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Short communication

Lead-210 and Beryllium-7 fallout rates on the southeastern coast of Brazil

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ABSTRACT

Total ^{210}Pb and ^{7}Be fallout rates were measured on the coastal region of Niteroi, Brazil. The monthly depositional flux of ^{210}Pb and ^{7}Be varied by a factor of 26, from 1.7 to 43.3 mBq cm^{-2} year^{-1} and ~27, from 7.5 to 203.5 mBq cm^{-2} year^{-1}, respectively. The relatively large oscillations in the depositional flux of ^{210}Pb at this study site were likely due to variations in air mass sources, while the ^{7}Be fluctuations may be driven by a combination of weather conditions. Local geology could support the periodic high fluxes of ^{210}Pb from continental air masses, as shifting oceanic wind sources were affirmed by the uncorrelated ^{210}Pb and ^{7}Be fallout activities and ^{7}Be/^{210}Pb ratios. The ^{210}Pb atmospheric deposition was found to be in agreement with local sediment inventories, an important consideration in geochemical studies that estimate sedimentation processes.

1. Introduction

Radium-226 within rocks and sediments produces ^{222}Rn as part of the ^{238}U decay series. Radon-222, an inert gas, diffuses into the atmosphere and decays through several short lived daughter nuclides to ^{210}Pb ($t_{1/2}$22.3y). Beryllium-7 ($t_{1/2}$53d) is a relatively short lived naturally occurring radionuclide of cosmogenic origin. It is formed in the upper atmosphere by spallation of light atmospheric nuclei, nitrogen (Z ¼ 7), oxygen (Z ¼ 8) and carbon (Z ¼ 6) when they interact with cosmic rays.

Since ^{7}Be is of atmospheric origin, it is an excellent tracer of atmospheric sources and transport processes. Both ^{210}Pb and ^{7}Be may be removed from the atmosphere via wet or dry deposition (Papastefanou and Ioannidou, 2004; Kulan et al., 2006; Caillet et al., 2001). Information on ^{210}Pb and ^{7}Be-derived fallout rates of aerosols is useful to determine the transport processes of other atmospheric contaminants that behave in a similar manner to these radionuclides, such as trace metals (Kim et al 2000; McNeary and Baskaran, 2003). Seasonal events, such as rain, temperature variability and winds influence the distribution of ^{210}Pb and ^{7}Be fallout in depositional areas (Al-Azmi et al., 2001; Leppanen et al., 2010).

Recent ^{7}Be atmospheric studies in Rio de Janeiro show that regional precipitation and irregular air-masses patterns influence the ^{7}Be variability in the near surface air concentrations (Pacini et al., 2011; Leppanen et al., 2010). In the present study, we report on variations in the total atmospheric fluxes of ^{210}Pb and ^{7}Be to Niteroi – Rio de Janeiro, Brazil. This study also compares ^{210}Pb annual flux rates in local sediment columns to the ^{210}Pb fallout rates determined in this work. Knowledge of regional ^{210}Pb and ^{7}Be fallout rates are particularly important in geochemical studies that determine depositional processes such as sediment mixing and accumulation (e.g., Smoak and Patchineelam, 1999; Sanders et al., 2010a; Sabaris and Bonotto, 2011).

The maximum rainfall in the state of Rio de Janeiro occurs between December and March and minimum between June and August. On a monthly basis, rough estimates from local meteorological station indicate prevailing south-southeastern winds between April and September and north-northeastern winds from October to May. The coastal regional geology in southeastern Brazil is lined by the Serra do Mar mountain range and dominated by igneous rocks such as granite and granodiorite.

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2. Methods

Using a modified approach of Schuler et al. (1991),a total deposition collector consisting of a 20-L polyethylene drum with surface area of 2500 cm^2 was setup in Nitreoi, RJ, Brazil (22°53.55′S, 43°08.95′W) on a stand, near mean sea level, to avoid contamination by resuspended dust (Fig. 1). The collector was continuously exposed to the atmosphere from June 2006 through May 2007 and sampled monthly (Table 1).

To prevent any potential adsorption of ^{7}Be and ^{210}Pb on to the walls of the collector, the container was acidified prior to deployment with concentrated HCl. Total deposition samples were collected roughly every month or when drum was nearly full. Immediately after collection, yield spikes [1 mL ICP- Standard solution (10 mg L^{-1} Be, 10 mg L^{-1} Pb)] were added to the sample. The drum was cleaned repeatedly with 6 M HCl to remove any adsorbed Be and Pb from the surface of the drum. Nine hundred mg of iron, in the form of $Fe(NH_4)_2(SO_4)_2$, was added to samples and allowed to equilibrate for 3 h. Lead and beryllium were coprecipitated with iron by adding NH_3OH to attain a pH of ~7. After a minimum of 24 h, the $Fe(OH)_3$ coprecipitate was collected by decanting the supernatant and centrifuging (modification of Schuler et al., 1991). The solution in the centrifuge flask was then dried and quantitatively transferred to a gamma counting vial.

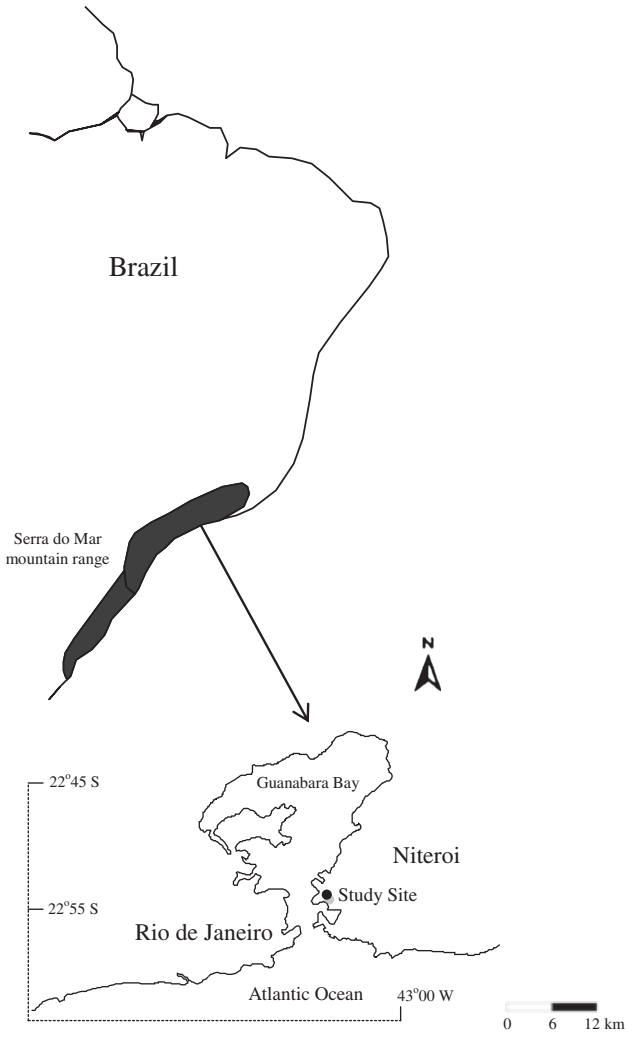

Fig. 1. Study site (Niteroi, Brazil).

Table 1
Sampling period (12 months) from June 1st 2006 to May 31st 2007, average sea level monthly temperatures (C°), rainfall (mm), ^{210}Pb, ^{7}Be and $^{7}Be/^{210}Pb$ (mBq cm^{-2} $year^{-1}$).

Sampling	Temp.	Rainfall	^{210}Pb	^{7}Be	$^{7}Be/^{210}Pb$
Jun	22	35	43.7	51.7	1.2
Jul	22	60	11.0	32.0	2.9
Aug	23	34	4.6	37.9	8.3
Sep	22	90	17.2	63.5	3.7
Oct	23	97	1.9	7.5	4.0
Nov	23	116	29.5	203.5	6.9
Dec	26	95	39.1	61.9	1.6
Jan	26	140	4.7	40.6	8.7
Feb	27	76	9.4	9.1	1.0
Mar	27	11	12.8	28.8	2.3
Apr	26	47	6.5	33.9	5.2
May	22	64	2.9	28.8	10.1
Yearly mean	24	72	15.3	49.8	4.6

Gamma counting was conducted in a high purity germanium coaxial high purity well coaxial detector, housed in a lead shield and coupled to a multichannel analyzer. Lead-210 and ^{7}Be were quantified by integrating gamma peaks at 46.5 and 477.6 keV respectively. Activities were calculated by multiplying the counts per minute by a factor that includes the gamma-ray intensity and detector efficiency. The ^{210}Pb counting efficiency was determined directly by the counting of a standard material. Due to its short half a ^{7}Be standard was not available. The ^{7}Be counting efficiency was interpolated from an exponential curve fit ($r^2 = 0.98$) of several detector efficiencies over an energy range from 46.5 to 911 keV. Counting errors were 2–3% for ^{7}Be and less than 5% for ^{210}Pb. Self absorption was evaluated by reducing the mass of a sample and repeating analysis. The sample mass was reduced seven times over a mass range from 1.7 to 0.08 g. The activity (2016.6 ± 90 $mBqg^{-1}$) remained the same with each reduction in mass. The net disintegrations per minute (dpm) vs. mass showed a linear relationship indicating no self adsorption occurred within this mass range.

The overall chemical yield of this procedure was determined after gamma analyses were completed. The sample was dissolved in 100 mL of 3 M HCl and 5 mL of this solution was analyzed for stable Be and Pb using an atomic absorption spectrometer (AAS). The chemical yield was used in the final calculation of ^{210}Pb and ^{7}Be.

3. Results and discussion

Collection intervals with specific fallout rates of ^{210}Pb and ^{7}Be for the 12 month period studied are shown in Table 1. The total ^{7}Be and ^{210}Pb depositional fluxes in the study site fluctuated over an order of magnitude during the sampling period. The ^{210}Pb fallout rate varied by a factor of 26, from 1.7 to 43.3 mBq cm^{-2} $year^{-1}$. The corresponding fallout rate of ^{7}Be varied by a factor of ~27, from 7.5 to 203.5 mBq cm^{-2} $year^{-1}$. The oscillations in the depositional fluxes do not appear to be influenced by rainwater flow or temperature fluctuations (Fig. 2). Indeed, no significant correlations were found between specific activities and the climatic variables shown in Fig. 2 or general wind direction (continental or oceanic).

Fallout rates depend on air mass source and a wide variation in air mass source can produce a relatively large variation in fallout rates. Due to low exhalation rates of ^{222}Rn over the ocean as compared to continental areas, oceanic air masses are typically depleted in ^{222}Rn and its daughter products, including ^{210}Pb (Baskaran and Swarzenski, 2007). As a consequence, atmospheric ^{210}Pb has a strong longitudinal dependency attributed to air mass origins (Turekian et al., 1977). However, ^{7}Be is of cosmogenic origin, and its flux to the Earth's surface has a latitudinal dependence,