

Partition coefficient of ^{234}Th , ^{233}Pa , ^{210}Pb , ^{210}Po , and ^7Be

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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)

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Abstract

Laboratory studies were conducted to examine the sorption of selected radionuclides (^{234}Th , ^{233}Pa , ^{210}Po , ^{210}Pb , and ^7Be) onto inorganic (pure silica and acid-cleaned diatom frustules) and organic (diatom cells with or without silica frustules) particles in natural seawater and the role of templating biomolecules and exopolymeric substances (EPS) extracted from the same species of diatom, *Phaeodactylum tricornutum*, in the sorption process. The range of partition coefficients (K_d , reported as $\log K_d$) of radionuclides between water and the different particle types was 4.78–6.69 for ^{234}Th , 5.23–6.71 for ^{233}Pa , 4.44–5.86 for ^{210}Pb , 4.47–4.92 for ^{210}Po , and 4.93–7.23 for ^7Be , similar to values reported for lab and field determinations. The sorption of all radionuclides was significantly enhanced in the presence of organic matter associated with particles, resulting in K_d one to two orders of magnitude higher than for inorganic particles only, with highest values for ^7Be ($\log K_d$ of 7.2). Results further indicate that EPS and frustule-embedded biomolecules in diatom cells are responsible for the sorption enhancement rather than the silica shell itself. By separating radiolabeled EPS via isoelectric focusing, we found that isoelectric points are radionuclide specific, suggesting that each radionuclide binds to specific biopolymeric functional groups, with the most efficient binding sites likely occurring in acid polysaccharides, iron hydroxides, and proteins. Further progress in evaluating the effects of diatom frustule-related biopolymers on binding, scavenging, and fractionation of radionuclides would require the application of molecular-level characterization techniques.

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Dataset Description

Laboratory studies were conducted to examine the sorption of selected radionuclides (^{234}Th , ^{233}Pa , ^{210}Po , ^{210}Pb , and ^7Be) onto inorganic (pure silica and acid-cleaned diatom frustules) and organic (diatom cells with or without silica frustules) particles in natural seawater and the role of templating biomolecules and exopolymeric substances (EPS) extracted from the same species of diatom, *Phaeodactylum tricornutum*, in the sorption process.

The range of partition coefficients (K_d , reported as $\log K_d$) of radionuclides between water and the different particle types was 4.78–6.69 for ^{234}Th , 5.23–6.71 for ^{233}Pa , 4.44–5.86 for ^{210}Pb , 4.47–4.92 for ^{210}Po , and

4.93–7.23 for ⁷Be, similar to values reported for lab and field determinations. The sorption of all radionuclides was significantly enhanced in the presence of organic matter associated with particles, resulting in K_d one to two orders of magnitude higher than for inorganic particles only, with highest values for ⁷Be (logK_d of 7.2). Results further indicate that EPS and frustule-embedded biomolecules in diatom cells are responsible for the sorption enhancement rather than the silica shell itself. By separating radiolabeled EPS via isoelectric focusing, we found that isoelectric points are radionuclide specific, suggesting that each radionuclide binds to specific biopolymeric functional groups, with the most efficient binding sites likely occurring in acid polysaccharides, iron hydroxides, and proteins. Further progress in evaluating the effects of diatom frustule-related biopolymers on binding, scavenging, and fractionation of radionuclides would require the application of molecular-level characterization techniques.

Methods & Sampling

Partition coefficient of ²³⁴Th, ²³³Pa, ²¹⁰Pb, ²¹⁰Po, and ⁷Be

Activity concentrations of ²³⁴Th, ²³³Pa, ²¹⁰Pb, ²¹⁰Po, and ⁷Be were measured by gamma counting the 63.5, 312, 46.5, and 477.6 keV lines, respectively, on a Canberra ultra-highpurity germanium well-type detector. The ²¹⁰Po activity was analyzed by liquid scintillation counting (Beckman Model 8100 Liquid Scintillation Counter). The filter samples (> 0.2 μm particulate phase) were soaked with 1 mol L⁻¹ HCl for 20 min in a counting vial, and the filtrate samples (< 0.2 μm dissolved phase) were transferred into counting vials directly. Both filter and filtrate samples were then counted for activities of each radionuclide. All reported activities were decay and geometry corrected. ²³³Pa was added in equilibrium with ²³⁷Np. Under a wide range of environmental conditions, aqueous neptunium speciation is believed to be dominated by the pentavalent cation NpO₂⁺. Np(V) is relative soluble and tends to remain in the water phase, unlike other actinides, such as Pu and Am, which are readily adsorbed by particles in the environment (Atwood 2013). In our absorption experiments, ²³⁷Np activities could be found only in the dissolved phase for all samples, supporting the assumption that ²³⁷Np would not adsorb onto particles during the time for which decay and in-growth corrections of ²³³Pa were applied. Traditional partition coefficients (K_d) between dissolved and particulate phases were used to quantify the sorption of radionuclides onto different particles in different experimental systems. K_d was defined here as

$$K_d = A_p \times (A_d \times C_p)^{-1} \quad (1)$$

where A_p and A_d represent particulate and dissolved activities (Bq L⁻¹) of radionuclides and C_p is the particle concentration (kg L⁻¹; Honeyman and Santschi 1989; Guo and Santschi 1997).

Data Processing Description

BCO-DMO Processing Notes:

- added conventional header with dataset name, PI name, version date
- modified parameter names to conform with BCO-DMO naming conventions
- combined the two submitted tables together on the pH column, indicating percent activity and percent composition in the parameter names.

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Data Files

File
log_kd.csv (Comma Separated Values (.csv), 287 bytes) MD5:d25a173f99293b7bfe668f64add7b820
Primary data file for dataset ID 764664

Related Publications

Alvarado Quiroz, N. G., Hung, C.-C., & Santschi, P. H. (2006). Binding of thorium(IV) to carboxylate, phosphate and sulfate functional groups from marine exopolymeric substances (EPS). *Marine Chemistry*, 100(3-4), 337-353. doi:[10.1016/j.marchem.2005.10.023](https://doi.org/10.1016/j.marchem.2005.10.023)

Methods

Baskaran, M., & Santschi, P. H. (1993). The role of particles and colloids in the transport of radionuclides in coastal environments of Texas. *Marine Chemistry*, 43(1-4), 95-114. doi:[10.1016/0304-4203\(93\)90218-d](https://doi.org/10.1016/0304-4203(93)90218-d)

Methods

Chuang, C.-Y., Santschi, P. H., Ho, Y.-F., Conte, M. H., Guo, L., Schumann, D., ... Li, Y.-H. (2013). Role of biopolymers as major carrier phases of Th, Pa, Pb, Po, and Be radionuclides in settling particles from the Atlantic Ocean. *Marine Chemistry*, 157, 131-143. doi:[10.1016/j.marchem.2013.10.002](https://doi.org/10.1016/j.marchem.2013.10.002)

Methods

Chuang, C.-Y., Santschi, P. H., Jiang, Y., Ho, Y.-F., Quigg, A., Guo, L., ... Schumann, D. (2014). Important role of biomolecules from diatoms in the scavenging of particle-reactive radionuclides of thorium, protactinium, lead, polonium, and beryllium in the ocean: A case study with *Phaeodactylum tricornutum*. *Limnology and Oceanography*, 59(4), 1256-1266. doi:[10.4319/lo.2014.59.4.1256](https://doi.org/10.4319/lo.2014.59.4.1256)

Results

Duan, S., & Bianchi, T. S. (2007). Particulate and dissolved amino acids in the lower Mississippi and Pearl Rivers (USA). *Marine Chemistry*, 107(2), 214-229. doi:[10.1016/j.marchem.2007.07.003](https://doi.org/10.1016/j.marchem.2007.07.003)

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Guo, L., & Santschi, P. H. (1997). Isotopic and elemental characterization of colloidal organic matter from the Chesapeake Bay and Galveston Bay. *Marine Chemistry*, 59(1-2), 1-15. doi:[10.1016/S0304-4203\(97\)00072-8](https://doi.org/10.1016/S0304-4203(97)00072-8)

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Guo, L., Santschi, P. H., Baskaran, M., & Zindler, A. (1995). Distribution of dissolved and particulate ^{230}Th and ^{232}Th in seawater from the Gulf of Mexico and off Cape Hatteras as measured by SIMS. *Earth and Planetary Science Letters*, 133(1-2), 117-128. doi:[10.1016/0012-821X\(95\)00063-i](https://doi.org/10.1016/0012-821X(95)00063-i)

[https://doi.org/10.1016/0012-821X\(95\)00063-i](https://doi.org/10.1016/0012-821X(95)00063-i)

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Hung, C.-C., & Santschi, P. H. (2001). Spectrophotometric determination of total uronic acids in seawater using cation-exchange separation and pre-concentration by lyophilization. *Analytica Chimica Acta*, 427(1), 111-117. doi:[10.1016/S0003-2670\(00\)01196-x](https://doi.org/10.1016/S0003-2670(00)01196-x)

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Jiang, Y., Yoshida, T., & Quigg, A. (2012). Photosynthetic performance, lipid production and biomass composition in response to nitrogen limitation in marine microalgae. *Plant Physiology and Biochemistry*, 54, 70-77. doi:[10.1016/j.plaphy.2012.02.012](https://doi.org/10.1016/j.plaphy.2012.02.012)

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Quigley, M. S., Santschi, P. H., Hung, C.-C., Guo, L., & Honeyman, B. D. (2002). Importance of acid polysaccharides for ^{234}Th complexation to marine organic matter. *Limnology and Oceanography*, 47(2), 367-377. doi:[10.4319/lo.2002.47.2.0367](https://doi.org/10.4319/lo.2002.47.2.0367)

Methods

Roberts, K. A., Xu, C., Hung, C.-C., Conte, M. H., & Santschi, P. H. (2009). Scavenging and fractionation of thorium vs. protactinium in the ocean, as determined from particle-water partitioning experiments with sediment trap material from the Gulf of Mexico and Sargasso Sea. *Earth and Planetary Science Letters*, 286(1-2), 131-138. doi:[10.1016/j.epsl.2009.06.029](https://doi.org/10.1016/j.epsl.2009.06.029)

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Robinson, R. S., Brunelle, B. G., & Sigman, D. M. (2004). Revisiting nutrient utilization in the glacial Antarctic: Evidence from a new method for diatom-bound N isotopic analysis. *Paleoceanography*, 19(3), n/a-n/a.

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Sambrook, J., Fritsch, E. F., & Maniatis, T. (1989). Molecular cloning: a laboratory manual (No. Ed. 2). Cold spring harbor laboratory press. <https://isbnsearch.org/isbn/0879693096>

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Schumann, D., Ayranov, M., Stowasser, T., Gialanella, L., Di Leva, A., Romano, M., & Schuermann, D. (2013). Radiochemical separation of ⁷Be from the cooling water of the neutron spallation source SINQ at PSI. *Radiochimica Acta*, 101(8), 509-514.

Methods

Van Loon, J. C. (1985). Selected methods of trace metal analysis: biological and environmental samples. John Wiley and Sons.

Methods

Walter, H. J., Rutgers van der Loeff, M. M., & Hoeltzen, H. (1997). Enhanced scavenging of ²³¹Pa relative to ²³⁰Th in the South Atlantic south of the Polar Front: Implications for the use of the ²³¹Pa/²³⁰Th ratio as a paleoproductivity proxy. *Earth and Planetary Science Letters*, 149(1-4), 85-100. doi:10.1016/S0012-821X(97)00068-X [https://doi.org/10.1016/S0012-821X\(97\)00068-X](https://doi.org/10.1016/S0012-821X(97)00068-X)

Methods

Xu, C., Zhang, S., Chuang, C., Miller, E. J., Schwehr, K. A., & Santschi, P. H. (2011). Chemical composition and relative hydrophobicity of microbial exopolymeric substances (EPS) isolated by anion exchange chromatography and their actinide-binding affinities. *Marine Chemistry*, 126(1-4), 27-36.

doi:[10.1016/j.marchem.2011.03.004](https://doi.org/10.1016/j.marchem.2011.03.004)

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Parameters

Parameter	Description	Units
Particle_type	radionuclide type	unitless
Be	activity concentration of Be as traditional partition coefficients	kilogram per liter (kg/L)
Be_sd	Standard deviation of Be activity concentration as traditional partition coefficients	kilogram per liter (kg/L)
Pa	activity concentration of Pa as traditional partition coefficients	kilogram per liter (kg/L)
Pa_sd	Standard deviation of Pa activity concentration as traditional partition coefficients	kilogram per liter (kg/L)
Th	activity concentration of Th as traditional partition coefficients	kilogram per liter (kg/L)
Th_sd	Standard deviation of Th activity concentration as traditional partition coefficients	kilogram per liter (kg/L)
Po	activity concentration of Po as traditional partition coefficients	kilogram per liter (kg/L)
Po_sd	Standard deviation of Po activity concentration as traditional partition coefficients	kilogram per liter (kg/L)
Pb	activity concentration of Pb as traditional partition coefficients	kilogram per liter (kg/L)
Pb_sd	Standard deviation of Pb activity concentration as traditional partition coefficients	kilogram per liter (kg/L)

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Instruments

Dataset-specific Instrument Name	Pharmacia Biotech Multiphor II
Generic Instrument Name	Electrophoresis Chamber
Dataset-specific Description	For IEF (isoelectric focusing electrophoresis), a Pharmacia Biotech Multiphor II with a EPS3500 XL power supply was used.
Generic Instrument Description	General term for an apparatus used in clinical and research laboratories to separate charged colloidal particles (or molecules) of varying size through a medium by applying an electric field.

Dataset-specific Instrument Name	Canberra ultra-highpurity germanium well-type detector
Generic Instrument Name	Gamma Ray Spectrometer
Dataset-specific Description	Activity concentrations of ²³⁴ Th, ²³³ Pa, ²¹⁰ Pb, ²¹⁰ Po, and ⁷ Be were measured by gamma counting the 63.5, 312, 46.5, and 477.6 keV lines, respectively, on a Canberra ultra-highpurity germanium well-type detector.
Generic Instrument Description	Instruments measuring the relative levels of electromagnetic radiation of different wavelengths in the gamma-ray waveband.

Dataset-specific Instrument Name	Beckman Model 8100 Liquid Scintillation Counter
Generic Instrument Name	Liquid Scintillation Counter
Dataset-specific Description	The ²¹⁰ Po activity was analyzed by liquid scintillation counting (Beckman Model 8100 Liquid Scintillation Counter).
Generic Instrument Description	Liquid scintillation counting is an analytical technique which is defined by the incorporation of the radiolabeled analyte into uniform distribution with a liquid chemical medium capable of converting the kinetic energy of nuclear emissions into light energy. Although the liquid scintillation counter is a sophisticated laboratory counting system used to quantify the activity of particulate emitting (β and α) radioactive samples, it can also detect the auger electrons emitted from ⁵¹ Cr and ¹²⁵ I samples.

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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 (^{230,234}Th, ²³¹Pa, ²¹⁰Po, ²¹⁰Pb and ^{7,10}Be). 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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