Experimental observations of radionuclide uptake by colloidal and particulate humic acids obtained from 14 soils collected worldwide

Website: https://www.bco-dmo.org/dataset/738833
Data Type: experimental
Version: 1
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Project
» Biopolymers as carrier phases for selected natural radionuclides (of Th, Pa, Pb, Po, Be) in diatoms and coccolithophores (Biopolymers for radionuclides)

Contributors	Affiliation	Role
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Abstract
The uptake and binding of six particle-reactive and/or redox-sensitive radionuclides (210Pb, 234Th, 7Be, 59Fe, 237Np and 233Pa) with different organic functionalities of three size fractions.

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Dataset Description

The uptake and binding of six particle-reactive and/or redox-sensitive radionuclides (210Pb, 234Th, 7Be, 59Fe, 237Np and 233Pa) with different organic functionalities of three size fractions.

Methods & Sampling

Basically, the humic acids (HAs) from different soils were isolated and further purified according to an alkaline extraction method from the International Humic Substance Society (IHSS) (Xu et al., 2011). Briefly, the dried soil was pre-treated with a 1 M HCl solution to separate the supernatant (i.e., fulvic acid) from the soil, followed by the addition of 0.1 M KOH under N2 purging and 0.3 M K+, as KCl. Then, the supernatant was acidified to pH of 1.0 using 6 M HCl to precipitate HAs, which were pelleted by centrifugation and then suspended in a 0.1 M HCl/0.3 M HF solution overnight for five times to minimize ash content. After HF digestion, Milli-Q water was used to wash HA with the purpose of minimizing ions. HA characterization data were previously reported elsewhere (Fujitake et al., 2012) and on IHSS website (http://humic-substances.org/13c-nmr-estimates-of-carbon-distribution-in-ihss-samples/).

The HA-groundwater suspension batch experiments were conducted basically similar to a previously reported

procedure (Xu et al., 2014). In brief, 5-6 mg of the purified HAs were pre-equilibrated in artificial groundwater in the centrifuge tubes for 48 h at room temperature (20 ºC) to reach the dissolution equilibrium, since the HAs were extraceted under pH < 1 but the pH of artificial groundwater is 5.5. Then, ~50 Bq of each gamma emitting radionuclide, including 234Th, 237Np-233Pa, 210Pb, 7Be and 59Fe was added to the HA-groundwater slurry to a final volume of 4 mL. The radiolabeled HA-groundwater slurry was then mixed continuously for 7 days in the dark with an end-over-end mixer to ensure that quasi-equilibrium was attained15. After one-week period, particulate (>0.45 µm), colloidal (3 kDa to 0.45 µm), and truly dissolved (<3 kDa) phases were size fractionationated by using 0.45 µm centrifugal filter tubes followed by ultrafiltration with 3 kDa Microsep centrifugal filter tubes (Millipore). Each fraction was collected for the measurement of radionuclide activity and organic matter concentrations. All the size fractions, including the particulate, colloidal and truly dissolved phases were corrected to the same volume and geometry for the counting of 234Th, 233Pa, 237Np, 210Pb, 7Be and 59Fe activity concentrations by a Canberra ultrahigh purity germanium well gamma detector.

Concentrations of organic carbon and nitrogen in the colloidal and truly dissolved fractions were determined using a Shimadzu TOC-L analyzer. The organic carbon and nitrogen concentrations in the particulate phase of the HA-groundwater suspension were calculated as the difference between the total carbon/nitrogen contents of the added HAs and the sum of colloidal and truly dissolved phases.

Data Processing Description

Microsoft Excel Ver. 15.15; KaleidaGraph Ver. 4.1.3

BCO-DMO Processing Notes:
* added conventional header with dataset name, PI name, version date
* modified parameter names to conform with BCO-DMO naming conventions
* reorganized data from three tables into one table
* added 'nd' for STD and AVG values which were not computed
* included additional site information as provided.

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Data Files

File
radionuclide_uptake.csv (Comma Separated Values (.csv), 2.67 KB) MD5:fb679e4c9c94c53d38bd082aebaeaf49 Primary data file for dataset ID 738833

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Related Publications

FUJITAKE, N., ASAKAWA, D., & YANAGI, Y. (2012). Characterization of Soil Humic Acids by 13C NMR Spectroscopy and High Performance Size Exclusion Chromatography. BUNSEKI KAGAKU, 61(4), 287–298. doi:10.2116/bunsekikagaku.61.287
Methods
,
Results

Lin, P., Xu, C., Xing, W., Sun, L., Kaplan, D. I., Fujitake, N., … Santschi, P. H. (2018). Radionuclide uptake by colloidal and particulate humic acids obtained from 14 soils collected worldwide. Scientific Reports, 8(1). doi:10.1038/s41598-018-23270-0
Results

Xu, C., Athon, M., Ho, Y.-F., Chang, H.-S., Zhang, S., Kaplan, D. I., … Santschi, P. H. (2014). Plutonium Immobilization and Remobilization by Soil Mineral and Organic Matter in the Far-Field of the Savannah River Site, U.S. Environmental Science & Technology, 48(6), 3186–3195. doi:10.1021/es404951y
Methods

Xu, C., Zhang, S., Ho, Y.-F., Miller, E. J., Roberts, K. A., Li, H.-P., … Santschi, P. H. (2011). Is soil natural organic matter a sink or source for mobile radioiodine (129I) at the Savannah River Site? Geochimica et Cosmochimica Acta, 75(19), 5716–5735. doi:10.1016/j.gca.2011.07.011
Methods

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Parameters

Parameter	Description	Units
Sample_ID	Identifier for the sample	unitless
part_210Pb	Particulate activity percentage for Lead 210	unitless
part_234Th	Particulate activity percentage for Thorium 234	unitless
part_7Be	Particulate activity percentage for Beryllium 7	unitless
part_59Fe	Particulate activity percentage for Iron 59	unitless
part_237Np	Particulate activity percentage for Neptunium 237	unitless
part_233Pa	Particulate activity percentage for Protactinium 233	unitless
col_210Pb	Colloidal activity percentage for Lead 210	unitless
col_234Th	Colloidal activity percentage for Thorium 234	unitless
col_7Be	Colloidal activity percentage for Beryllium 7	unitless
col_59Fe	Colloidal activity percentage for Iron 59	unitless
col_237Np	Colloidal activity percentage for Neptunium 237	unitless
col_233Pa	Colloidal activity percentage for Protactinium 233	unitless
logKd_210Pb	partitioning coefficient (logKd) for Lead 210	unitless

logKd_234Th	partitioning coefficient (logKd) for Thorium 234	unitless
logKd_7Be	partitioning coefficient (logKd) for Beryllium 7	unitless
logKd_59Fe	partitioning coefficient (logKd) for Iron 59	unitless
logKd_237Np	partitioning coefficient (logKd) for Neptunium 237	unitless
logKd_233Pa	partitioning coefficient (logKd) for Protactinium 233	unitless
logKdc_210Pb	partitioning coefficient in colloidal fraction (logKdc) for Lead 210	unitless
logKdc_234Th	partitioning coefficient in colloidal fraction (logKdc) for Thorium 234	unitless
logKdc_7Be	partitioning coefficient in colloidal fraction (logKdc) for Beryllium 7	unitless
logKdc_59Fe	partitioning coefficient in colloidal fraction (logKdc) for Iron 59	unitless
logKdc_237Np	partitioning coefficient in colloidal fraction (logKdc) for Neptunium 237	unitless
logKdc_233Pa	partitioning coefficient in colloidal fraction (logKdc) for Protactinium 233	unitless
POC	Concentration of Particulate Organic Carbon (POC) after one week HAs-groundwater resuspension	miligrams per liter (mg/L)
PN	Concentration of Particulate Nitrogen (PN) after one week HAs-groundwater resuspension	miligrams per liter (mg/L)
COC	Concentration of Colloidal Organic Carbon (COC) after one week HAs-groundwater resuspension	miligrams per liter (mg/L)
CON	Concentration of Colloidal Organic Nitrogen (CON) after one week HAs-groundwater resuspension	miligrams per liter (mg/L)
pcnt_POC	Percentage of Particulate Organic Carbon (POC) after one week HAs-groundwater resuspension	unitless
pcnt_PN	Percentage of Particulate Nitrogen (PN) after one week HAs-groundwater resuspension	unitless

pcnt_COC	Percentage of Colloidal Organic Carbon (COC) after one week HAs-groundwater resuspension	unitless
pcnt_CON	Percentage of Colloidal Organic Nitrogen (CON) after one week HAs-groundwater resuspension	unitless
No	site number	unitless
HA_ID	Sample description	unitless
Site	Name of site.	unitless
Soil_Order	Soil Order	unitless
Land_use	Land use description	unitless

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Instruments

Dataset-specific Instrument Name	Beckman Coulter Allegra X-12 centrifuge
Generic Instrument Name	Centrifuge
Dataset-specific Description	Beckman Coulter Allegra X-12 centrifuge
Generic Instrument Description	A machine with a rapidly rotating container that applies centrifugal force to its contents, typically to separate fluids of different densities (e.g., cream from milk) or liquids from solids.

Dataset-specific Instrument Name	Canberra ultrahigh purity germanium well gamma detector
Generic Instrument Name	Gamma Ray Spectrometer
Dataset-specific Description	Canberra ultrahigh purity germanium well gamma detector Model GCW3024
Generic Instrument Description	Instruments measuring the relative levels of electromagnetic radiation of different wavelengths in the gamma-ray waveband.

Dataset-specific Instrument Name	Shimadzu TOC-L analyzer
Generic Instrument Name	Shimadzu TOC-L Analyzer
Dataset-specific Description	Concentrations of organic carbon and nitrogen in the colloidal and truly dissolved fractions were determined using a Shimadzu TOC-L analyzer.
Generic Instrument Description	A Shimadzu TOC-L Analyzer measures DOC by high temperature combustion method. Developed by Shimadzu, the 680 degree C combustion catalytic oxidation method is now used worldwide. One of its most important features is the capacity to efficiently oxidize hard-to-decompose organic compounds, including insoluble and macromolecular organic compounds. The 680 degree C combustion catalytic oxidation method has been adopted for the TOC-L series. http://www.shimadzu.com/an/toc/lab/toc-l2.html

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Project Information

Biopolymers as carrier phases for selected natural radionuclides (of Th, Pa, Pb, Po, Be) in diatoms and coccolithophores (Biopolymers for radionuclides)

NSF Award Abstract:

Particle-associated natural radioisotopes are transported to the ocean floor mostly via silica and carbonate ballasted particles, allowing their use as tracers for particle transport. Th(IV), Pa (IV,V), Po(IV), Pb(II) and Be(II) radionuclides are important proxies in oceanographic investigations, used for tracing particle and colloid cycling, estimating export fluxes of particulate organic carbon, tracing air-sea exchange, paleoproductivity, and/or ocean circulation in paleoceanographic studies. Even though tracer approaches are considered routine, there are cases where data interpretation or validity has become controversial, largely due to uncertainties about inorganic proxies and organic carrier molecules. Recent studies showed that cleaned diatom frustules and pure silica particles, sorb natural radionuclides to a much lower extent (by 1-2 orders of magnitude) than whole diatom cells (with or without shells). Phytoplankton that build siliceous or calcareous shells, such as the diatoms and coccolithophores, are assembled via bio-mineralization processes using biopolymers as nanoscale templates. These templates could serve as possible carriers for radionuclides and stable metals.

In this project, a research team at the Texas A & M University at Galveston hypothesize that radionuclide sorption is controlled by selective biopolymers that are associated with biogenic opal (diatoms), CaCO3 (coccolithophores) and the attached exopolymeric substances (EPS), rather than to pure mineral phase. To pursue this idea, the major objectives of their research will include separation, identification and molecular-level characterization of the individual biopolymers (e.g., polysaccharides, uronic acids, proteins, hydroquinones, hydroxamate siderophores, etc.) that are responsible for binding different radionuclides (Th, Pa, Pb, Po and Be) attached to cells or in the matrix of biogenic opal or CaCO3 as well as attached EPS mixture, in laboratory grown diatom and coccolithophore cultures. Laboratory-scale radiolabeling experiments will be conducted, and different separation techniques and characterization techniques will be applied.

Intellectual Merit : It is expected that this study will help elucidate the molecular basis of the templated growth of diatoms and coccoliths, EPS and their role in scavenging natural radionuclides in the ocean, and help resolve debates on the oceanographic tracer applications of different natural radioisotopes (230,234Th, 231Pa, 210Po, 210Pb and 7,10Be). The proposed interdisciplinary research project will require instrumental approaches for molecular-level characterization of these radionuclides associated carrier molecules.

Broader Impacts: The results of this study will be relevant for understanding biologically mediated ocean scavenging of radionuclides by diatoms and coccoliths which is important for carbon cycling in the ocean, and will contribute to improved interpretation of data obtained by field studies especially through the GEOTRACES program. This new program will enhance training programs at TAMUG for postdocs, graduate and

undergraduate students. Lastly, results will be integrated in college courses and out-reach activities at Texas A&M University, including NSF-REU, Sea Camp, Elder Hostel and exhibits at the local science fair and interaction with its after-school program engaging Grade 9-12 students from groups traditionally underrepresented.

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Funding

Funding Source	Award
NSF Division of Ocean Sciences (NSF OCE)	OCE-1356453

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