI reacted with NOM to form adsorbable organic iodine (organic-I). Increasing the chlorine dose further did not enhance iodine removal, owing to the formation of nonadsorbable iodate ion (IO_3^-). Co-existing Br^- depressed iodine removal, particularly in NOM-free water, because hypobromous acid (HOBr) formed and catalyzed the oxidation of HOI to IO_3^- . However, the effect of Br^- was small in the NOM-containing water because organic-I formed instead of IO_3^- . SPAC (median particle diameter, 0.62 μm) had a higher equilibrium adsorption capacity for organic-I than did conventional PAC (median diameter, 18.9 μm), but the capacities of PAC and SPAC for HOI were similar. The reason for the higher equilibrium adsorption capacity for organic-I was that organic-I was adsorbed principally on the exterior of the PAC particles and not inside the PAC particles, as indicated by direct visualization of the solid-phase iodine concentration profiles in PAC particles by field emission electron probe microanalysis. In contrast, HOI was adsorbed evenly throughout the entire PAC particle.

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1. Introduction

The Tohoku Earthquake on 11 March 2011 resulted in the release of large amounts of radioactive materials including iodine-131 (^{131}I) into the environment from the Fukushima Daiichi nuclear power plant. After the accident, ^{131}I was detected in drinking water in 15 out of the total 47 prefectures in Japan: the highest concentration in a village of Fukushima Prefecture was 1000 Bq/kg-water and the highest concentration in Tokyo Metropolitan was 200 Bq/kg-water (Ikemoto and Magara, 2011), which points to the importance of technologies for removal of radioactive substances during water purification processes. The radioactive iodine index levels for the restriction on drinking water intake in emergency by the Nuclear Safety Commission of Japan was 300 Bq/kg for the general public and 100 Bq/kg for infants (babies who take breast feeding or formula milk). The removal of radioactive iodine by 50–90 % was required.

The addition of powdered activated carbon (PAC) may be a feasible method for removal during conventional water treatment when accidents or incidents give rise to high levels of contamination in drinking water sources (Brown et al., 2008; Lettinga, 1972). However, the efficacy of ^{131}I removal by PAC was not sufficiently high. Kosaka et al. (2012) surveyed ^{131}I removal in water purification plants after the Fukushima accident and found that the removal percentages achieved by granular activated carbon and PAC were merely 30–40% or less. The authors also conducted laboratory tests and attained the removal efficiencies of 60–70% by PAC of the dosage 50 mg/L and the contact time 30 min after chlorination of the dosage 0.5–1 mg- Cl_2 /L and reaction time 10 min.

Dissolved I (including ^{131}I) exists in various forms: molecular iodine (I_2), hypoiodite ion (IO^-), hypoiodous acid (HOI), iodide ion (I^-), iodate ion (IO_3^-), and organic iodine (organic-I). At neutral pH iodine can be present in the latter four forms (Bichsel and von Gunten, 2000a; Lettinga, 1972), while iodine is commonly found as I^- , IO_3^- , and organic-I in environmental waters (Davis et al., 2009; Gong and Zhang, 2013; Hansen et al., 2011). It has been reported that $^{131}\text{I}^-$ is poorly adsorbed by activated carbon (Ikeda and Tanaka, 1975; Lettinga, 1972). HOI is adsorbed on activated carbon to a greater extent than I^- : the adsorptive property of HOI is utilized in the iodine number, one of the most fundamental indicator widely used to characterize activated carbon as an adsorbent (AWWA, 1974). The removal of HOI is due to adsorption while that of chlorine is reductive reaction. Because HOI is formed by oxidation of I^- , the combination of chlorination and PAC adsorption could be effective for $^{131}\text{I}^-$ removal (Lettinga, 1972). However, $^{131}\text{IO}_3^-$, which is formed by oxidation of HO^{131}I , is reportedly not adsorbed by carbon (Lettinga, 1972). Therefore, chlorination of $^{131}\text{I}^-$ at a high chlorine dose, a long reaction time, or both would reduce the extent of adsorptive removal compared to that at a low dose and a short reaction time. During aqueous oxidation in the presence of natural organic matter (NOM), HOI reacts with NOM to form iodo-organic byproducts (Bichsel and von Gunten, 1999), which can be adsorbed by activated carbon (Summers et al., 1989). No systematic experimental study has been conducted to clarify how chlorination time, carbon type, and carbon dose

affect iodine species distribution and, thus, iodine removal efficacy.

In this study, we systematically investigated the formation of iodine species by chlorination and their removal from water by activated carbon adsorption. The effects of chlorine dose, chlorination time, carbon particle size, NOM content, and co-existing bromide on iodine removal were evaluated to determine the optimum conditions for removal of radioactive iodine from water. We used ^{127}I rather than ^{131}I . But the adsorption behavior of ^{127}I is similar to that of ^{131}I . Therefore, information regarding ^{127}I removal can be expected to provide important insights for the removal of ^{131}I . The removal of iodide in water is also of great significance for controlling the formation of iodinated disinfection byproducts (Ding and Zhang, 2009; Plewa et al., 2004). Hereafter, we refer to ^{127}I simply as I (or iodine).

2. Materials and methods

2.1. Sample water

NOM-free water was prepared by adding inorganic ions to ultrapure water (Milli-Q Advantage, Millipore) so that the ionic composition was equal to that of water from Lake Hakucho, Hokkaido, Japan (Table S1, Supplementary Material) (Ando et al., 2010). NOM water was prepared by adding Suwannee River NOM (International Humic Substance Society) to the NOM-free water to bring the dissolved organic carbon (DOC) concentration to 1.5 mg-C/L unless otherwise noted. Potassium iodide was added to the NOM-free water and the NOM water, and then the pH was adjusted to 7.0 with HCl or NaOH; the iodine concentration was 10 $\mu\text{g-I/L}$, which is typical of natural surface waters. In the experiments on iodine removal in the presence of bromide ion (Br^-), KBr was also added, at concentrations ranging from 0 to 2000 $\mu\text{g-Br/L}$ (Jones et al., 2011; Richardson et al., 2008). All chemicals used were reagent grade (Wako Pure Chemical, Osaka, Japan).

2.2. Activated carbon

Two types of activated carbon were used. A commercially available, thermally activated wood-based PAC (median particle diameter, 18.9 μm) was obtained from Taiko-W, Futamura Chemical (Nagoya, Japan). A superfine PAC (SPAC, median particle diameter, 0.62 μm ; Figures S1 and S2, Supplementary Material) was prepared by microgrinding the PAC with a bead mill (Metawater, Tokyo) (Ando et al., 2010). PAC and SPAC were stored as slurries in ultrapure water at 4 °C and used after dilution. The particle size distributions of PAC and SPAC were determined with a laser-light scattering instrument (Microtrac MT3300EXII, Nikkiso, Tokyo) following the addition of a dispersant (Triton X-100, Kanto Chemical, Tokyo; final concentration, 0.08 vol%) and ultrasonication.

2.3. Batch oxidation and adsorption tests

Sample water was treated with chlorine (in the form of sodium hypochlorite, Wako Pure Chemical) at concentrations of 0.01–50 mg- Cl_2 /L for 1–60 min, and then activated carbon was added. After a carbon contact time of up to 30 min, the

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