

Multi-isotopic constraints on contamination history, contaminant migration and structure of the F-Area acidic plume, Savannah River Site

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Seepage basins in the F-Area of the Savannah River Site were used from 1955 to 1989 for the disposal of low-level radioactive acidic (ave. pH ~2.9) waste solutions from site operations involving irradiated uranium and other materials used in the production of radionuclides. These disposal activities resulted in a persistent acidic groundwater plume (pH as low as 3.2) beneath the F-Area including contaminants such as ³H, HNO₃, ⁹⁰Sr, ¹²⁹I and U that has impinged on surface water about 600 m from the basins. After cessation of disposal in 1989, the basins were capped in 1991. Since then, remediation consisted of a pump-and-treat system that was recently replaced by *in situ* treatment using a funnel-and-gate system with injection of alkaline solutions to neutralize pH.

In order to delineate the history of contamination and the current mobility and fate of contaminants in F-Area groundwater, we have undertaken a study of variations in the isotopic compositions of U (²³⁴U/²³⁸U, ²³⁵U/²³⁸U, ²³⁶U/²³⁸U), Sr (⁸⁷Sr/⁸⁶Sr), Nd (¹⁴³Nd/¹⁴⁴Nd) water (δ¹⁸O, δD) and nitrate (δ¹⁵N, δ¹⁸O) within the contaminant plume. The chemical and isotopic variations in the plume all delineate upper (0-15ft below water table) and lower (15-35 ft) zones within the upper aquifer. Together, the data suggest that the lower zone represents the effects of seepage from the basins during operation, while the upper zone represents mostly meteoric water contaminated by infiltration through the sub-basin vadose zone since closure/capping. Through U isotopic analysis, we have been able to detect recent migration as the plume expands laterally, with the greatest extent of U contamination near the top of the aquifer but decreasing with depth. Comparing the U isotopic compositions of groundwater samples collected within the plume two years apart tracks the migration of U from up-gradient portions of the plume. Nd isotopic compositions indicate that REE concentrations were controlled by progressive interaction between acid solutions and natural trace minerals in the sediments. Nitrate has an isotopic signature of processed waste, but no sign of nitrate bioreduction within the plume.

Vertical distribution of ²³⁶U in the western equatorial Atlantic Ocean

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During the Pelagia Geotraces cruise PE321 in summer 2010 two depth profiles of ²³⁶U were sampled in the western equatorial Atlantic Ocean (WEA) and subsequently analyzed at ETH Zurich using low energy accelerator mass spectrometry (AMS). These data represent the first ²³⁶U-measurements in the open Atlantic Ocean.

²³⁶U is almost exclusively produced by neutron capture on ²³⁵U. While the natural background of ²³⁶U/²³⁸U is estimated to be in the 10⁻¹² - 10⁻¹⁴ range anthropogenic ²³⁶U/²³⁸U ratios are much higher. For example, it can be estimated that as a consequence of the atmospheric nuclear bomb explosions about 1-2 tons of ²³⁶U were blown into the atmosphere. Mixed with the upper few hundred meters of the Ocean this would result in ²³⁶U/²³⁸U ratios in the 10⁻⁹ range. Although the expected signal dynamics of ²³⁶U/²³⁸U in the environment is very large, currently only AMS-systems have the capability for a fast and quasi background free detection of ²³⁶U/²³⁸U ratios significantly below 10⁻⁹.

At stations 39 and 40 of the PE321 cruise two depth profiles were sampled for ²³⁶U (25 m, 2500 m, and 4250 m). The ²³⁶U/²³⁸U ratios decrease from about 10⁻⁹ at the surface down to about 10⁻¹⁰ in the Antarctic Bottom Water (AABW). The advective contribution of anthropogenic ²³⁶U from the North Sea (from nuclear reprocessing plants) seems unlikely at this location. The most likely explanation for the elevated ratios in the AABW is the transport and the subsequent release of particle bound ²³⁶U from the biologically active surface waters. Extrapolating from the calculated ²³⁶U-inventories a total input of 1.5 - 2 tons of ²³⁶U can be estimated for the global fallout. The inventory calculations are consistent with global fallout as the sole source for ²³⁶U at the WEA. However, simple box model results show that very high U-export rates from the surface layer to the deep sea (>50 ng cm⁻² yr⁻¹) would be necessary to reproduce the measured concentration profile.

Furthermore, a close correlation of salinity and ²³⁶U was found, which is much steeper than the relation with natural U-isotopes in the open ocean. This indicates that, in contrast to natural U, ²³⁶U has not yet reached steady state. Our results indicate that ²³⁶U might have a large potential as a new conservative and transient tracer in Oceanography.