Journal of Environmental Radioactivity xxx (2014) 1–9

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Investigations of the partitioning and residence times of Po-210 and Pb-210 in a riverine system in Southeast Michigan, USA

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ARTICLE INFO

Article history:
Received 26 May 2013
Received in revised form
12 January 2014
Accepted 15 January 2014
Available online xxx

Keywords:
^{210}Po
^{210}Pb
Partitioning coefficient K_d
Fractionation factor
Residence time

ABSTRACT

Some of the daughter products in the ^{222}Rn-decay series, such as ^{210}Po and ^{210}Pb, have been widely used as tracers and chronometers in aqueous systems. We measured the concentrations of ^{210}Pb and ^{210}Po in the dissolved (≤0.5 μm), bulk (unfiltered) and particulate phases (≥1 μm) collected in the Clinton River in the Lake St. Clair watershed in Southeast Michigan in order to investigate their partitioning between particulate and dissolved phases. Activity measurements of the dissolved and particulate phases revealed that an average of 38% (range: 12–59%) and 33% (range: 12–66%) of the total ^{210}Pb and ^{210}Po, respectively, in the water column was found in the particulate phase. The activity of dissolved and total ^{210}Pb was higher than that of ^{210}Po because of the higher atmospheric depositional fluxes of ^{210}Pb compared to ^{210}Po. Although the calculated K_d values of ^{210}Pb and ^{210}Po were similar, there was an inverse relationship between the K_d and suspended particulate matter concentration, indicating the presence of a particle concentration effect and we attribute this observation to the presence of significant amounts of colloidal ^{210}Po and ^{210}Pb in the dissolved phase. The fractionation factors for Po and Pb were found to be less than 1 in most cases. The first-order box model calculation-based residence times with respect to scavenging varied from 2 to 25 days for ^{210}Pb and 19–78 days for ^{210}Po, indicating higher particle-reactivity of ^{210}Pb compared to ^{210}Po.

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1. Introduction

Polonium-210 and ^{210}Pb have been extensively used as tracers and chronometers in marine system. Indeed, ^{210}Pb is the most widely used chronometer for dating of sediments in aqueous system that are <150 yrs old. Polonium, a sulfur group element which is fundamental to organic matter, is also a particle-reactive element with a wide range of K_d values, 103–105 cm^3 g^{-1} (Carvahlo, 1997; Nozaki et al., 1997; Hong et al., 1999; Kim et al., 2005; Yang et al., 2013). There is relatively a large body of data on the distribution of ^{210}Po and ^{210}Pb between particulate and dissolved phases in marine system, but their distribution in freshwater system is relatively sparse.

The major source of ^{210}Pb to the surface waters is atmospheric fallout; however, most of the dissolved ^{210}Po in the upper waters is from the decay of ^{210}Pb which is ultimately derived from atmospheric fallout with ^{210}Po/^{210}Pb activity ratio in precipitation of ~0.1 (e.g., except in areas that are highly influenced by volcanic eruption; Hussain et al., 1998; Baskaran, 2011). Thus, the concentration of ^{210}Pb in the surface waters primarily depends temporally and geographically on varying inputs of atmospheric deposition. Subsequent to the deposition, most of the ^{210}Pb is removed by sorption on to particulate matter. In a dynamic riverine system where terrigenous particulate fraction dominates (>90%; Jweda et al., 2008), most of the ^{210}Pb and ^{210}Po are expected to be controlled by lithogenic particulate matter. The ^{210}Po-^{210}Pb disequilibria in open ocean marine system have been utilized to determine the export fluxes of particulate organic carbon (POC) (Shimmield et al., 1995; Friedrich and Rutgers van der Loeff, 2002; Murray et al., 2005; Verdeny et al., 2009). This study aims to provide significant insight on their partitioning in a dynamic freshwater riverine system and intends to answer the following questions:

1) What is the range of distribution coefficients of ^{210}Po and ^{210}Pb in the riverine system and how do they vary with the changes in the concentrations of suspended particulate matter (SPM)?

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0265-931X/$ – see front matter © 2014 Published by Elsevier Ltd.
http://dx.doi.org/10.1016/j.jenvrad.2014.01.007

Please cite this article in press as: Mudbidre, R., et al., Investigations of the partitioning and residence times of Po-210 and Pb-210 in a riverine system in Southeast Michigan, USA, Journal of Environmental Radioactivity (2014), http://dx.doi.org/10.1016/j.jenvrad.2014.01.007

2) How do the removal rate constants of ^{210}Po and ^{210}Pb vary in a riverine system compared to other freshwater systems? and
3) How do the residence times of dissolved, particulate and total residence time of ^{210}Po and ^{210}Pb vary in a riverine water column?

The answers to these questions should help in understanding the fate and transport of other biogenic and abiogenic elements (such as Se, S group elements for ^{210}Po and lithogenic elements for ^{210}Pb) and other contaminants that have similar geochemical properties as those of ^{210}Pb and ^{210}Po.

2. Materials and methods

2.1. Study area

The Clinton River, located in Southeastern Michigan, drains ~1980 km^2 of the northern suburbs of Detroit including portions of 4 different counties (Macomb Co, Oakland Co, St. Clair Co, and Lapeer Co) and flows 127 km before it drains into Lake St. Clair, which is a very important water body of the Great Lakes System. Lake St. Clair is a relatively shallow (3.6 m mean depth) body of water in the Great Lakes system which is part of the connecting channels between Lake Huron and Lake Erie and serves as an important economic water resource, from which about 4.5 million people receive their drinking water.

2.2. Water sampling and preparation

Water samples for the determination of the concentration of the suspended particulate matter (SPM) and radionuclides were collected at four sampling stations monthly (Fig. 1) over a period of 4 months (around 15th of the month). The SPM concentration was determined by filtering 1 L of the river water collected at ~15 cm below the air–water interface through a pre-weighed Nuclepore filter paper (0.4 μm pore size and 47 mm diameter). Filtration was carried out using a covered Nalgene filtration set-up connected to a vacuum pump to ensure that there was no accumulation of dust on the filter paper. The filter paper was carefully removed and stored between two Petri dishes and allowed to air-dry for ~2–3 days at 80 °C. Once the filter paper was completely dry it was weighed (precision of 0.1 mg and the mass of the blank filter 15–20 mg) and the difference in weight in the concentration of SPM was determined. The error associated with the SPM determination is <1%.

Filtered water samples were collected by pumping 20 L of water using a peristaltic pump through pre-cleaned polypropylene filter cartridge (0.5 μm median nominal pore size). For particulate ^{210}Pb, large volumes of water samples (200–300 L) were filtered through the filter cartridge and the fiber was separated from the core and ashed in a furnace at 550 °C for approximately 6 h. The residual ash was weighed and packed into a vial for gamma counting. Unfiltered water was collected by pumping into cubitainers without the filter cartridge. Filtered and unfiltered water samples were brought back to the laboratory and were transferred into a 25 L low density polyethylene bucket. The cubitainers were rinsed with 50 mL of conc. HCl twice to remove any adsorbed radionuclides of interest on the walls of the cubitainers and added to the 25 L bucket. To the above solution, 250 μL of ^{209}Po (134.8 mBq/mL), 1.5 mL of $FeCl_3$ (50 mg Fe/mL) and 1000 μg of $Pb(NO_3)_2$ AAS Standard (Baker) were added. The ^{210}Pb level (with ^{210}Po in equilibrium) was found to be 10.3 mBq/g Pb and appropriate blank corrections for ^{210}Po and ^{210}Pb were applied (Baskaran et al., 2013). The solution was stirred continuously for 30 min using Easy-load masterflex circulator and then allowed to equilibrate for 3 h. After the solution had equilibrated, concentrated NH_4OH solution was added to achieve a pH of ~4 after which 1 mL of 10% Na_2CrO_4 was added. Addition of Na_2CrO_4 enhanced the recovery of ^{210}Po and ^{210}Pb, similar to the method adopted in Church et al. (2012). Eventually the pH was brought to ~7 and $Fe(OH)_3$ was allowed to precipitate for ~8 h. We have shown from laboratory experiments with separate aliquots of particulate and dissolved phases, that there is mass balance and removal of particulate matter is quantitative. The precipitate and solution were separated by decanting followed by filtration. The precipitate along with the filter paper was transferred to a pre-cleaned 100 mL Teflon beaker, and then 6 M HCl was added to dissolve the precipitate (heated if required). The filter paper was removed from the beaker and was rinsed with deionized water. About 0.5 g of ascorbic acid ($C_6H_8O_6$) was added and the pH was brought to ~1.5 by adding NH_4OH. The solution was heated for 15 min and the polished silver planchet was carefully placed at the bottom of the Teflon beakers and plated for 3 h at about 90 °C while the solution was constantly stirred using a bar magnet (Flynn, 1968). After 3 h of plating is complete, the silver planchet was removed, rinsed with distilled water, air dried and counted in the alpha detector for ~2–10 days (Jweda and Baskaran, 2011).

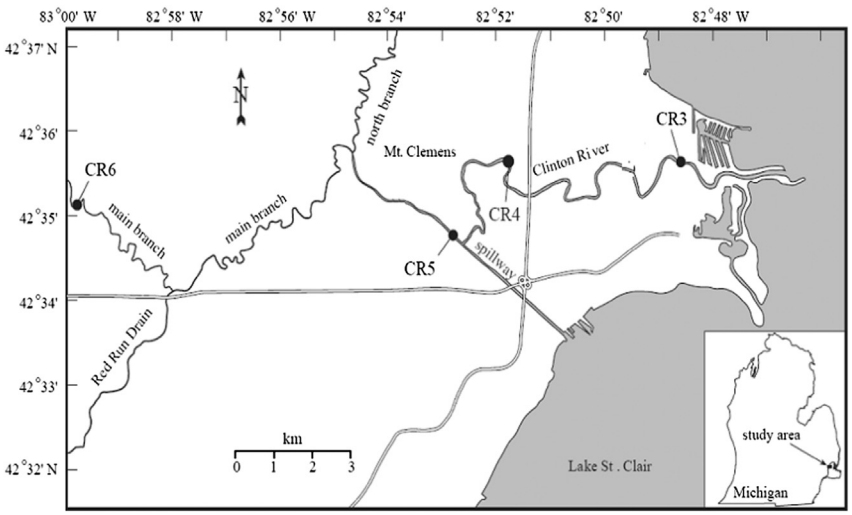

Fig. 1. Sample locations in Clinton river.

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