



Factors controlling mobility of ^{127}I and ^{129}I species in an acidic groundwater plume at the Savannah River Site

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ABSTRACT

In order to quantify changes in iodine speciation and to assess factors controlling the distribution and mobility of iodine at an iodine-129 (^{129}I) contaminated site located at the U.S. Department of Energy's Savannah River Site (SRS), spatial distributions and transformation of ^{129}I and stable iodine (^{127}I) species in groundwater were investigated along a gradient in redox potential (654 to 360 mV), organic carbon concentration (5 to $60\ \mu\text{mol L}^{-1}$), and pH (pH 3.2 to 6.8). Total ^{129}I concentration in groundwater was $8.6 \pm 2.8\ \text{Bq L}^{-1}$ immediately downstream of a former waste seepage basin (well FSB-95DR), and decreased with distance from the seepage basin. ^{127}I concentration decreased similarly to that of ^{129}I . Elevated concentrations of ^{127}I or ^{129}I were not detected in groundwater collected from wells located outside of the mixed waste plume of this area. At FSB-95DR, the majority (55–86%) of iodine existed as iodide for both ^{127}I and ^{129}I . Then, as the iodide move down gradient, some of it transformed into iodate and organo-iodine. Considering that iodate has a higher K_d value than iodide, we hypothesize that the production of iodate in groundwater resulted in the removal of iodine from the groundwater and consequently decreased concentrations of ^{127}I and ^{129}I in downstream areas. Significant amounts of organo-iodine species (30–82% of the total iodine) were also observed at upstream wells, including those outside the mixed waste plume. Concentrations of groundwater iodide decreased at a faster rate than organo-iodine along the transect from the seepage basin. We concluded that removal of iodine from the groundwater through the formation of high molecular weight organo-iodine species is complicated by the release of other more mobile organo-iodine species in the groundwater.

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1. Introduction

From 1955 to 1988, the F-area facilities at the U.S. Department of Energy's Savannah River Site (SRS F-area) produced radionuclides, including tritium (^3H) and plutonium-239 (^{239}Pu) for nuclear weapons, medical, industrial and scientific purposes. Seepage basins of F-area (Fig. 1) routinely received acidic (pH=1–4) wastewater containing low levels of radionuclides and chemicals from the F-area separations facilities. In addition to ^3H and Pu isotopes, fission products (e.g., ^{90}Sr , ^{95}Zr , ^{106}Ru , ^{129}I and ^{137}Cs), heavy metals (e.g., Cu, Hg and Pb), and synthetic organic compounds are reported as the main contaminants in the wastewater (Ryan, 1984; Kaplan et al., 1994; SRNS, 2008). The wastes were from nitric acid recovery units and evaporators used to concentrate uranium nitrate solutions (WSRC, 2001). The $2.7 \times 10^4\ \text{m}^2$ seepage basins were originally

unlined, 10-m deep ditches that received the acidic wastewaters (Killian et al., 1987). The F-area seepage basins are located about 600 m upgradient from Fourmile Branch, which is a secondary tributary that flows about 6.4 km to drain into the Savannah River.

Seepage basins at the SRS F-area were closed in 1988 by adding limestone and blast furnace slag to the basin bottom and then covered with a low permeability engineered barrier system to reduce groundwater infiltration. Limestone was used to promote the precipitation of trivalent, tetravalent, pentavalent and hexavalent metals, and blast slag was used to reduce radionuclides to immobile species such as Pu(IV) and Tc(IV). Since then, the pH values in groundwater near the seepage basins have increased at a rate of 0.02 units year⁻¹, while concentrations of most radionuclides significantly decreased (WSRC, 2006). One exception has been ^{129}I , where remobilization from soil has been detected and attributed to rising pH values of the groundwater (Kaplan et al., 2011) and flushing the seepage basin with large volumes with background, pH 5.5, groundwater (Denham and Vangelas, 2008).

In 2004, barrier walls and base injection gates (Fig. 1) were installed 300–500 m downgradient of the primary seepage basin to

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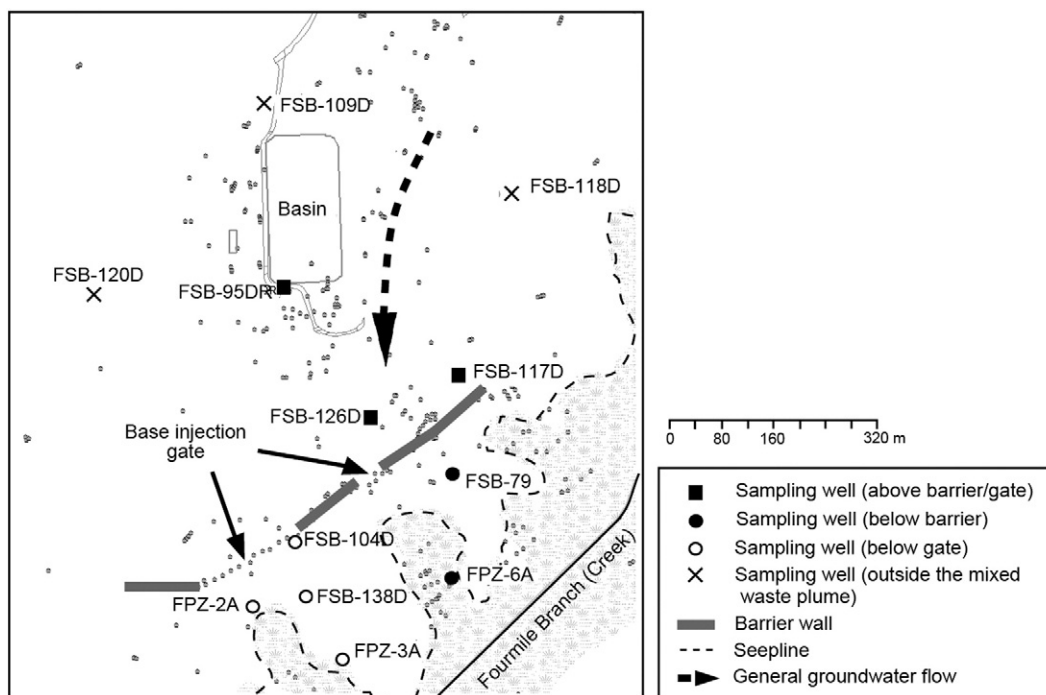


Fig. 1. Locations of the SRS F-area seepage basin, barrier walls, base injection gates and sampling wells in this study.

adjust groundwater pH for better management of ^3H and metal releases to the seep line. Since July 2005 a total of 4×10^7 l of base solution ($\text{pH} = 10$) has been added through the gates. After the installation of barrier walls, it was found that the water table upstream from the walls was elevated and that in the downstream water table was lowered (WSRC, 2006). This was presumed to be due to a “flattening” effect by blocking of the groundwater stream.

Iodine, a biophilic elements, has a stable isotope with mass number 127 (^{127}I) and 23 radioisotopes with mass number ranging from 125 to 135 that are mainly produced by atomic fission of uranium and trans-uranium isotopes. Because of its long half-life, excessive inventory and toxicity, iodine-129 (^{129}I ; half-life 15.7×10^6 years) is among the top three risk drivers for waste disposal at the Yucca Mountain, Idaho National Laboratory and Savannah River Sites (DOE, 2002, 2011; Santschi and Schwehr, 2004), and is also regarded as one of the most important radionuclides that should be assessed for its biogeochemical cycling on a global scale (UNSCEAR, 2000; Hou et al., 2008; Hu et al., 2009).

Iodine in aquatic systems on the earth's surface can be grouped into three species; iodide, iodate and organo-iodine. Since the speciation of iodine is affected by various parameters of water chemistry, such as temperature, pH, redox potential and organic matter productivity, it is necessary to understand the factors that affect its speciation to address the behavior of ^{129}I in the environment. With regard to inorganic iodine, iodine should theoretically exist as iodide in groundwater and anoxic surface waters (Fig. 2). Recent studies on SRS groundwater, however, reported that a significant proportion of ^{129}I existed as iodate (Schwehr et al., 2009; Zhang et al., 2010). The processes that produce ^{129}I -iodate under these conditions have not been quantitatively assessed.

Several studies have demonstrated that significant proportions of iodine in groundwater and surface water exist as organo-iodine (Oktay et al., 2001; Schwehr et al., 2005, 2009). Some laboratory experiments have reported that the primary product of iodide oxidation (e.g., hypiodous acid) is immediately incorporated into organic matter in soils (Bichsel and von Gunten, 1999; Warner et al., 2000). Significant amounts of iodine are eventually incorporated into soils on time scales ranging from a few days to several months

(Ashworth et al., 2003; Hou et al., 2008; Shimamoto and Takahashi, 2008; Schwehr et al., 2009; Hu et al., 2009; Yamaguchi et al., 2010). Furthermore, Amachi et al. (2001) have shown that a wide range of bacteria are capable of producing volatile methyl iodide in soils, which, however, are orders of magnitude lower in concentration than iodide, iodate or non-volatile organo-iodine species.

The degree of incorporation of dissolved iodine into soil depends on the species. Distribution coefficients (K_d) of iodide ($\sim 1 \text{ Lkg}^{-1}$) are much lower than those of iodate and organo-iodine (10^3 Lkg^{-1}) (Fukui et al., 1996; Kaplan et al., 2000; Schwehr et al., 2009). It is also known that concentrations of iodine in soils are more than three orders of magnitude higher than that in the base rocks, which is

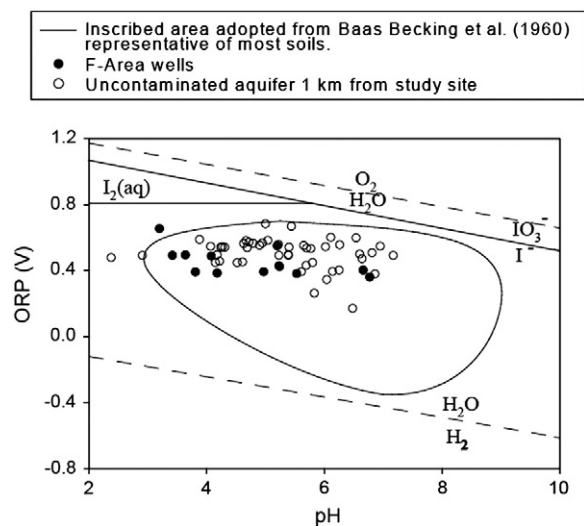


Fig. 2. Relationships between pH and oxidation-reduction potential (ORP) in the SRS F-area groundwater (closed symbols) and from the upper aquifer in the SRS E-area, adjacent to the F-area. The inscribed area was adopted from Baas-Becking et al. (1960) and is representative of most sediments. Lines representing $\text{O}_2/\text{H}_2\text{O}$ and the $\text{H}_2/\text{H}_2\text{O}$ couples provide limits of oxidation-reduction potentials in aqueous systems (organo-iodine was not considered in these calculations).

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