Evaluation of a Low Cost Humate Solution as an In Situ Amendment to Enhance Attenuation of Uranium in an Acidic Plume – 15445

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ABSTRACT

A single well injection test was conducted in the acidic portion of a groundwater contamination plume beneath the F-Area Seepage Basins on the U.S. Department of Energy's Savannah River Site to evaluate humate as a viable amendment to enhance attenuation of uranium and other radionuclides. Humate is typically used as an inexpensive organic fertilizer. A solution of humate was injected into a monitoring well in the heart of the F-Area Seepage Basins plume. The geochemical conceptual model suggests humate adsorbs to mineral surfaces creating a subsurface treatment zone in which uranium adsorption is enhanced. Following humate injection, groundwater was sampled from the injection well at regular intervals to determine the composition of groundwater and to evaluate the sorption of contaminants within the treatment zone. Attenuation of uranium, radioiodine and Strontium-90 was determined by comparing concentrations to analytes unaffected by the humate, specifically tritium, nitrate, and specific conductance. The post-injection monitoring data showed enhanced uranium attenuation in the treatment zone as uranium concentrations rebounded to only 68% of the pre-test concentrations. Radioiodine attenuation was complicated by changing speciation during the test. When pH was elevated by the humate injection, iodate dominated and enhanced attenuation was observed. As pH decreased, iodide dominated and enhanced attenuation decreased. Humate did not significantly attenuate Stronium-90. Results of the humate injection test demonstrate that humate strongly adsorbs to the aquifer sediments at acidic pH. Humate desorption curves suggest that a substantial fraction of humate will remain adsorbed to the sediments for long-periods of time.

INTRODUCTION

The conceptual approach of using derivatives of natural humic substances to enhance the attenuation of contaminant metals and radionuclides evolved from the tendency of metals to bond with organic matter leading to the formation of ore deposits in natural systems. The association of uranium with natural organic matter has long been recognized [1, 2, 3, 4]. Similarly, organic matter may play a role in formation of other metal ores [5, 6, 7]. Paralleling studies of metal accumulation with organic matter were studies demonstrating the effect of humic substances on metal behavior in natural waters [8, 9, 10, 11, 12, 13]. In the simplest terms, the mobility of a metal with affinity for humic acid is determined by the mobility of the humic acid. In groundwater, humic acid can decrease mobility of metals in which conditions are conducive to sorption of the humic acid to mineral surfaces. When conditions favor mobility of the humic acid, metal mobility can be enhanced by the presence of humic acid. This observation suggests in-situ approaches for remediation of metal-contaminated groundwater by injection of humic amendments into the aquifer. Petrović et al. [14] suggested humic substances could be used to mobilize some metals and enhance the sorption of other metals. Remediation based on mobilizing metals is considered riskier than remediation by immobilization and more effort has been focused on the latter. Oeste and Kempfert [15] describe a method for creating a permeable reactive barrier in a two-step process by first precipitating injected dissolved humic acid followed by a subsequent injection of acid. To avoid injection of acids, a method of creating humic derivatives that adhere more strongly to soil or other substrates under a variety of conditions was described by Perminova et al. [16]. However, under the acidic groundwater conditions typical of many metal and radionuclide contaminant plumes, humic substances sorb well to mineral surfaces. In such cases, acid injections or specialized humic derivatives may not be necessary to create a subsurface treatment zone for many metals.

Wan et al. [17] demonstrated that peat and soil humic acid reference materials, when sorbed to Savannah River Site soils, enhanced the sorption of uranium under acidic conditions (pH=3.0 to 4.5). The sorption of the humic acid was rapid with minimal reversibility in both batch and column studies. The column studies simulated injection of humic acid into the subsurface to remediate uranium contamination. Wan et al. [17] found that there was an initial increase in uranium concentration upon transition from injection of humic acid to flushing with simulated acidic groundwater, likely due to humic-uranium aqueous complex formation. After the initial increase, uranium in the effluent of the humic acid treated columns decreased rapidly to very low concentration, whereas the uranium concentration in the effluent of untreated columns remained high. This demonstrated the potential use of humic acid solutions as an injectable amendment to treat uranium contamination in acidic aquifers.

A potential challenge to the use of soluble humic substances is the cost of the materials. Most groundwater contamination sites will require several thousand kilograms of concentrated humic substances to remediate a plume. Processing and purifying commercial sources of humic substances increases their cost. Hence, a single well injection test was conducted with an inexpensive organic amendment at a site with acidic groundwater contaminated with uranium, Stronium-90 (Sr-90), tritium, nitrate, and Iodine-129 (I-129). The amendment solution is an alkaline mixture of humic compounds, hereafter referred to as “humate” because it is not acidic and the components are the anions of humic acid compounds.

DESCRIPTION

Field Test Site

The F-Area Seepage Basins (FASB) on the Savannah River Site (located near Aiken, South Carolina) was chosen as the field test site. The site consists of three basins that were originally unlined, earthen surface impoundments used to dispose liquid wastes from the F-Area Separations facility (Fig. 1). From 1955 through 1988, these unlined basins received approximately 7.1 million cubic meters of low-level waste solutions originating from the processing of uranium slugs and irradiated fuel. The liquid wastes were acidic (wastewater with nitric acid) and low activity waste solutions containing a wide variety of radionuclides and dissolved metals [18, 19]. The wastewater was allowed to evaporate and to seep into the underlying soil. The purpose of the basins was to take advantage of the interaction with the basin soils to minimize the migration of contaminants to exposure points. Though the seepage basins essentially functioned as designed for many potential contaminants, some metals and radionuclides reached the water table resulting in the formation of groundwater contaminant plumes. The primary contaminants having concentrations that consistently exceed regulatory standards are tritium, uranium, Stronium-90, Iodine-129, and nitrate. In the most acidic portions of the contamination plume pH is near 3.

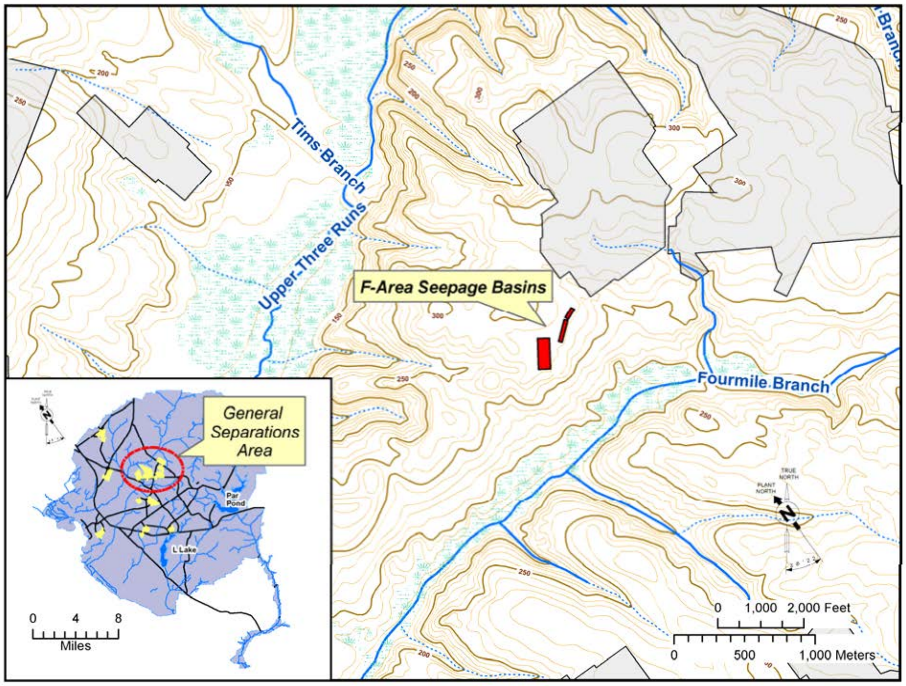

Fig. 1: Location of the F-Area Seepage Basins at the Savannah River Site

A detailed discussion of the selection of the test well, as well as upgradient and downgradient wells can be found in Millings et al. [20]. Briefly, well FOB-16D was chosen as the test well because it is in the heart of the plume both laterally and vertically. Another important factor was that the well is not regularly sampled for regulatory compliance purposes. Therefore the test could be performed in FOB-16D without interfering in any site operations. The upgradient wells, FSB-95DR and FSB-94DR, and the downgradient well, FSB-126D, were chosen because they are located approximately along a flow path from Basin 3 through the test well. Fig. 2 shows the locations of the wells relative to the FASB and the general flow direction of the groundwater.

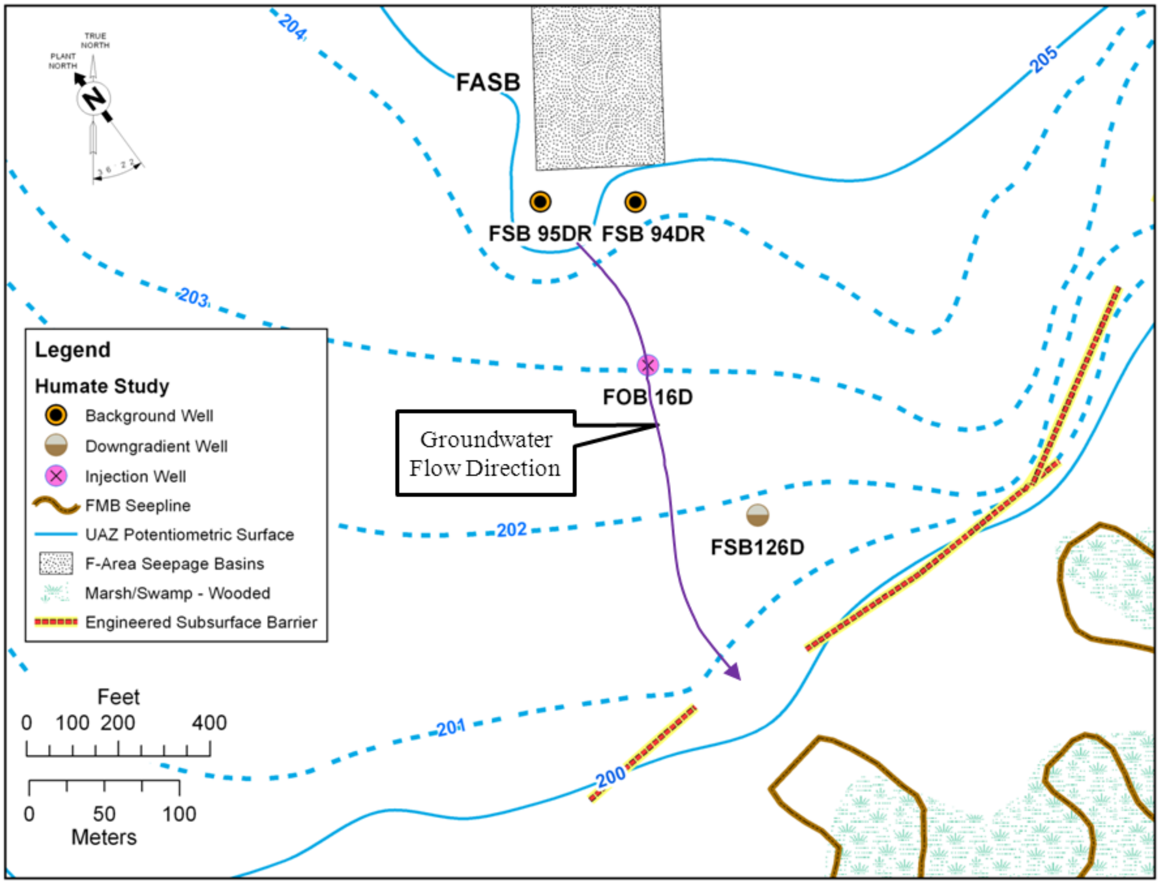

Fig. 2: Map showing location of the pilot test wells. The potentiometric surface of the upper aquifer is shown in blue and the general direction of groundwater flow is indicated by the purple arrow.

Humate Amendment

For the field test, it was important to choose a relatively inexpensive and commercially available product to demonstrate whether or not humate amendments could be a realistic remedial option for large groundwater plumes typical of U.S. Department of Energy sites. For this field study, Huma K© was chosen as the humate amendment because of its ease of use in dry flake form, as well as existing data and experience with the amendment [20, 21]. Huma K© is a commercially available, dry flake, organic amendment manufactured by Land and Sea Organics. The amendment is high in humic and fulvic compounds and is just one of several brands produced for large scale use as soil conditioners to boost productivity in organic agriculture. It is certified by the Organic Materials Review Institute for use in organic farming, based on a number of stringent criteria including low concentrations of trace metals.

Single Well Injection Test

The idealized conceptual model for the single well injection test is that as the humate solution is injected into the subsurface, the humate will sorb to sediments in the affected volume around the injection well screen. Once injection is complete, the contaminated plume water will begin to flow back through the upgradient portion of the affected volume, or treatment zone. An idealized diagram of the concept is shown in Fig. 3. As the groundwater moves through the injection zone, contaminants with an affinity for humate adsorb to the humate bound on minerals surfaces, thereby lowering the concentrations of the contaminant in the impacted groundwater. Over time, the upgradient portion of the treatment zone becomes saturated with contaminant, and concentrations of contaminants in the well bore increase. This is the simplest of field tests with two goals: 1) to determine if the humate solution can be injected into the subsurface without clogging the well, and 2) to determine if the treatment zone created by sorbed humate enhances sorption of contaminants.

Additional information can be derived if the field test is supplemented with laboratory tests on sorption of the humate used in the field test. This is important because sorption of the humate injected into an acidic plume is complicated by the varying pH in the injection volume as the alkaline humate solution interacts with acidified mineral surfaces. Humate sorption to sediment minerals varies with pH, sorbing more strongly at low pH than high [22, 23]. The humate solution has a pH that approaches 10, whereas the aquifer at F-Area typically has a pH of 3.2. As the humate solution is injected, a pH gradient is established that moves away from the borehole until injection is complete.

Humate adsorption-desorption studies on cored sediments from the test site support the conceptual model of injected humate adsorbing strongly onto aquifer minerals [24] Desorption studies were done in which the sediments used in the adsorption studies were eluted 4 times with fresh water. These showed hysteresis in the adsorption-desorption curves for all sediments and indicated that desorption of humate from an in situ treatment zone would be slow.

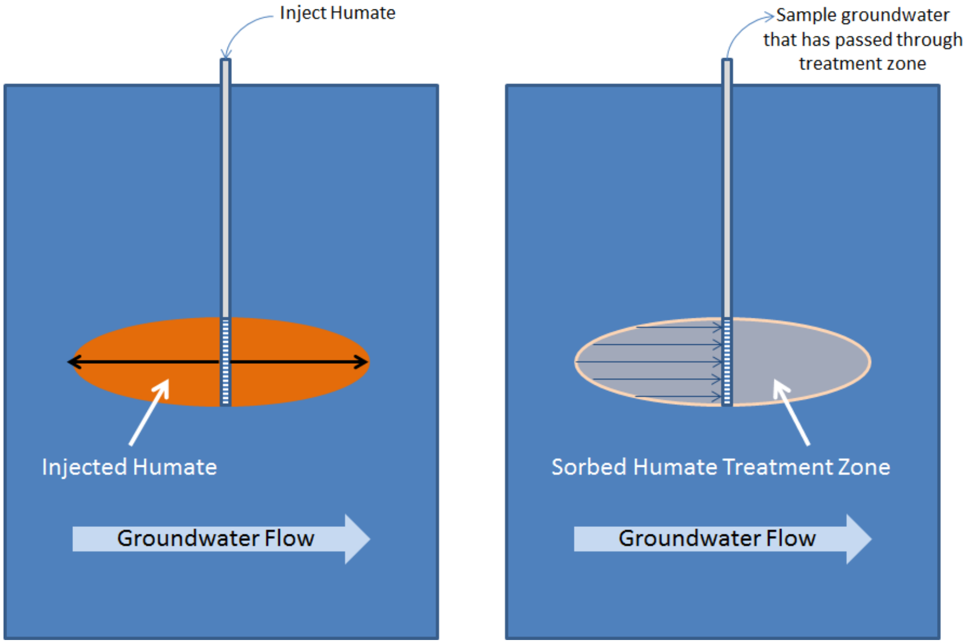

Fig. 3: Idealized conceptual model of the single well humate injection test.

The humate injectate solution contained approximately 10,000 mg/L Huma-K© which is equal to approximately 3200 mg/L organic carbon. The pH and specific conductance of the injectate solution were measured periodically during injection using a YSI sonde. The mean pH was 9.9 and the specific conductance was 1840 uS/cm. A large volume peristaltic pump was used to transfer the humate injectate from totes to the screen zone of the well. The humate injectate was not pressurized but rather allowed to flow under a gravity feed. The tote tank was connected to the peristaltic pump via a valved outlet on the tote. A 100 micron filter was connected to the outlet side of the peristaltic pump to remove any large humate debris in an effort to prevent clogging of the well.

At the well, the humate injectate flowed through a 2.5-cm schedule 40 PVC casing with a 1.5 meter screen attached at the bottom. This screen was set within the screen zone of the well at the desired elevation in order to accurately emplace the material during injection. Monitoring of the water level in the well was conducted to evaluate the amount of head placed on the humate injectate. Mixing and injection took place over the course of seven hours.

The injection of humate was followed by monitoring the same well for a period of 4 months to sample groundwater that had passed through the upgradient portion of the treated injection volume. Sampling was more frequent early in the monitoring period to capture transient effects and graded to monthly samples for the final 2 months.

Standard field parameters (pH, specific conductance, oxidation-reduction potential, dissolved oxygen, and temperature) were measured during sampling using a YSI sonde with a flow-through cell. Cations, anions, Stronium-90, tritium, uranium isotopes, and total organic carbon were analyzed by an E.P.A. Certified laboratory using U.S. Environmental Protection Agency-approved methods. Speciation of radioactive iodine was done by Savannah River National Laboratory.

DISCUSSION

Monitoring Results

Normal temporal variations in contaminant concentrations over a 4 month period at the test site are sufficient that they must be considered when evaluating the effect of the injected humate. For example, Fig. 4 shows systematic decrease in specific conductance values measured at the test well over the course of the field test. It can be assumed that a similar pattern in concentrations of contaminants also occurred and temporal variations in tritium and nitrate (reported as nitrate-nitrite as nitrogen) concentrations support this assumption. These contaminants would not react with humate and their concentrations should return to ambient values after dilution effects of the humate injection have dissipated. In contrast, concentrations of contaminants that are attenuated by humate should remain lower than ambient concentrations after dilution effects have dissipated. To truly evaluate attenuation by humate, the decrease in ambient concentrations must be considered.

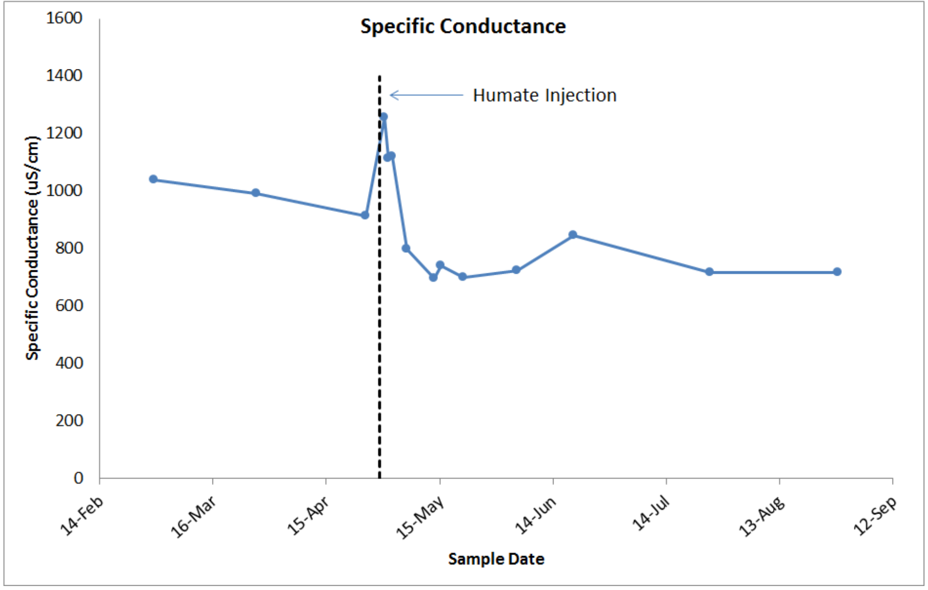

Fig. 4: Specific conductance values in groundwater extracted from the test well during the humate injection field test.

Specific conductance was used as a baseline parameter to quantitatively evaluate effectiveness of humate 4 months after the injection. Specific conductance was chosen because it is a composite parameter, less subject to short-term variations, sampling artifacts, and analytical error than specific contaminants. The evaluation was done by calculating a predicted “untreated” concentration for the contaminant at 4 months based on the specific conductance at 4 months. The 3 pre-test measurements of specific conductance were averaged and the ratio of specific conductance at 4 months to the pre-test average was calculated. This ratio was used to calculate the predicted untreated concentration of a contaminant assuming this ratio was equal to the corresponding ratio of contaminant concentrations.

Nitrate and chloride were used to test this method. These constituents are not likely to be sorbed in the humate treatment zone and thus the predicted untreated concentrations should be equal to the measured concentrations. Fig. 5 shows this to be the case. The blue diamonds are measured data and the orange squares are the predicted untreated concentrations at 4 months, based on specific conductance.

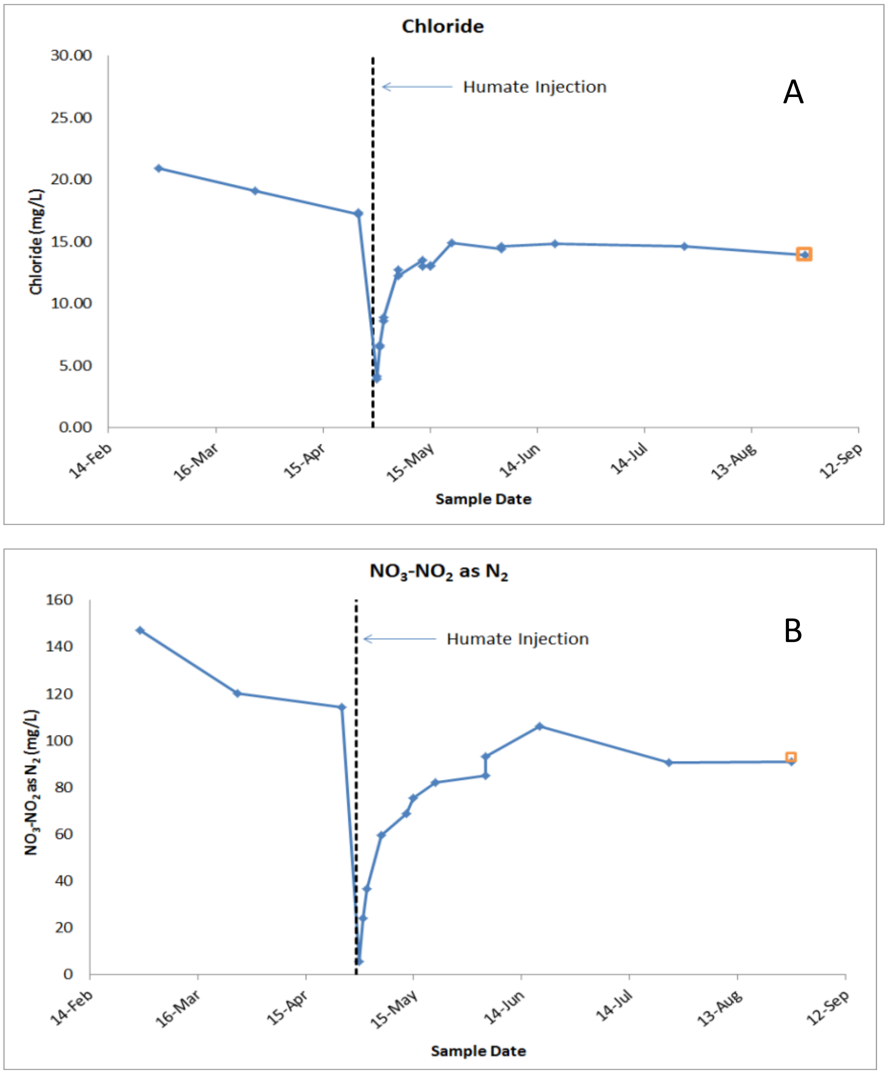

Fig. 5: A) Chloride concentration at test well during humate injection test, B) Nitrate-nitrite as nitrogen concentration at test well during humate injection test (orange squares are predicted untreated values).

Nitrate was used in a similar way as specific conductance to qualitatively evaluate the effects of humate during post-injection rebound of contaminant concentrations. Specific conductance cannot be used for this purpose because the injected humate solution had a high specific conductance. In contrast, nitrate concentration was negligible in the injection solution as indicated by the concentration during injection (Fig. 5B).

The value of pH during the test is important because uranium and Sr-90 sorption in the presence or absence of humate is dependent on pH. Fig. 6 shows the pH values in groundwater extracted during the test. The injection of the alkaline humate solution brought the pH from 3.3 to near 10. As acidic groundwater flushed through the treatment zone the pH decreased reaching 3.6 by the end of the test.

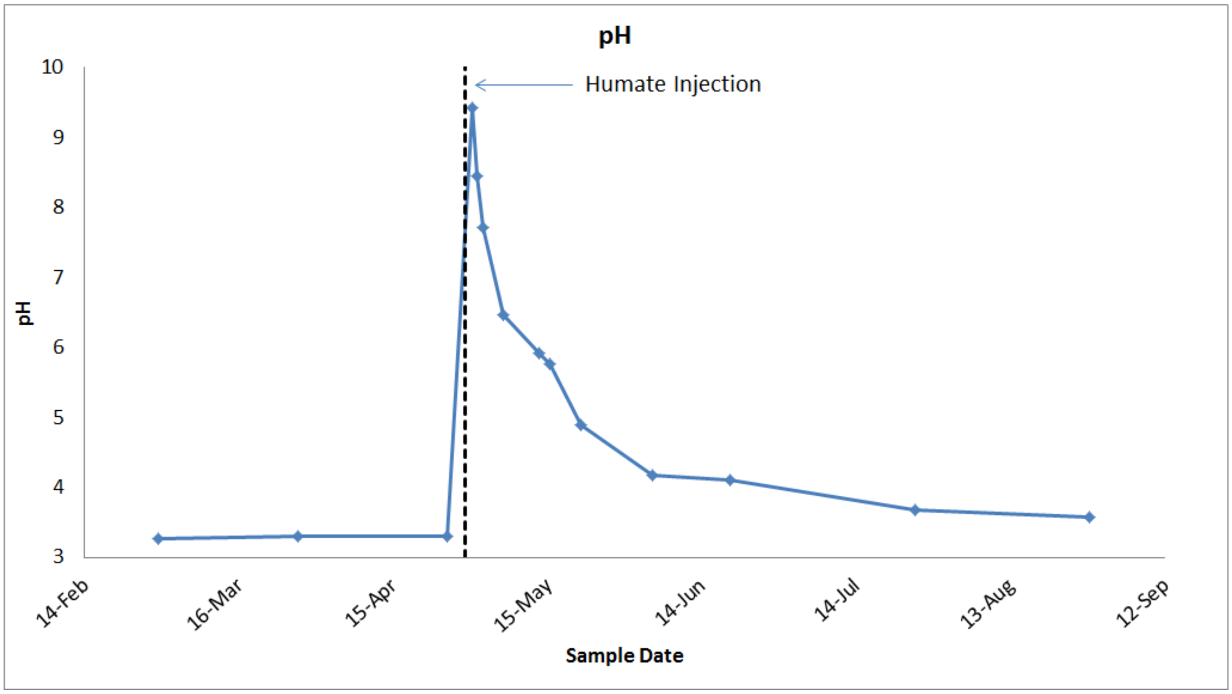

Fig. 6: Values of pH during the humate injection field test.

Fig. 7 shows the concentration of Sr-90 in groundwater from the injection test well, FOB-16D, during the humate injection field test. The pattern is similar to the non-reactive constituents and, by the last sampling event, the Sr-90 concentration is equal to that projected using specific conductance. Nevertheless, when the rebound of Sr-90 concentrations is compared to the projected rebound (based on rebound of nitrate) there does appear to be minor sorption of Sr-90. However, the sorption capacity of the treatment zone for Sr-90 was low, and the concentration returned to the projected plume concentration by the last sampling event.

The measured uranium rebound curve in Fig. 8 is compared to the projected rebound curve calculated from the rebound of nitrate. The curve indicates that humate enhanced sorption of uranium. The uranium concentrations in the early portion of the test have an interesting pattern that differs from other constituents. The uranium concentration decreases from the injection dilution, but 3 days after the injection increases from 135 ug/L to 458 ug/L and remains essentially the same until 14 days after the humate injection, when it begins to decrease. Uranium continues to decrease to 31 ug/L at 22 days after injection and then begins to increase again, with the rate of increase slowing at 51 days after injection. At the end of the 4 month monitoring period, the uranium concentration rebounded to 68% of the untreated concentration projected using the specific conductance.

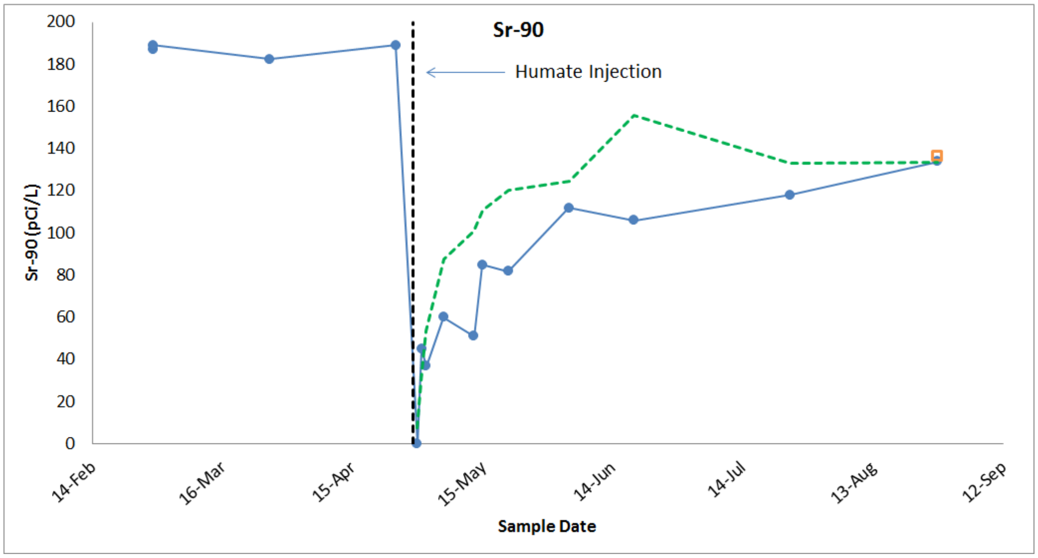

Fig. 7: Sr-90 concentrations during the humate injection field test (green dashed line is projected untreated rebound curve based on nitrate; orange square is the projected untreated concentration based on specific conductance).

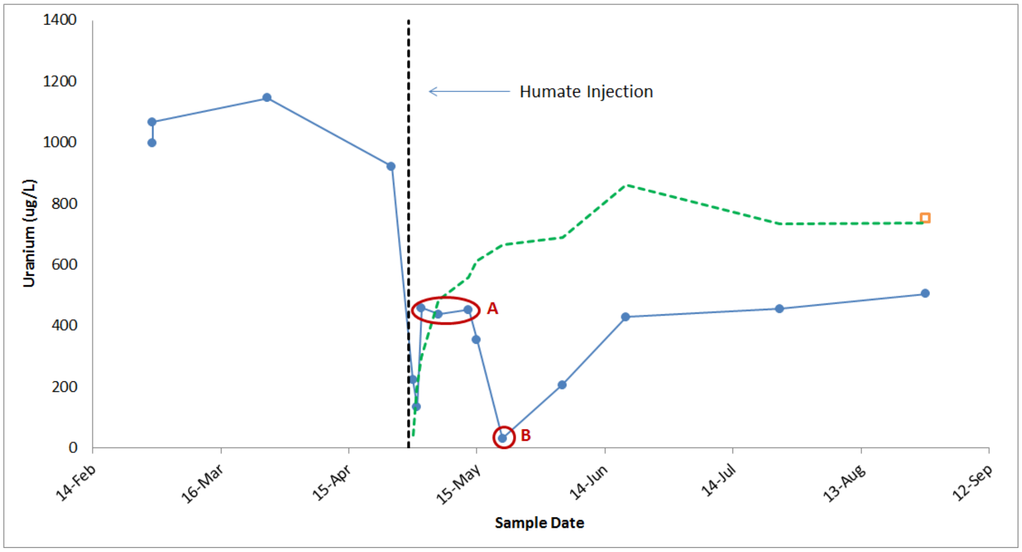

Fig. 8: Uranium concentrations in groundwater from the injection well, FOB-16D, during the humate injection field test compared to projected rebound concentrations calculated by the nitrate (green dashed) rebound curve; orange square is projected untreated concentration based on specific conductance.

The cause of the unusual pattern of uranium concentrations is unclear, but is likely the result of aqueous complexing of uranium by dissolved humate. During the time period highlighted in the region labeled “A” in Fig. 8, the pH decreased from 7.7 to 5.9 and dissolved organic carbon concentration was high. These conditions would favor formation of aqueous uranium-humate complexes. At the point labeled “B” the pH had decreased to 4.9 and 98% of the dissolved total organic carbon had flushed from the treatment zone or had been adsorbed. This point is the maximum sorption of uranium in the humate treatment zone. Over the next 3 months uranium concentration increased, but never reached the projected untreated concentration, indicating that humate effectively enhanced the attenuation of uranium. The effectiveness lasted for greater than 18 to 72 pore volumes of groundwater passing through the humate treatment zone, based on a range of estimated flow velocities at the site compiled by Tokunaga et al [25].

In general, iodine concentrations in groundwater from the extraction well behaved similarly to other constituents during the humate injection field test. Stable I-127 results are reported here as a surrogate for I-129. Iodine can exist in organic and inorganic species with inorganic iodine dominated by iodide (I^-) and iodate (IO_3^-). Unfortunately, organic iodine species were not measured in time to be reported here. Fig. 9 shows the results for total inorganic iodine and iodate during the field test. The inorganic speciation of pre-test I-127 was dominated by iodide, but as rebound of I-127 began after the injection of humate, the inorganic iodate dominated. During this time, pH was also elevated. In a study of iodine speciation in the F-Area Seepage Basins plume, Otosaka et al. [26] observed that, in acidic portions of the plume, the dominant inorganic iodine species was iodide. In the portions downgradient of the base injection, where pH was elevated, iodate dominated. In the humate injection field test, it is likely that the elevated pH introduced by the humate injection controlled speciation of I-127, favoring iodate. As the pH decreased below 6.5, the fraction of iodide increased. At pH below 5.8, iodide dominated. During early rebound, when speciation was dominated by iodate, there was minor attenuation relative to the rebound curve projected using nitrate rebound (green dashed line). Once iodide dominated, attenuation was minimal.

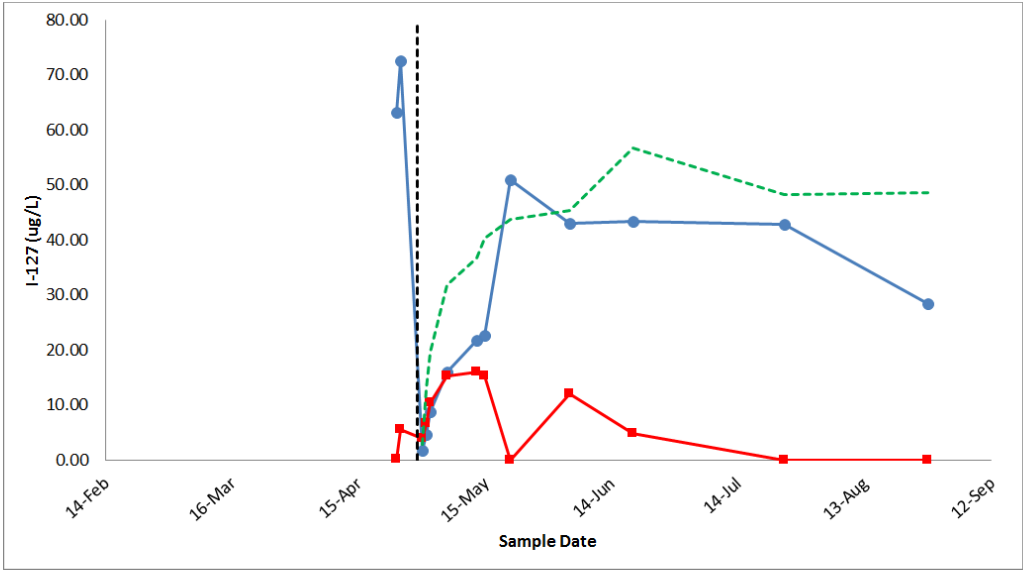

Fig. 9: Total inorganic I-127 (blue circles) and iodate I-127 (red squares) concentrations in groundwater from the injection well, FOB-16D, during the humate injection field test compared to untreated rebound curve projected using nitrate (green dashed line).

CONCLUSIONS

A humate solution was injected into an acidic aquifer contaminated with uranium, Sr-90, tritium, nitrate, and I-129 in a single well injection test to assess the feasibility of creating an in situ adsorbed humate treatment zone that would enhance attenuation of uranium and other radionuclides. The humate injection field test demonstrated that a concentrated solution of unrefined humate can be injected into an acidic aquifer with little difficulty. The injection of 2000 liters through a 1.5 meter screen was completed by gravity feed in less than 8 hours with no indication of formation or well-screen clogging. It is anticipated that much larger injections, as required in a full-scale deployment, could be done with no issues.

The treatment zone created by the humate solution injection substantially attenuated uranium. Four months after humate injection, the uranium concentration had rebounded to only 68% of the pre-injection concentration, accounting for a decreasing trend in uranium concentrations over the course of the test. In contrast, tritium and nitrate concentrations rebounded completely. The effects of the treatment zone on Sr-90 were negligible. Iodine speciation was affected by the treatment, but long-term attenuation was not significant.

Humate adsorption studies indicate that humate strongly adsorbs to mineral surfaces at a pH of 4. Desorption studies indicate that a large fraction of the humate injected would remain adsorbed for long periods of time.

The results of this study suggest that is feasible to use inexpensive sources of humate to create full-scale, in-situ treatment zones for remediation of uranium and other metals that bind to humate in acidic groundwater.

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