

THE EFFECT OF ISOTOPIC EQUILIBRATION TIME ON THE
DETERMINATION OF ^{234}Th IN SEAWATER

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This study is aimed at the determination of the isotopic equilibration time that is needed when measuring ^{234}Th activity in seawater. It is found that a period of at least 4 h should be given after the yield tracer is added to acidified seawater.

INTRODUCTION

The distribution of naturally occurring ^{234}Th ($T=24.2$ d) in seawater has been used by oceanographers to study scavenging phenomena in the ocean¹⁻⁵. To measure ^{234}Th activity in seawater, it is common practice to spike seawater samples with known amounts of other Th isotope as yield tracer (e.g., ^{230}Th , ^{229}Th). After the yield tracer is added to the acidified seawater, a time span of hours is empirically given to allow the isotopic equilibrium, then procedures of preconcentration, purification, and counting are carried out. How-

ever, the length of time for isotopic equilibration between ^{234}Th and Th yield tracer has not been investigated. The goal of this study is to determine how long an isotopic equilibration time is required for measuring ^{234}Th in seawater.

EXPERIMENTAL

A large volume (100 liter) of seawater obtained from the subsurface ocean off southwestern Taiwan was filtered through 0.45 μm glass fiber filters into a garbage can. Salinity of the filtrate is 32.344 psu measured by AutoSal[®] salinometer. The seawater is evenly divided into 14 subsamples of 5 liter each and acidified to pH~2 with 5 ml concentrated HCl. The acidified samples were stored for ~1 year to allow radioactive equilibrium of ^{234}Th and ^{238}U .

The samples were spiked with ^{230}Th yield tracer along with 15 mg Fe carrier (as FeCl_3) and homogenized by stirring. Isotopic equilibration times ranging from 30 min to 30 h were applied for different samples. About 5 ml concentrated NH_4OH was then added to raise the pH to precipitate $\text{Fe}(\text{OH})_3$. The $\text{Fe}(\text{OH})_3$ precipitate was separated from the supernatant by centrifugation and dissolved in concentrated HCl. Separation of uranium and thorium was accomplished by passing the solution through an anion exchange column (1X8 Dowex resin) pre-conditioned with 9N HCl. Thorium passed through the column while uranium and iron were retained by the resin.

Thorium samples were purified further by evaporating the sample on a hot plate and passing the sample through three anion exchange columns pre-conditioned with 8N HNO_3 . The sample was evaporated down to one

drop and was ready for extraction. ^{234}Th and the yield tracer, ^{230}Th , were extracted into a TTA (thenoyltri-fluoroacetone)-benzene solution and stippled on a stainless steel disc.

The activities of ^{234}Th were counted by a low background (<0.3 cpm) anticoincidence counter (Tennelec LB-5100) via its β -emitting daughter ^{234}Pa . Spiked ^{230}Th was counted using silicon surface-barrier detectors (EG&G Ortec 576). The counting efficiencies of the α -detectors were calibrated against NIST traceable ^{241}Am (Isotope Products Laboratory 387-67-2-2) and ^{230}Th (Isotope Products Laboratory 387-67-3) standard plates. Activities of ^{234}Th reported here were corrected back to the time of spiking after the ingrowth of ^{234}Th from ^{238}U was subtracted.

RESULTS AND DISCUSSION

The results of the experiment are shown in Fig. 1. ^{234}Th activities were reported as dpm l^{-1} . Error bars in Fig. 1 represent uncertainties ($\pm 1\sigma$) from the propagation of counting errors. Since the samples were stored for ~ 1 year so that secular equilibrium between ^{234}Th and ^{238}U should be reached. Hence, the expected ^{234}Th activity is 2.290 dpm l^{-1} calculated from the salinity- ^{238}U relationship of Ku et al.⁶.

Except for the data for 8 h all samples with isotopic equilibration times longer than 4 h have the same activities with ^{238}U within the range of counting uncertainties. The average ^{234}Th activity for these samples is $2.281 \pm 0.054 \text{ dpm l}^{-1}$, which agrees with expected value. The relative standard deviation of $\sim 3\%$ indicates that the analytical uncertainty is

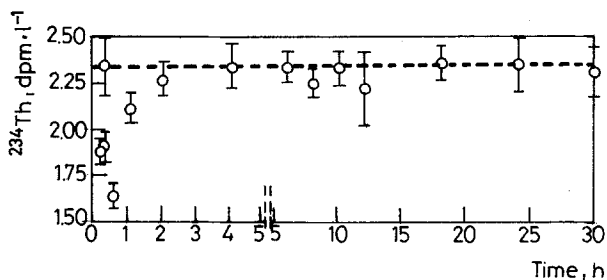


Fig. 1. Results of ^{234}Th activity vs. isotopic equilibration time. The dashed line represents expected ^{234}Th activity calculated from the salinity- ^{238}U relationship

lower than the counting uncertainty (~6%). Those samples with <4 h isotopic equilibration time tend to be lower than expected activities. Accordingly, we suggest that a time span of at least 4. h should be given for ^{234}Th measurement in seawater.

It should be noted that the samples had been filtered through 0.45 μm membrane filters, the yield tracer (^{230}Th as nitrate solution) was essentially equilibrated with ^{234}Th in colloids and/or dominant Th^{4+} ions⁷ in seawater sample. If unfiltered seawater was to be measured, an even longer period of time might be needed because exchange processes for ^{234}Th incorporated in particles are expected to take place before isotopic equilibration.

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