



Natural uranium-series radionuclide inventories in coastal and oceanic waters of the south-western Pacific - insights into trace metal flux and removal pathway analysis

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Summary

Particulate scavenging of trace metals plays a major role in determining their ecosystem flux and incident dissolved concentrations. Differences in the half-lives and biogeochemical behaviour of natural uranium series radioisotope pairs (eg. $^{238}\text{U}/^{234}\text{Th}$, $^{210}\text{Pb}/^{210}\text{Po}$) allow their application as oceanic process tracers. Coincidental measurements of dissolved and particulate trace element concentrations and inventories of radionuclides in the Noumea coral lagoon and adjacent offshore waters were used to quantify water column flux rates and provide insights on removal pathway analysis. Understanding prevailing pathways and respective flux rates of pollutants in specific coastal and oceanic systems will assist to establish the fate and consequence of pollutants and allow sustainable management strategies to be developed.

Introduction

The distribution, flux rates and pathways of trace metals, radionuclides and other chemical species in the marine environment are often extremely complex. Factors such as differences in sources, geochemistry and the influence of numerous in situ processes (eg. biological productivity, upwellings, resuspension) vary greatly and are important determinants of incident concentrations.

Biogeochemical behaviour

Differences in geochemical behaviour (Table 1) manifest as differences in environmental behaviour.

Geochemical Behaviour	Trace Metals	Radionuclides
Conservative	gold, rubidium, tungsten	uranium
Nutrients	arsenic, cadmium, copper, iron, silver, nickel	polonium
Scavenged	aluminium, arsenic, cobalt, copper, iron, manganese	thorium
Redox-controlled	arsenic, chromium, iron, manganese, selenium	iodine

Table 1. Oceanic geochemical behaviour of some trace metals and radionuclides.

Several elements may also fall into more than one behavioural category (eg. iron) or oscillate between categories as a result of changes in redox conditions

(eg. arsenic) and speciation (eg. copper), further complicating their redistribution. Both natural and pollutant chemical species introduced to the marine environment may either remain benign in solution or undergo physiological uptake by biota, but most often associate with colloids and fine particles which subsequently undergo aggregation, sedimentation and removal to the sea floor. These particles may be clay minerals, carbonates, quartz, feldspar, ferromanganous oxyhydroxides or organic solids formed by either chemical or biological processes. Solution to particulate phase transitions, by any mechanism significantly influence the incident residence times and consequently the environmental impact of many chemical species.

Tracing removal rates and mechanisms

An understanding of several oceanic processes has been developed through studies using naturally occurring uranium series radionuclides as tracers (Cochran, 1982; Szymczak, 1997). Particularly suited to studies of chemical scavenging in marine systems is the observed disequilibrium in $^{234}\text{Th}/^{238}\text{U}$ activities, where ^{234}Th is the daughter isotope of ^{238}U (Fig.1). Differences in the half-life and biogeochemical behaviour of natural uranium series radioisotopes ^{238}U (conservative) and ^{234}Th (scavenged) allow their application as oceanographic process tracers. This tracer pair provides a mechanism to trace removal processes with temporal ranges of days to months.

Assessments of the in situ inventories of $^{234}\text{Th}/^{238}\text{U}$ have been previously applied to determine the residence times and flux of particles in seawater

(Cochran, 1982) and interpret the behaviour of several dissolved trace elements (Szymczak, 1997; Jeffree & Szymczak, 2000).

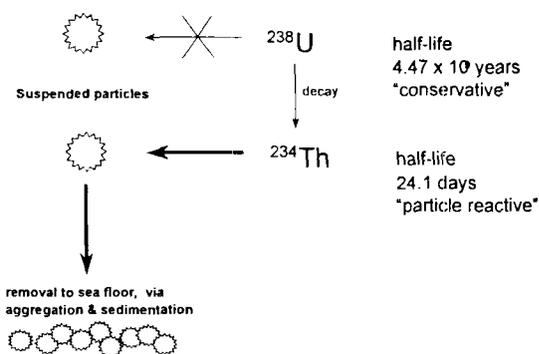


Figure 1. Physicochemical behaviour of ^{234}Th and ^{238}U radionuclides in seawater.

Other members of the ^{238}U decay series, ^{210}Po and ^{210}Pb are also useful isotopes for gaining an understanding of surface ocean particle transport processes. Analyses of $^{210}\text{Po}/^{210}\text{Pb}$ inventories are well suited to these studies as their rates of input to the euphotic zone are known and they vary appreciably in relation to particle reactivity and bioavailability (Fig. 2). Oceanic surface water inventories of ^{210}Po and ^{210}Pb (Peck and Smith, 2000) identified depletion of ^{210}Po (the daughter nuclide) in the dissolved phase and consequent relative enrichment in particulate phase. These results have been used for the determination of particle residence times longer than $^{234}\text{Th}/^{238}\text{U}$ ratios allow. and removal mechanism analysis.

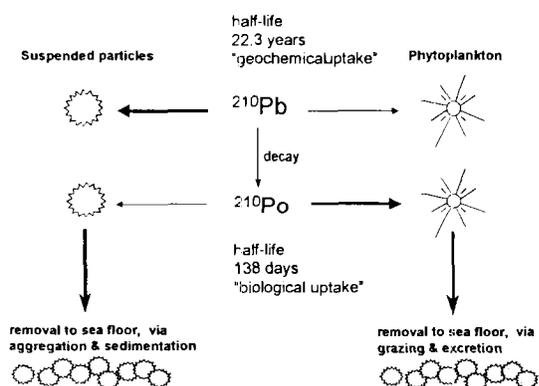


Figure 2. Physicochemical behaviour of ^{210}Pb and ^{210}Po radionuclides in seawater.

Conclusions

Scavenging and release of trace metals and other pollutants by particles plays an important role in determining their flux and incident concentrations in marine systems. Riverine and resuspended seafloor

particles are very important in these processes inshore, whereas further offshore biological processes become increasingly significant, dominating oligotrophic regions.

In the past many chemical oceanographic studies have characterised marine systems based on information gained on single observations. However, transitional kinetics between dissolved and particulate phases of heavy metals in the marine environment span several orders of magnitude and several mechanisms are involved depending on the chemical species (Honeyman and Santschi, 1988). Studies utilising a kinetic, or mechanistic approach to environmental analysis will greatly assist in the interpretation of several key issues associated with the biogeochemical fate and behaviour, hence environmental consequence, of pollutants in coastal and oceanic systems and allow sustainable management strategies to be developed.

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