29. THORIUM AND URANIUM ISOTOPES IN UPPER PLEISTOCENE SEDIMENTS OF ODP SITES 645 (BAFFIN BAY), 646, AND 647 (LABRADOR SEA)¹

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ABSTRACT

Concentrations and activity ratios of uranium and thorium isotopes $(^{234}U/^{238}U, ^{230}Th/^{232}Th)$ were determined at about 5-m intervals through the composite top 22-m sequence of Ocean Drilling Program (ODP) Hole 645 in Baffin Bay and, in the Labrador Sea, at 1-m intervals through the top 11 m of Core 84-030-003 (TWC and P) collected by the *Hudson* during a preliminary survey of Site 647, and also at about 2-m intervals through the composite top 22-m sequence of Hole 646. In the Labrador Sea, surficial sediments show unsupported ^{230}Th having a $^{230}Th/^{234}U$ activity ratio of about 3. At Site 647, a regular decrease in the $^{230}Th/^{232}Th$ activity ratio was observed downcore from about 1.2 (at 1 mbsf) to about 0.4 (at ~ 8 mbsf), through a sequence spanning over ^{18}O stages 2 through 8. The correlative thorium/uranium chronology and ^{18}O stratigraphy indicate relatively constant sedimentation rates throughout the sequence. At Site 646, down Greenland slope, and at Site 645, in Baffin Bay, highly variable uranium and thorium concentrations and isotopic ratios were observed in relation to highly variable sedimentation rates. As a whole, the lower ^{230}Th -excess observed in Baffin Bay records is indicative of very high absolute sedimentation rates. As a whole, the lower ^{230}Th -excess (over the ^{230}Th -excess (over the ^{230}Th -rain from the water column) was found.

INTRODUCTION

Since the early work of Piggot and Urry (1939, 1942), several authors developed the ²³⁰Th-excess method for studying deep sea sediments. Picciotto and Wilgain (1954), then Goldberg and Koide (1958, 1962), showed that the downcore-decreasing ²³⁰Th/²³²Th ratio could be used to estimate sedimentation rates. This approach is based on the presence of a significant amount of ²³⁴U and unsupported ²³⁰Th in surficial sediments, produced by the decay of uranium in the water column. Its validity depends on the fulfillment of several prerequisites, of which the most important are:

1. A homogeneous source of detrital particles during the sedimentation period under consideration (e.g., clays with constant "initial" uranium and thorium concentrations and isotopic ratios).

2. A constant production of 230 Th in the water column from its parent isotope (234 U; i.e., a constant uranium-content in sea water; see Turekian and Chan, 1971b; and Ku et al., 1977).

3. A constant rate of speciation (removal/precipitation) of thorium (which in turn implies a constant planktonic activity; see Fisher et al., 1987).

4. A negligible or constant uptake of uranium in the surficial sediments (with biogenic minerals, see Holmes et al., 1968; Ku, 1966; and with organic matter, see Veeh, 1967; Turekian and Bertine, 1971a; Baturin, 1973, etc.).

When these conditions are encountered, surficial sediments show an excess of ²³⁰Th activity over ²³⁴U as an effect of the "thorium rain" from the water column and possibly as a consequence of an "initial" excess of ²³⁰Th in detrital particles originating from intensively leached soil profiles on surrounding continents (Fig. 1; see Rosholt et al., 1966; Moreira-Nordemann, 1980). Therefore, the decay of the unsupported ²³⁰Th through time allows one to determine the sedimentation rates. Unfortunately, ideal conditions are rarely encountered (Coch-



Figure 1. Sketch of ²³⁰Th accumulation and decay in deep-sea sediments.

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ran, 1982; Lalou, 1982; and other review papers in Ivanovich and Harmon, 1982) and, in most cases, the excess-thorium method can simply provide empirical first-order information about sedimentation rates. Recently, precise time-marks were established for δ^{18} O stage boundaries, and generally the ¹⁸O stratigraphy provides better interpolations of deep-sea sedimentation rates. However, in the Labrador Sea and Baffin Bay basins, large-amplitude ¹⁸O changes (Hillaire-Marcel et al., and Aksu et al., this volume) in relation to direct meltwater influx from surrounding ice caps do not unequivocally correlate with the "open ocean" ¹⁸O stratigraphy; furthermore, access to time-dependent data beyond the ¹⁴C-method limits was of a great interest. On these premises, we undertook a preliminary study of uranium and thorium isotopes in cores from ODP Sites 645 (Baffin Bay), 646, and 647 (Labrador Sea; Fig. 2).

SAMPLING AND METHODS

Leg 105 Site locations in Baffin Bay and in the Labrador Sea are indicated in Figure 2. From ODP Hole 645, in Baffin Bay, four $\sim 2-0 \text{ cm}^3$ samples were analyzed and two were duplicated. These samples were collected at ~ 5 -m intervals through the composite top 22 m sequence. From the Labrador Sea, 12 samples taken at ~ 1 -m intervals through the top 11 m of Core 84-030-003 (TWC and PC) collected by the *Hudson* during a preliminary survey of Site 647, as well as 12 samples at ~ 2 -m intervals through the composite top 22-m sequence of Hole 646, were analyzed. Eleven of the latter were cut off the core sections of Hole 646B.

In all samples, the $< 120 \ \mu m$ fraction was separated by sieving after shaking in distilled water. The fine fraction was then completely dissolved, first in aqua regia, and then in a mixture of perchloric and hydrofluoric acids. Uranium and thorium isotopes present in the HCl 6N solution obtained are preconcentrated by coprecipitation with iron and aluminium hydroxides. Separation of uranium isotopes was made on anion exchange column (Dowex AG1X8, 200-400 mesh), and further purification was conducted by selective extraction on organic solvents: iron is removed by hexone from the HCl 6N solution, and specific extraction of heavy elements by thenoyltrifluoroacetone (TTA) in 4M solution in benzene from a nitric solution at pH 3 usually achieves a suitable degree of purification. Before the separation of thorium isotopes on the cation exchange column (Dowex 50WX8, 200-400 mesh), most of the aluminium was removed by dissolution in 3N NaOH solution. Final purification was conducted in the TTA-benzene solution from a nitric solution at pH 1. Uranium and thorium sources were electrodeposited from an ethyl alcohol solution in slightly acid form. Resolution of alpha spectra (detectors were silicon surface barrier-type, 100 µm depletion depth, and 350 mm² area) was better than 50 KeV.

This chemical procedure is relatively conventional, as can be concluded from Lally's review paper (1982). A less usual feature is the choice of the spike: the conventional $^{232}U_{-228}$ Th couple at secular equilibrium was added to a recent 234 Th preparation. The 234 Th is a natural radioactive isotope, which is thus present in samples, but there is no problem of interference with the 234 Th activity of the sample because 234 Th is a β -emitter, and a large excess (three or four orders of magnitude) is used.

The ²³⁴Th is isolated from the uranium solution once it has been cleaned of its thorium isotopes, and then allowed to produce its daughters. After two or three months, the ²³⁴Th/²³⁸U activity ratio is high enough to obtain a preparation usable for 40 or 50 samples. This ²³⁴Th solution, checked for its null alpha activity, is evaporated to dryness in a pyrex erlen into which the 232U-228Th alpha spike has been poured. After homogeneization under ultrasonic conditions, the triple spike is ready to be introduced at the beginning of each analysis. One weighed quantity of ~100 μ L (micropipetted) is allowed to dry at laboratory temperature on a platinum disk and is used as an external reference for each thorium source. This reference disk is β -counted at least 10 times, and at least 600,000 counts are counted each time. The β -activity of each thorium source is checked twice, with a total measured activity greater than 600,000 counts. β -activity is registered with a silicon surface barrier detector partially depleted (300 µm depth) and protected from ambient radioactivity by a lead shield. The background is equal to 2 cpm. Under these conditions, the thorium chemical efficiency is determined with a standard deviation equal to 2‰. The source geometry is constant, as ensured by the electrodeposition. The alpha counting efficiency, necessary for knowing the total efficiency of the thorium, was determined by analyses of uraninite in secular equilibrium for 238 U- 234 U- 230 Th isotopes.

This method presents one disadvantage: one must prepare the spike every two or three months. But two advantages are significant: (1) the risk of a wrong thorium efficiency calculated in the case of disequilibrium between ²³²Th and ²²⁸Th is eliminated and (2) even in the case of equilibrium greater accuracy is obtained for thorium efficiency determination.

URANIUM AND THORIUM ISOTOPES IN LABRADOR SEA

At Site 647 in south-central Labrador Sea (Table 1), uniform ²³²Th concentrations ($X = 4.89 \pm 0.62 \,\mu g/g$) are observed downcore, indicating a homogeneous source of detrital particles. The uranium uptake by organic matter and biogenic minerals was apparently low: an average uranium concentration of 0.62 \pm 0.08 μ g/g characterizes the studied sequence, with two exceptions (UQT-532 and UQT-545; Table 1). These two samples show higher proportions of uranium (>1 μ g/g). Their ²³⁴U/ 238 U activity ratio (respectively 1.07 \pm 0.02 and 1.16 \pm 0.03) confirms the uranium uptake from seawater. Actually, all the other samples show $^{234}U/^{238}U$ ratios <1 (X = 0.94 ± 0.05), typical of marine clays that have undergone preferential ²³⁴Uleaching during sedimentation. As expected, the 230Th/232Th activity ratio decreases regularly downcore (Figs. 3 and 4). Very minor changes in the sedimentation rate may be inferred between isotopic stages 7 and 5 for an interpolated average value of $\sim 5 \times 10^{-3}$ cm/yr.

At Site 646, a more irregular pattern is observed (Table 2). This can be seen, for example, by the ²³⁸U/²³²Th activity ratio variations downcore ($X = 0.66 \pm 0.26$) and also by the wide fluctuations in the ²³⁰Th-excess values (above ²³⁴U; Figs. 5 and 6). By comparison with Site 647, the latter is also characterized by lower ²³²Th concentrations ($X = 3.3 \pm 0.6 \,\mu g/g$) and a proportionally higher uranium uptake with 234U/238U activity ratios more often exceeding the equilibrium value of 1. As a whole, sedimentation rate changes from $\sim 5 \times 10^{-3}$ to $\sim 13 \times 10^{-3}$ cm/yr, about a mean value of $\sim 9 \times 10^{-3}$ cm/yr (Fig. 7 and Table 3), may be calculated using time-marks obtained from the 18O-stratigraphy (Aksu et al., this volume) and assuming an initial ²³⁰Th/²³²Th ratio of about 0.36 in the detrital particles before any ²³⁰Th increment from the "thorium rain." This initial value equal to 0.36 is suggested by the lowest isotopic ratios observed in the regional deep-sea clays, notably those of Baffin Bay (see below: Sample UQT-490; Table 4), where very high sedimentation rates result in an almost negligible ²³⁰Th uptake from the ²³⁰Th-rain.

At both sites, some similar geochemical processes appear. All samples having relatively high proportions of uranium (vs. ²³²Th) show correlatively higher ²³⁴U/²³⁸U ratios that occasionally (e.g., UQT-532) reach the typical value of seawater (~ 1.15 ; Thurber, 1962). In such samples, uranium uptake from seawater (with organic matter or biogenic minerals) can be inferred. Indeed, these differ from all other samples with a uranium content essentially related to that of the detrital particles: these samples have ²³⁴U/²³⁸U activity ratios lower than 1.0, in a few cases, as low as ~0.84. The ²³⁴U-preferential leaching certainly occurs here when the clay particles are in suspension and during the sedimentation phase, rather than during their early continental genesis and history, considering the climate in the surrounding continents, which is cold and unfavorable for efficient leaching processes. Because of the large sampling intervals, it is hazardous to correlate the high uranium-content samples with climatic episodes as a whole. However, the highest value falls into isotopic stage 5 (according to ¹⁸O data; de Vernal and Hillaire-Marcel, 1987), i.e., into an interglacial period when higher primary productivity and organic sedimentation may be ex-



Figure 2. Location map of Sites 645, 646, and 647.

pected. This trend apparently contradicts that observed by Kalil (1976).

Another characteristic of the surficial sediments in the Labrador Sea may also be mentioned. At both sites, a large difference exists between the ²³⁰Th-excess (²³⁰Th over equilibrium with ²³⁴U) measured in surficial sediments (~ 2.8 dpm/cm at Site 646

and ~2.6 dpm/cm at Site 647) and the expected theoretical value. The latter can be calculated from the ²³⁰Th production in the overlying water column and the average sedimentation rates (for Site 646, ~9 cm × 10^{-3} yr; for Site 647, ~5 cm × 10^{-3} yr) interpolated above from the ¹⁸O-stratigraphy. The theoretical ²³⁰Th-excess calculates to ~0.85 dpm/cm and ~1.7 dpm/cm,

Table 1. Thorium and uranium concentrations and isotope activity ratios at Site 647.

Field ¹ number	Laboratory number	²³² Th (μg/g)	²³⁸ U (μg/g)	$\frac{^{234}U}{^{238}U}$	$\frac{^{238}U}{^{232}Th}$	$\frac{^{230}\mathrm{Th}}{^{232}\mathrm{Th}}$	$\frac{^{234}U}{^{232}Th}$	²³⁰ Th (dpm/g)	²³⁴ U (dpm/g)
3T-101/103	UQT-530	4.67	0.73	0.92	0.47	1.24	0.43	1.42	0.50
		±0.20	±0.02	±0.03	± 0.02	±0.03	0.02	±0.06	±0.01
3T-186/188	UQT-531	4.90	0.67	0.97	0.41	1.08	0.40	1.30	0.48
		±0.30	± 0.02	±0.03	±0.03	±0.03	±0.03	± 0.08	± 0.01
3P-103/105	UQT-532	5.03	1.49	1.16	0.90	0.95	1.04	1.18	1.29
		±0.35	±0.04	±0.03	±0.07	±0.03	± 0.08	±0.08	±0.03
3P-204/206	UQT-533	4.15	0.55	1.00	0.40	0.85	0.40	0.87	0.41
		±0.21	± 0.01	±0.03	± 0.02	± 0.03	± 0.02	±0.04	±0.01
3P-302/304	UQT-534	5.70	0.76	0.89	0.40	0.63	0.36	0.88	0.51
		±0.17	±0.01	± 0.02	± 0.01	±0.01	±0.01	±0.03	± 0.01
3P-400/402	UQT-535	4.27	0.53	0.94	0.38	0.70	0.36	0.74	0.38
		±0.23	±0.01	±0.03	±0.02	±0.02	±0.02	±0.04	±0.01
3P-500/502	UQT-543	4.73	0.58	0.90	0.37	0.54	0.33	0.63	0.39
1917509000000000000000000000000000000000	Section - Constraint Constraints	±0.13	±0.01	±0.03	± 0.01	± 0.01	±0.01	±0.02	± 0.01
3P-603/605	UQT-544	3.93	0.53	0.84	0.41	0.53	0.34	0.51	0.33
		±0.10	±0.01	±0.03	±0.02	±0.01	±0.01	±0.01	±0.01
3P-696/698	UQT-545	5.49	1.07	1.06	0.59	0.54	0.63	0.72	0.85
	15	±0.16	± 0.02	± 0.02	±0.02	±0.01	± 0.02	±0.02	±0.01
3P-801/803	UQT-546	4.42	0.56	0.99	0.38	0.42	0.38	0.46	0.41
	1403 (1 997) 2 (1979) (1979)	±0.13	± 0.01	±0.03	± 0.01	±0.01	±0.01	±0.02	±0.01
3P-911/913	UQT-547	5.49	0.66	0.97	0.36	0.43	0.35	0.57	0.48
		±0.16	± 0.01	±0.03	±0.01	±0.01	±0.01	±0.02	±0.01
3P-1008/10	UQT-548	5.92	0.70	0.94	0.36	0.44	0.34	0.64	0.49
	13	± 0.18	± 0.02	± 0.04	± 0.02	± 0.01	± 0.02	± 0.02	± 0.02

¹ T = Trigger weight core; P = Piston core; sampling interval in centimeters below seafloor. Error data represent one σ from counting statistics.

respectively, at Sites 646 and 647. These values are based on the 230 Th "classical" scavenging rate of

$$2.2 \text{ dpm/}D \text{ km/cm} \times 10^3 \text{ yr}$$
 (1)

where D = the water depth (in kilometers)(see Lalou, 1985). The ²³⁰Th-excess over that accounted by the decay of ²³⁴U in the water column must be introduced into the sediment without the parent isotope, i.e., without uranium uptaking. Two probably complementary causes can be invoked. Beyond the particle ²³⁰Th/ ²³⁴U disequilibrium due to uranium leaching, a large proportion of unsupported ²³⁰Th is probably incorporated into the sediment as an effect of organic activity. Algae (Mann and Fyfe, 1984) and bacteria (Beveridge et al., 1983) are known to concentrate uranium from ambient water. Recently, Fisher et al. (1987) also showed significant thorium uptake by nano- and picoplankton. Plankton and their debris are oxidized while sinking and depositing. When ²³⁰Th remains trapped and accumulates within the sediment, uranium may still be released into the ambient water. Consequently, surficial sediments may receive a ²³⁰Th-excess above that due to the classical ²³⁰Th-rain from the decay of dissolved ²³⁴U in the water column. The relative weight of this bio-induced ²³⁰Th accumulation increases where the water column depth decreases and also at places where the primary productivity increases: i.e., larger anomalies should be observed in shallower sites and near neritic environments. Moreover, because turbidity as a whole is higher in marginal basins compared to "open oceans," ²³⁰Th scavenging rates should increase proportionally. A horizontal flux of ²³⁰Th, from the latter to the former, should be the consequence of this difference. Beyond other considerations (see Cochran, 1982, for a review of literature), this phenomenon could account at least for some negative anomalies observed in 230Th excesses in open oceans and, in this case, for the positive anomaly of the Labrador Sea surficial sediments. Both effects, i.e., the bio-induced ²³⁰Th accumulation and the ²³⁰Th scavenging rates, should increase in shallower basins. In fact, this is the case in the Labrador Sea, where the ac-

terglacial successions (de Vernal and Hillaire-Marcel, 1987), the bio-induced ²³⁰Th-excess may have changed drastically through time. Consequently, the latter might here be an indicator of paleoproductivities rather than of sedimentation rates. **URANIUM AND THORIUM ISOTOPES IN BAFFIN BAY** At Site 645, the sediments are characterized by ²³²Th concentrations as high as 17 $\mu g/g$ (Sample UQT-490; Table 4). Although deep-sea clays are known to have an average thorium content of ~10 $\mu g/g$ (Lalou, 1982), we have already seen that the Labrador sediments are much poorer in thorium (with concentrations of ~3 to 5 $\mu g/g$). From this viewpoint, the sedimen-

the Labrador seements are much pooler in thoritain (with concentrations of ~3 to 5 μ g/g). From this viewpoint, the sedimentation history of Baffin Bay has to be considered independently from that of the Labrador Sea. Relatively high thorium contents are also observed in shallow marginal environments of the Laurentide Ice Sheet (e.g., in Lake Deschaillons varves of St. Lawrence Lowland; see Hillaire-Marcel and Causse, 1987), where the fine fraction originates from the glacial floor. On sedimentological grounds, the Baffin Bay sediments show large similarities with the latter: notably their coarseness and mineralogical composition (see Aksu and Piper, 1979).

tual ²³⁰Th-excess in surficial sediments vs. the theoretical value

increases from a ratio of ~ 1.5 at Site 647 in the central Labrador Sea (depth ~ 3850 m) to ~ 3.3 at Site 646, on the Greenland slope (depth ~ 3450 m). As a result, any attempt to calculate sedimentation rates from measured ²³⁰Th-excesses and theoreti-

cal ²³⁰Th-rains in the water columns fails. According to Lalou

(1985), the ²³⁰Th anomalies described above are considered to be

uniform through time at any specific site. Hence, when few time controls exist (e.g., through ¹⁸O-stage boundaries), short-term

changes in the sedimentation rate can be determined from ²³⁰Th-

excess fluctuations, the mean sedimentation rate being provided otherwise. However, we would like to add a note of caution: in

marine basins such as the Labrador Sea, where primary produc-

tivity experienced significant changes in relation to glacial/in-





As a whole, uranium uptake by surficial sediments seems negligible: uranium/thorium mass ratios of 0.1 to 0.2 were measured. Whereas the very modern surficial sediments show a ²³⁰Th-excess (~2.5 dpm/g above ²³⁴U activity; Sample UQT-675) not unlike that observed on the Greenland slope in the Labrador Sea, all other samples are characterized by 230Th/234U activity ratios near the equilibrium value (within the standard deviation: Fig. 8). This difference may be attributed to the very large sedimentation rate change that occurred there at the Holocene glacial/interglacial transition. Actually, in most piston cores collected in the deepest part of the Bay, the post-glacial sediments were rarely recovered and they were represented by no more than a few inches in trigger-weight cores. On the contrary, a high pre-Holocene mean sedimentation rate of ~ 8 to 10 \times 10⁻³ cm/yr was interpolated between ¹⁸O-stage boundaries and a few ¹⁴C (AMS) ages of foraminifers (de Vernal et al., 1987; Hillaire-Marcel et al., this volume). During most of the sedimentary episode sampled here, the primary productivity was extremely low, if not nonexistent, in this basin notably because of a dense sea-ice cover (de Vernal, 1986). As a result, the only source of ²³⁰Th-excess at the surface of the sediments was the ²³⁰Th-rain from the decay of dissolved ²³⁴U in the water column. Using the ²³⁰Th-scavenging rate formula mentioned above, and assuming a sedimentation rate of $\sim 10 \times 10^{-3}$ cm/yr, the ²³⁰Thexcess activity (above that of ²³⁴U) in the surficial sediments

when deposited was ~0.5 dpm/cm³. Because ²³⁴U-activities of ~4.5 dpm/cm³ are actually measured in the sediments, the ²³⁰Th-excess did not originally exceed about 10% (above the ²³⁴U-supported thorium 230). Within less than 10,000 yr, this excess decreased to a value falling within the analytical standard deviation around the equilibrium value. We have to conclude that, unfortunately, the thorium/uranium method is of no help here in establishing sedimentation rate changes through time, and *a fortiori* that it cannot provide any assessment of the absolute rate values. One can conclude that sedimentation rates (from ~8 to 20 cm × 10⁻³ yr; Hillaire-Marcel et al., this volume) were high enough to prevent any significant accumulation of unsupported ²³⁰Th, at least until the last glacial/interglacial transition.

CONCLUSIONS

This study confirms, once again, that uranium and thorium concentrations and isotopic ratios often do not provide direct time controls in deep-sea sediments (this statement is certainly valid for all types of sediments). Rather, these should be considered as geochemical indicators that depend on time and environment. These Leg 105 sites also confirm that sedimentation rates can be estimated, provided the continental and margin influences are negligible. Indeed, at Site 647 in the southeastern central Labrador Sea, the regular decrease of ²³⁰Th/²³²Th ratios downcore can be compared to that observed in North Atlantic deep-sea cores. This reflects minor changes in sedimentation rates. At other sites, more influenced by the surrounding ice fluctuations, major changes in sedimentation rates were observed throughout the sampled sequences. On the whole, both Sites 646 and 645 experienced very high mean sedimentation rates, which in Baffin Bay (Site 645) resulted in a negligible initial ²³⁰Th-excess (over its parent isotope). Fortunately, in the Labrador Sea, down Greenland slope (Site 646), a large ²³⁰Thexcess over the "classical 230Th-rain" from the water column exists. This possibly bio-induced excess allows us to evaluate the sedimentation rate changes with some reservations (notably due to possible variations in primary productivity through time, which led to fluctuating initial ²³⁰Th-excesses). Actually, the most significant outcome of this study concerns the peculiarities of ²³⁰Th geochemistry in such marine basins that, despite their depth, constitute marginal-type environments. Ongoing studies on organic matter vs. ²³⁰Th contents in surficial sediments from the Labrador Sea should help to clarify these peculiarities.

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Figure 4. The 230 Th/ 232 Th vs. 234 U/ 232 Th activity ratio variations down Core HU-84-030-003, collected during the *Hudson* Site 647 survey.

Field number	Laboratory number	²³² Th (µg/g)	²³⁸ U (µg/g)	²³⁴ U ²³⁸ U	$\frac{^{238}U}{^{232}Th}$	$\frac{^{230}Th}{^{232}Th}$	$\frac{^{234}U}{^{232}Th}$	²³⁰ Th (dpm/g)	²³⁴ U (dpm/g)
		± 0.12	±0.01	±0.03	±0.01	±0.03	±0.01	±0.04	± 0.01
646B-1-3	UQT-665	4.24	0.55	0.93	0.40	1.28	0.37	1.33	0.38
		±0.12	±0.01	±0.03	±0.01	±0.03	±0.01	± 0.04	± 0.01
646B-1-4	UQT-666	2.81	0.45	1.05	0.49	1.21	0.51	0.84	0.35
		±0.09	±0.01	±0.03	±0.02	±0.04	±0.02	±0.03	± 0.01
646B-2-3	UQT-668	3.17	1.14	1.10	1.09	1.74	1.20	1.36	0.94
		±0.09	±0.02	±0.03	±0.04	±0.05	±0.04	±0.04	± 0.02
646B-1-6	UQT-667	2.66	0.33	0.83	0.38	1.28	0.32	0.84	0.21
		± 0.08	± 0.01	±0.04	±0.02	±0.04	±0.01	±0.02	±0.01
646B-2-4	UQT-669	2.63	0.80	1.00	0.92	1.44	0.92	0.93	0.60
		±0.09	± 0.02	±0.03	±0.04	± 0.04	±0.04	±0.03	± 0.01
646B-2-5	UQT-670	3.64	0.90	1.13	0.75	1.13	0.85	1.01	0.76
		± 0.11	± 0.02	±0.03	±0.03	± 0.04	±0.03	±0.03	± 0.02
646B-2-6	UQT-671	4.46	0.48	0.90	0.33	0.84	0.30	0.92	0.32
		±0.13	±0.01	±0.03	±0.01	±0.02	± 0.01	±0.03	±0.01
646B-3-1	UQT-672	3.24	0.91	1.06	0.85	1.03	0.90	0.82	0.72
		± 0.10	± 0.02	±0.03	±0.03	±0.03	±0.03	±0.02	± 0.02
646B-3-2	UQT-673	2.85	0.80	1.02	0.85	1.18	0.87	0.82	0.61
		± 0.10	± 0.02	±0.03	±0.04	±0.04	± 0.04	±0.03	±0.01
646B-3-4	UQT-674	2.94	0.80	1.10	0.83	1.05	0.91	0.76	0.66
		±0.09	±0.02	±0.03	±0.03	±0.03	±0.04	± 0.02	± 0.02
6B36-98-10	UQT-492	6.14	0.92	1.01	0.46	0.55	0.46	0.83	0.70
		±0.40	± 0.02	±0.03	±0.03	±0.01	±0.03	± 0.05	± 0.02

Table 2. Thorium and uranium concentrations and isotope activity ratios at Site 646.

Error data represent one σ from counting statistics.





Figure 6. Log $(^{230}Th/^{232}Th)$ changes downcore at Site 646.





Table 3. Initial ²³⁰Th/²³²Th values in sediments from Site 646 calculated using approximative control provided by ¹⁸O-stratigraphy and indicative values of ²³⁰Th-excess/²³²Th activity ratio and of sedimentation rate changes.

Field Laboratory Depth number number (cm)		Chronology from ¹⁸ O-stratigraphy	$\begin{bmatrix} 230 \text{Th} \\ \hline 232 \text{Th} \end{bmatrix}^1$	$\left[\frac{230_{\text{Th}}}{232_{\text{Th}}}\right]^2$	Sedimentation rate (cm/k.y.) ³	
646B-1-1	UQT-664	100	8,000 ± 2,000	1.42 ± 0.02	1.06	~ 12.7
646B-1-3	UQT-665	400	$32,000 \pm 3,000$	1.59 ± 0.03	1.23	~10.9
646B-1-4	UQT-666	540	$40,000 \pm 5,000$	1.53 ± 0.05	1.17	~11.5
646B-2-3	UQT-668	770	$65,000 \pm 5,000$	2.18 ± 0.05	1.82	~7.4
646B-1-6	UQT-667	860	$78,000 \pm 5,000$	2.29 ± 0.10	1.93	~7.0
646B-2-4	UQT-669	980	$110,000 \pm 10,000$	2.35 ± 0.14	1.99	~6.8
646B-2-5	UQT-670	1093	$130,000 \pm 5,000$	1.75 ± 0.04	1.39	~9.7
646B-2-6	UQT-671	1207	$150,000 \pm 10,000$	2.48 ± 0.21	2.12	~6.3
646B-3-1	UQT-672	1559	$180,000 \pm 15,000$	1.55 ± 0.09	1.19	~11.3
646B-3-2	UQT-673	1718	$215,000 \pm 15,000$	3.06 ± 0.32	2.70	~ 5.0
646B-3-4	UQT-674	1999	$220,000 \pm 10,000$	1.79 ± 0.08	1.43	~9.4
6B36-98-10	UQT-492	2200	$260,000 \pm 20,000$	$1.44~\pm~0.20$	1.08	~12.5
Mean values				1.95 ± 0.49	1.6 ± 0.5	~9

¹ Extrapolate initial values of ²³⁰Th/²³²Th activity ratios, calculated according to ¹⁸O-stratigraphy chronology.
² Indicative initial values of ²³⁰Th-excess/²³²Th activity ratios, related to "²³⁰Th-rain" (assuming a uniform ²³⁰Th/²³²Th activity ratio equal to 0.36 for detrital particles).
³ [mean sedimentation rate from ¹⁸O data] × [mean (²³⁰Th-excess/²³²Th) initial ratio] ÷ [calculated (²³⁰Th-excess/²³²Th) initial ratio].

Error data represent one σ from counting statistics.

Table 4. Thorium and uranium concentrations and isotope activity ratios at Site 645.

Field number	Laboratory number	²³² Th (µg/g)	²³⁸ U (µg/g)	²³⁴ U ²³⁸ U	$\frac{^{238}\text{U}}{^{232}\text{Th}}$	$\frac{^{230}\mathrm{Th}}{^{232}\mathrm{Th}}$	$\frac{^{234}U}{^{232}Th}$	²³⁰ Th (dpm/g)	²³⁴ U (dpm/g)	Depth (cm)
		±0.41	±0.03	±0.03	± 0.02	±0.04	± 0.02	±0.16	± 0.02	
5G14-59-61	UQT-490	24.12	2.06	1.05	0.26	0.36	0.27	2.11	1.61	660
		±7.24	±0.06	±0.03	± 0.08	± 0.01	± 0.08	±0.64	±0.05	
5G14-59-61	UQT-503	17.29	1.73	0.99	0.30	0.35	0.30	1.49	1.27	660
		±1.07	±0.19	±0.12	±0.04	± 0.01	± 0.04	±0.09	± 0.14	
5F32-34-36	UQT-488	10.80	2.32	1.01	0.65	0.51	0.66	1.34	1.74	1500
		±2.66	±0.05	± 0.02	±0.16	± 0.01	±0.16	±0.33	± 0.04	
5F35-76-78	UQT-491	10.95	1.66	1.16	0.46	0.53	0.53	1.42	1.43	2000
		±2.17	±0.05	±0.03	±0.09	±0.01	±0.11	±0.28	±0.04	
5F35-76-78	UQT-504	10.77	2.19	0.85	0.62	0.53	0.52	1.41	1.38	2000
		±0.45	± 0.27	± 0.13	± 0.08	± 0.01	± 0.07	± 0.06	± 0.18	

Error data represent one σ from counting statistics.



Figure 8.The ²³⁰Th/²³⁴U activity ratios downcore at Site 645.