Development of a Sequential Extraction Technique to Define the Fractionation of Radioactive Elements in NIST Natural Matrix Standards

Michael Schultz and Bill Burnett, Environmental Radioactivity Measurement Facility, Department of Oceanography, Florida State University, Tallahassee, FL 32306-3048, Tel: (904) 644-6700, Fax: (904) 644-2581
e-mail: mschultz@ocean.fsu.edu, burnett@ocean.fsu.edu
and
Kenneth G. W. Inn, Joylene W. L. Thomas, and Zhichao Lin, National Institute of Standards and Technology, Ionizing Radiation Division, Building 245, Gaithersburg, MD, 20899-0001
Tel: (301) 975-5541 e-mail: keninn@micf.nist.gov

The NIST radionuclide standard reference material (SRM) program has made available natural-matrix materials that are used for the evaluation of radiochemical methods. Traditionally, these standards have been certified for total radionuclide concentration. Although a useful tool for the initial characterization of a radiologically-contaminated sample, it is clear that total concentration alone does not describe the environmental behavior of contaminating radionuclides. Characterizing and monitoring of radiologically-impacted areas and formulating optimal remediation strategies requires more sophisticated knowledge and information concerning the "partitioning" or "speciation" of radionuclides in soils and sediments. Speciation in this context is defined as an identifiable physico-chemical form (oxyhydroxide, carbonate species, organic complex, etc.) in which a specific radioactive element might be found within a given soil or sediment sample.

A workshop was convened June 13-15, 1995 at NIST to discuss the problem of how to address the speciation of radionuclides in natural matrix standard reference soils and sediments. The workshop participants comprised experts in the fields of geochemistry, soil science, transition-metal and radionuclide speciation, actinide chemistry, oceanography, as well as representatives from the EPA, DOE, NRC, and other agencies. The approach chosen for the identification of the physico-chemical associations of radionuclides in NIST standards is the application of a sequential-geoselective extraction technique. The results can be used as interpretive indicators of the environmental availability of radionuclides in the environment. When the technique is established, NIST will explore the possibility of certifying some SRM's by "fraction" along with "total" concentration.

One of the principal outcomes of the meeting was a recommended "consensus protocol" for sequential extractions (Table 1). A collaborative project between investigators at NIST and Florida State University is now underway to evaluate the optimum conditions for the recommended protocol.

Table 1. The consensus method for the speciation of radionuclides in soils and s[illegible]nents. The optimum conditions (reagent concentrations, duration of extr[illegible]on period and temperature of reaction) are to be determined by systematically varying the experimental conditions.

Fraction	Reagent	Reagent/Sample Ratio	Reagent Conc. (M)	Temp. (°C)	Time (Hrs.)
H_2O/Exchangeable	$MgCl_2$	5:1/9:1/15:1	0.1/0.4/1.0	25	1/2/4
Organics	NaOCl pH 8.5	5:1/9:1/15:1	5%	25/50/95	0.5/1/2
Carbonates	NaOAc in 25% HOAc pH 5	5:1/9:1/15:1	1	25/50/95	2/4/6
Oxides	NH_2OH•HCl in 25% HOAc pH 2 w/HNO_3	15:1	0.01/0.04/0.1	25/50/95	1/4/16
Acid/Sulfide	HNO_3	5:1/9:1/15:1	4/8/16	95	1/4/16
Residue	HF-$HClO_4$-HCl or NaOH fusion	* 5:1	na na	Fumed 500	* 3

Notes: i. Each fraction is extracted two times, followed by a rinse with extractant and then two deionized water rinses which are combined with the extractant.
ii. Each reagent-solid mixture will be separated by high-speed (>10,000g if possible) centrifugation followed by 0.1 µm filtration.

*The dissolution time is sample dependent.