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ScienceDirect
Marine Chemistry 104 (2007) 27–42
MARINE CHEMISTRY
www.elsevier.com/locate/marchem

Seasonal variations on the residence times and partitioning of short-lived radionuclides (^{234}Th, ^{7}Be and ^{210}Pb) and depositional fluxes of ^{7}Be and ^{210}Pb in Tampa Bay, Florida

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Received 18 January 2006; received in revised form 23 June 2006; accepted 23 June 2006
Available online 9 August 2006

Abstract

Historically, Tampa Bay has been impacted heavily by a wide range of anthropogenic perturbations that may include, agricultural-, shipping-, phosphate mining/distribution-related activities, as well as a burgeoning coastal population. Due to the presence of U-rich underlying sediments, elevated activities of U- and Th-series daughter products may be naturally released into this system. This region is also known for summer thunderstorms and corresponding increases in precipitation and surface water runoff. Only limited work has been conducted on the partitioning of particle-reactive radionuclides (such as ^{7}Be, ^{210}Pb, and ^{234}Th) in such a dynamic coastal system. We investigated both the removal residence time and partitioning of these radionuclides between filter-retained particulate matter ($\geq$0.5 μm) and the filtrate (<0.5 μm) phase during late spring (June 2003) and mid summer (August 2003) in the water column of Tampa Bay.

Our results indicate that the partitioning of ^{7}Be, ^{210}Pb, and ^{234}Th between filtrate and filter-retained phase is controlled foremost by enhanced bottom resuspension events during summer thunderstorms. As a consequence, no significant relationship exists between the distribution coefficients (K_d values) of these isotopes and the concentration of suspended particulate matter (SPM). Relatively faster recycling rates of atmospheric water vapor derived from the ocean results in lower atmospheric depositional fluxes of ^{210}Pb to the study site than predicted. The relationship between ^{7}Be and ^{210}Pb in bulk (wet+dry) deposition is compared to their respective water column activities. The residence times of particulate and dissolved ^{234}Th, ^{7}Be and ^{210}Pb, as well the distribution coefficients of these radionuclides, are then compared to values reported in other coastal systems.

Keywords: Tampa Bay; Scavenging of particle-reactive radionuclides; Be-7; Pd-210; Th-234; Distribution coefficients; Residence times of nuclides

1. Introduction

Particle-reactive radionuclides in the U- and Th-series and ^{7}Be, have been used extensively as tracers in estuarine and coastal environments to evaluate the biogeochemical reactivity of chemical constituents, as well as the provenance of suspended particulate matter, SPM (Broecker et al., 1973; Santschi et al., 1979, 1980; Aller et al., 1980; Olsen et al., 1982, 1986; Baskaran and Santschi, 1993; Baskaran et al., 1997; Feng et al., 1999; Swarzenski et al., 1999; Turner and Millward, 2002; Radakovitch et al.,

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doi:10.1016/j.marchem.2006.06.012

2003). The particulate matter composition within an estuary may consist of lithogenic (weathered crustal material, mainly quartz and silicate minerals), hydrogenic (coatings of oxyhydroxides of Fe and Mn, carbonates, sulfides and humic aggregates), biogenic (produced by biological processes from organisms such as bacteria, fungi, protozoans, plankton, fecal matter and other remains of decaying organisms, lipids, proteins, etc.), and anthropogenic (particulate pollutants such as sewage solids, surfactants, trace elements, coal dust, fly ash, etc.) components (Turner and Millward, 2002). The partitioning of reactive chemical species depends directly on the concentration, composition, and fate of SPM (Turekian, 1977). SPM concentrations are typically 2–4 orders of magnitude higher in coastal waters (1–100 mg L^{-1}) than in the open ocean (10–100 μg L^{-1}), and as SPM has a much larger surface area per unit mass compared to larger particles that may undergo faster settling, SPM tends to stay in the water column long enough to effectively scavenge particle-reactive chemical species. Thus, most of the particle-reactive radionuclides in estuarine and coastal waters are associated with SPM, and their dynamic interaction can be investigated using a suite of particle-reactive radionuclides.

One of the more useful techniques for quantifying the fate of particle-reactive radionuclides is to apply information from natural U- and Th-series and cosmogenic radionuclides, either by making use of parent–daughter nuclide disequilibria, or by measuring precise fluxes and standing crops (inventories) of atmospherically-delivered radionuclides (i.e., ^{210}Pb, ^{7}Be). For example, the disequilibrium between ^{234}Th ($t_{1/2}$=24.1 days) and ^{238}U (or ^{228}Th/^{228}Ra), and the determination of fluxes and standing crops of atmospherically-delivered ^{210}Pb ($t_{1/2}$=22.1 years) and cosmogenic ^{7}Be ($t_{1/2}$=53 days) are powerful tools for estimating the rates of processes that affect the estuarine distribution of pollutants (Broecker et al., 1973; Aller and Cochran, 1976; Bacon et al., 1976; Simpson et al., 1976; Turekian, 1977; Olsen et al., 1980, 1982; Li et al., 1979; Santschi et al., 1980; Minagawa and Tsunogai, 1980; Aller et al., 1980; Benninger and Krishnaswami, 1981; Kaufman et al., 1981; Baskaran and Santschi, 1993; Baskaran et al., 1997; Feng et al., 1999).

In estuarine environments, the primary sources of these radionuclides include, *i)* radiogenic production from the parent (i.e., ^{234}Th from ^{238}U, and ^{210}Pb from ^{222}Rn after several daughter products), *ii)* direct atmospheric deposition (fall out of ^{210}Pb and ^{7}Be), *iii)* erosional input from the watershed through the discharge of streams and rivers, and *iv)* radionuclides present in off-shore waters that may enter the estuary or bay through physical exchange processes. Seasonal events, such as major summer thunderstorms, appear to dominate the distribution of these radionuclides both in the water column and bottom sediments. Their distribution between the particulate and dissolved phases in the water column provides information on the role of seasonal events on the distribution of other particle-reactive species. In addition, the present study site, Tampa Bay, is among the largest estuaries in Florida (drainage basin = ~7500 km^2), and may be utilized as a model for the investigation of the fate of U- and Th-series radionuclides in an estuary where U activities may be both naturally and anthropogenically enriched. This bay receives very low sediment input (Schoellhamer, 1995) with a very low drainage basin area/sediment input ratio. The presence of two phosphate distribution facilities directly adjacent to Tampa Bay may provide an additional source of U- and Th-series radionuclides above levels present in ubiquitous U-rich sediments (Fanning et al., 1982; Swarzenski and Baskaran, 2007). We have analyzed water samples from 17 stations throughout Tampa Bay to test the hypothesis that in such a particle-depleted estuarine system, the particle–concentration effect (Honeyman and Santschi, 1989) will not be as pronounced as compared to other coastal systems where SPM concentrations are relatively high. Specific objectives were to investigate the range in calculated particle residence times using ^{7}Be, ^{210}Pb, and ^{234}Th, and to evaluate the partitioning of ^{7}Be, ^{210}Pb and ^{234}Th between the dissolved water and SPM during two seasons, late spring and summer of 2003. We also report variations in the atmospheric depositional input of ^{210}Pb and ^{7}Be to the water column and assess the relation between ^{7}Be and ^{210}Pb in the bulk atmospheric depositional flux as well as in the water column.

2. Materials and methods

2.1. Measurements of ^{7}Be and ^{210}Pb from Tampa Bay

Tampa Bay is a shallow (less than 3 m) estuarine system and the water column is generally well mixed vertically. Water samples from 17 stations within the bay were collected and analyzed for U- and Th-series radionuclides and ^{7}Be (Fig. 1) during June and August 2003. For ^{7}Be and ^{210}Pb analyses, ~150 L of water samples from Tampa Bay were filtered through a precleaned 0.5 μm (median pore size) prefilter and collected in precleaned 20-L containers and transferred into a 200-L drum for $Fe(OH)_3$ precipitation at the shore-based laboratory. The cubitainers were rinsed with 6 M HCl to remove any adsorbed ^{7}Be or ^{210}Pb. To the solution, 1 mL of stable Pb (≡1 mg, Aldrich 05618DR) and Be (≡1 mg,