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Model of radioiodine speciation and partitioning in organic-rich and organic-poor soils from the Savannah River Site

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ABSTRACT

Radioiodine biogeochemistry was investigated by developing an integrated comprehensive model describing multiple physiochemical and enzyme catalyzed reactions based on detailed iodine speciation data obtained previously from laboratory experiments using organic-rich and organic-poor soils from the Savannah River Site, South Carolina. The model accounted for iodine speciation, inter-conversion kinetics (I^-, IO_3^-, organo-I, and colloidal-I), reversible partitioning to soil organic matter (SOM) and mineral surfaces, irreversible covalent bonding to SOM, and abiotic and biotic (enzymatic/catalyst-type) reactions. Modeling results strongly supported the assertion that iodine–SOM interactions dominate iodine geochemistry; the iodine uptake coefficient for SOM was an order-of-magnitude greater than that for mineral surface. The proposed model simulated well the iodine partitioning among the soil, colloid, and solution phases. The previously proposed process of soil reduction of IO_3^- to I^- was strongly supported through model simulations. The model revealed that during the first 14 days of contact most iodine in soil was comprised of I^- or IO_3^- associated with mineral surfaces and reversibly bound to SOM. After 14 days, the continued uptake of iodine by soil was attributed primarily to the irreversible bonding of organo-I to SOM. Finally, the model was successfully validated using an independent experimental data set. This biogeochemical modeling study underscores the importance of capturing the dynamic nature of iodine speciation transformations and the importance of treating SOM as a sink (irreversible covalent bonding) and a source (colloidal- and organo-iodine mobile species) for subsurface iodine.

Introduction

Radioactive iodine, especially ^{129}I, is a key risk driver at radioactive waste disposal facilities and contaminated sites within the Department of Energy complex, including the F-Area on the Savannah River Site (SRS; Aiken, SC) and 200-West Area on the Hanford Site (Richland, WA). Much of the risk associated with ^{129}I can be attributed to its long half-life ($t_{1/2} = 15.7$ million years), large inventory, high toxicity, high mobility in the environment, and most importantly biophilic nature [1]. Radioiodine can be easily incorporated into the human body because stable iodine, ^{127}I, is an essential element used by the thyroid gland in the production of hormones, whose concentration levels may cause several health issues such as growth and developmental problems in children and goiter in adults [2]. Thus, the maximum contaminant level (MCL) of ^{129}I in groundwater issued by the US-EPA is the lowest among all monitored radionuclides (1 pCi L^{-1}).

The fate and transport of released iodine in the subsurface environment are not well understood, primarily because of problems associated with the analytical detection of the various iodine species, which profoundly alter iodine biogeochemistry [1]. Iodine in the natural environment exists mainly as three species, iodide (I^-), iodate (IO_3^-), and organo-iodine (Org-I). Slight changes in environmental conditions, such as pH, E_h, microbial activity, and dissolved organic carbon (DOC), have been shown to readily promote transformations between these species or other less common species, such as I_2 or hypoiodous acid (HOI). The degree of sorption or uptake of the various iodine species can also vary greatly. Generally, the degree of I^- sorption tends to be much less than the other species, i.e. IO_3^- and Org-I. Reported K_d values (iodine concentration ratio of solids/

Abbreviations: AFW, artificial freshwater; CA, component additive; CBD, citrate–bicarbonate–dithionite; COC, colloidal organic carbon; COI, colloidal organo-iodine species; COM, colloidal organic matter; DOC, dissolved organic carbon; HA, humic acid; LSC, liquid scintillation counter; OC, organic carbon; OP soil, organic-poor soil; OR soil, organic-rich soil; Org-I, organo-iodine; SOM, soil organic matter; SRS, Savannah River Site; TOC, total organic carbon.

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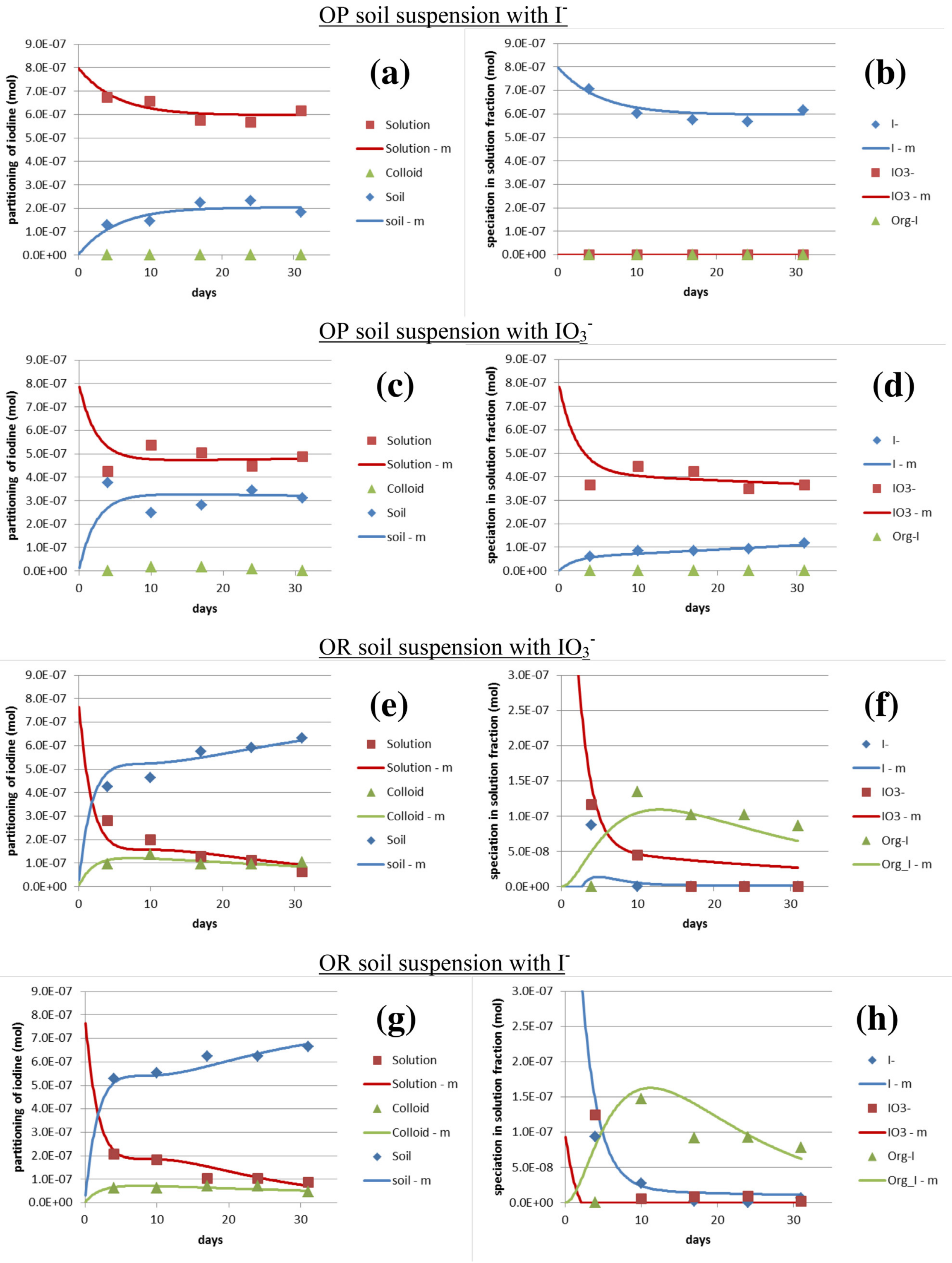

Fig. 1. Partitioning and speciation of added iodine in soil suspensions. Plots (a)–(d) used the OP soil (<0.01% OM and pH = ~6.9). Plots (e)–(h) used the OR soil (24.1% OM, pH = ~5.2). Solution phase = <3 kDa; colloidal phase = 3 kDa – 0.45 μm; particulate soil = >0.45 μm. Symbols = experimental data [11]; lines = model simulation (spike = 20 μM (equivalent of 8.0×10^{-7} mol) iodine, 10 g soil, and 40 mL artificial freshwater).