ABSTRACT: Iodine occurs in multiple oxidation states in aquatic systems in the form of organic and inorganic species (iodide and iodate). This fact leads to complex biogeochemical cycling of I and its long-lived isotope, $^{129}$I, a major by-product of nuclear fission $^{[1,2]}$. In order to assess the distribution of $^{129}$I and stable $^{127}$I in environmental systems, a sensitive and rapid method was developed which enabled us to determine isotopic ratios ($^{129}$/I/$^{127}$/I) and speciate I via GC-MS $^{[3]}$. Results using this new method demonstrate that the mobility of $^{129}$I species greatly depends on the type of I species and its concentration $^{[4,5]}$, pH $^{[6,7]}$, and sediment redox state $^{[7]}$, with equilibration times taking up to 12 weeks $^{[6,7]}$. For example, at ambient concentrations ($\sim10^{-7}$ M), I$^{-}$ and IO$_3$- are significantly retarded by sorption to mineral surfaces and covalent binding to natural organic matter (NOM), while at concentrations traditionally examined in sorption studies (i.e., $10^{-4}$M or higher), I$^{-}$ travels along with the water $^{[5]}$. Iodate removal can also occur through incorporation into CaCO$_3$ crystal lattice, e.g., at the Hanford Site $^{[8]}$. Iodide and iodate interactions with NOM leads to covalent binding of I to a limited number of aromatic carbon moieties on the particle surface $^{[6,9,10,11]}$. Iodine association with NOM is important in sediments, even when organic carbon are very low (e.g., $<$0.2% at Hanford Site) $^{[8,12]}$. Removal of iodine from the groundwater through interaction with NOM is complicated by the release of mobile organo-I species $^{[9,10,11]}$. A small fraction of NOM that is bound to iodine can behave as a mobile organo-I source $^{[9]}$, a process that we were able to numerically simulate using kinetic Michaelis-Menton-type redox-reactions and kinetic uptake reactions $^{[2]}$. Field $^{[13]}$ and laboratory studies evaluating the cause for steady increases in $^{129}$I concentrations (up to 1000 pCi L$^{-1}$, 3 orders of magnitude greater than drinking water limits of 1 pCi L$^{-1}$ $^{129}$I) emanating from radiological basins at SRS indicate that an increase of 0.7 pH units in groundwater over 17 years may explain the observed increased groundwater $^{129}$I concentrations $^{[14]}$. Bacteria from a $^{129}$I-contaminated aerobic aquifer at the F-area of SRS can accumulate I$^{-}$ at environmentally relevant concentrations ($10^{-7}$ M), but account for only a minor fraction of total added iodide (0.2-2.0%) $^{[15]}$, indicating that bacterial I$^{-}$ accumulation likely does not account for the high fraction (up to 25% of total I) of measured organo-I in groundwater $^{[4,16]}$. However, enzymatic oxidation of I$^{-}$ likely plays a greater role in iodination of NOM, and is currently being investigated further $^{[15,17]}$. Iodide oxidizing activity of bacteria and organic acid produced by bacteria significantly contribute to organo-iodine formation $^{[15,17]}$. Our references: [1] Kaplan et al. 2013. Crit. Rev. Environ. Sci. Technol., subm. [2] Chang et al. 2013. ES&T, subm. [3] Zhang et al. 2010. ES&T, 44, 9042. [4] Schwehr et al. 2009. ES&T 43, 7258. [5] Zhang et al. 2011. ES&T 45, 5543. [6] Xu et al. 2011. ES&T 45, 9975. [7] Emerson et al. 2013. Chem. Geol., subm. [8] Zhang et al. 2013. ES&T, in subm. [9] Xu et al. 2011. GCA 75, 5716. [10] Xu et al., 2013. STOTEN 449, 244. [11] Xu et al. 2012. GCA 97, 166. [12] Xu et al., 2013. ES&T, in subm. [13] Otosaka et al. 2011. STOTEN 409, 3857. [14] Kaplan et al. 2011. ES&T 45, 489. [15] Li et al. 2011. Appl. Environ. Microbiol. 77, 2153. [16] Li et al. 2012. p. 89-97. In: Interdisc. Stud. Environ. Chem. Vol. 6 - Environ Poll. Ecotox. Terra Sc. Publ. Comp. Tokyo. [17] Li et al. 2012. ES&T 46, 4837.