

Uranium isotopic variations as a tracer of environmental contamination in sediment cores

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This study investigates potential environmental uranium contamination from the Fernald Feed Materials Production Center (FFMPC) located in SW Ohio. In 1951, the FFMPC site was established to process high purity uranium products from natural uranium ore and recycled recoverable residues, including enriched (EU) and depleted uranium (DU). The facility is estimated to have inadvertently released 200,000 to 1 million pounds of uranium into the environment during its operation from 1952 to 1985 [1]. Previous studies conducted in proximity to the FFMPC site have found elevated uranium concentrations in sediment cores [2] and non-natural uranium isotopic signatures indicative of an anthropogenic source in local tree bark [3].

This study tests the utility of lake sediment as a proxy for the temporal and aerial extent of uranium contamination, as well as to constrain the sources of uranium. A sediment core 80 cm in length was collected from an impoundment located 6.7 km south of the FFMPC site. The core was divided into 2 cm sections, resulting in forty sediment samples. Three samples that span the length of the core (top, middle and bottom) were analyzed using gamma spectrometry to determine ²¹⁰Pb and ¹³⁷Cs ages. The relative ²¹⁰Pb excess and the lack of measurable ¹³⁷Cs in the deepest sample suggest that it dates to the early 1950's and thus may pre-date any FFMPC activity.

Forty samples representing the entire sediment core were analyzed by ICP-MS, and were found to have U concentrations ranging from 0.33 to 1.33 ppm. Most samples exhibit a strong positive correlation between U and Th concentration. However, anomalously elevated U concentrations were found in nine consecutive samples from immediately above the base of the core (62 to 78 cm depth). In order to determine whether the elevated U was due to natural or anthropogenic sources potentially related to the FFMPC, twenty-one samples were analyzed for U isotopic composition by TIMS. The deepest sample, inferred to have been deposited in the early 1950's, has a natural ²³⁵U/²³⁸U signature and no measurable ²³⁶U, consistent with this sediment pre-dating FFMPC activity. Furthermore, the samples with anomalously high U concentrations have variable ²³⁵U/²³⁸U ranging from DU to EU signatures (0.00645-0.00748), and all contain measurable ²³⁶U with ²³⁶U/²³⁸U ranging from 2.1x10⁻⁶-3.6x10⁻⁵. Samples in the upper 0 to 40 cm of the core display natural ²³⁵U/²³⁸U signatures, but measurable ²³⁶U/²³⁸U, suggesting a continued presence of contaminant uranium in the local environment.

[1] Makhijani (2000) *Inst. Energy & Environ* 5. [2] Hardesty (1991) *Miami University*. 1-70. [3] Widom (2010) *Goldschmidt Conference Abstracts*.

H₂CO₃ and its Anions and Oligomers: Updates on Experimental and Theoretical Studies

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Most chemistry textbooks assert that H₂CO₃ is the dominant species in aqueous carbonic acid. Yet, geochemists know that H₂CO₃ is present at only very low concentration in aqueous solution of carbonic acid, which is instead dominated by hydrated CO₂. H₂CO₃ has recently been characterized in cryoscopic environments and a species which is probably H₂CO₃-HCO₃⁻ has been created and characterized in aqueous solution. The dianion (HCO₃⁻)₂ has been characterized as an impurity in calcite. All the oligomeric carbonate species are stabilized by H-bonding. Oligomeric H₂CO₃ has been suggested as a form in which anthropogenic CO₂ can be sequestered. Recent quantum studies on the structures and properties of these species will be presented.