Iodine dynamics with humic acid

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The anionic fission product iodine-129 is of specific concern to policy makers regarding human and environmental risk assessments. Previous research has demonstrated a strong link between soil organic matter (SOM) content and reduced mobility of iodine within the environment. Negatively charged humic substances, such as humic acid (HA), may constitute 80% of organic matter and the mechanisms that enable an anionic radionuclide to interact with HA are not well understood. Here we show how the change in surface charge development of HA with pH affects the consequent reduction and binding of the anions iodide ($^{129}I^-$) and iodate ($^{129}IO_3^-$). Using inductively coupled plasma mass spectrometry (ICP-MS) linked to high performance liquid chromatography (HPLC) we monitored the changes in iodine speciation as it was incoporated into native pools of organic iodine. Over time the more oxidised iodine species, $^{129}IO_3^-$, was rapidly transformed to intermediate species that then formed both $^{129}I^-$ and organically bound iodine (Org-^{129}I). As predicted, when pH decreased, the rate of this reduction reaction increased. Thirty days after spiking with 10 ppb $^{129}IO_3^-$ the quantity removed from solution in the presence of HA was 71, 76, 84, 91, 97, 100 and 100% at pH 7, 6.5, 6, 5.5, 5, 4.5 and 4 respectively, showing a strong reaction rate dependence on pH. The same was seen for HA concentration where increasing HA concentration increased the rate of $^{129}IO_3^-$ reduction, and consequent I removal from solution. Previous research has suggested that that the most likely mechanism is $^{129}IO_3^-$ reduction to $^{129}I^-$ which then goes on to bind with phenolic groups on organic matter forming Org-^{129}I species. However using size exclusion chromatography (SEC) it can be seen that $^{129}IO_3^-$ was rapidly bound to HA forming Org-^{129}I species with no initial evidence of $^{129}I^-$ formation. Over time $^{129}I^-$ concentration increased. This was observed as rapid formation of Org-^{129}I, followed by a decline as $^{129}I^-$ concentration increased in the solution phase. This information is currently being used to develop new models to predict the behaviour of iodine within soil systems.