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Recent $^{210}$Pb, $^{137}$Cs and $^{241}$Am accumulation in an ombrotrophic peatland from Amsterdam Island (Southern Indian Ocean)

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Abstract

Over the past 50 years, $^{210}$Pb, $^{137}$Cs and $^{241}$Am have been abundantly used in reconstructing recent sediment and peat chronologies. The study of global aerosol climate interaction is also partially depending on our understanding of $^{222}$Rn $^{210}$Pb cycling, as radionuclides are useful aerosol tracers. However, in comparison with the Northern Hemisphere, few data are available for these radionuclides in the Southern Hemisphere, especially in the South Indian Ocean. A peat core was collected in an ombrotrophic peatland from the remote Amsterdam Island (AMS) and was analyzed for $^{210}$Pb, $^{137}$Cs and $^{241}$Am radionuclides using an underground ultra low background gamma spectrometer. The $^{210}$Pb Constant Rate of Supply (CRS) model of peat accumulations is validated by peaks of artificial radionuclides ($^{137}$Cs and $^{241}$Am) that are related to nuclear weapon tests. We compared the AMS $^{210}$Pb data with an updated $^{210}$Pb deposition database. The $^{210}$Pb flux of $98 \pm 6$ Bq m$^{-2}$ y$^{-1}$ derived from the AMS core agrees with data from Madagascar and South Africa. The elevated flux observed at such a remote location may result from the enhanced $^{222}$Rn activity and frequent rainfall in AMS. This enhanced $^{222}$Rn activity itself may be explained by continental air masses passing over southern Africa and/or Madagascar. The $^{210}$Pb flux at AMS is higher than those derived from cores collected in coastal areas in Argentina and Chile, which are areas dominated by marine westerly winds with low $^{222}$Rn activities. We report a $^{137}$Cs inventory at AMS of $144 \pm 13$ Bq m$^{-2}$ (corrected to 1969). Our data thus contribute to the under represented data coverage in the mid latitudes of the Southern Hemisphere.

1. Introduction

Lead $^{210}$ (T$_{1/2}$ 22.3 years) dating is the most common method employed to estimate short term (from years to decades) chronologies in peat, estuarine, fluvial, and lacustrine environments (Le Roux and Marshall, 2011; Robbins and Edgington, 1975; Benoit and Rozan, 2001; Humphries et al., 2010). $^{210}$Pb originates from the decay of gaseous $^{222}$Rn, which escapes from the Earth’s continental crust to the atmosphere (Graustein and Turekian, 1990). $^{210}$Pb adsorbs strongly to the surface of aerosols in the 0.1–0.5 μm diameter size range as soon as it is produced in the air (Knuth et al., 1983). $^{210}$Pb bearing aerosols are distributed globally by general atmospheric circulation and can be deposited on the Earth’s surface mainly by precipitation, but also by dry fallout as well as convective updrafts (Knuth et al., 1983; Baskaran, 2011). The $^{210}$Pb deposited from the atmosphere is called “unsupported $^{210}$Pb” or “excess $^{210}$Pb” (denoted $^{210}$Pb$_{ex}$), which should be distinguished from the $^{210}$Pb produced inside the matrix (e.g. lake sediment) and which is named “supported $^{210}$Pb” (Guevara et al., 2003).

The Constant Rate of Supply (CRS) model based on $^{210}$Pb$_{ex}$ flux, which could be validated by nuclear fallout studies (e.g. $^{137}$Cs, $^{241}$Am), is by now widely used (e.g. Appleby et al., 2001, 2008). Another radionuclide that is widely used to derive ages is $^{137}$Cs. With a half life of 30.2 years, $^{137}$Cs is considered as one of the important radionuclides among those from nuclear emissions (e.g. atmospheric nuclear weapon tests in the 1950s 1970s with the peak in 1963 in the Northern Hemisphere and the fallout from the...
Chernobyl accident in 1986), with respect to being a persistent tracer and an indicator of single event chronology (Aoyama et al., 2006; Rodway Dyer and Walling, 2010). In contrast, $^{241}$Am another artificial radionuclide is strictly related to nuclear bomb testing in remote areas.

Up to now, most studies about the inventory of sediment radionuclides and radiochronology have been conducted in the Northern Hemisphere. Limited work has been carried out in the Southern Hemisphere, especially in the Indian Ocean. The scarcity of studies conducted in the Southern Hemisphere is partly due to the lower fallout of $^{210}$Pb and $^{137}$Cs, fewer continental surfaces and fewer man made radionuclide emissions and fallout, which generally result in lower activities bordering on analytical detection limits (Owens and Walling, 1996; Bonotto and De Lima, 2006). In the Southern Hemisphere nuclear weapons fallout is about three times lower than that in the Northern Hemisphere. Consequently, the $^{137}$Cs fallout peak is usually more difficult to identify in cores due to the relatively high measurement uncertainties associated with the low $^{137}$Cs concentrations (Hancock et al., 2011). Southern Hemisphere investigations on $^{210}$Pb and $^{137}$Cs have been confined primarily to large land masses, such as South America (Sanders et al., 2006; Guevara et al., 2003; Cisternas et al., 2001), Australia and New Zealand (Pfitzer et al., 2004; Hancock et al., 2011) and South Africa and Madagascar (Humphries et al., 2010; Kading et al., 2009; Iva novitch and Harmon, 1992; Rabesiranana et al., 2016).

No studies on terrestrial sediment radionuclides and radiochronology exist for the Southern Indian Ocean, although the island is an important part of the global atmospheric and oceanic circulation patterns. Amsterdam Island (AMS, 37° S) is located just north of the Subtropical Front (at approximately 40° S, Orsi et al., 1995), where cool, low salinity subpolar water submerges beneath warm, saline subtropical water (Prell et al., 1979). The island is located at the northern edge of both the Southern Westerly wind belt and the Antarctic Circumpolar Current.

The main objectives of this study were to investigate (1) to what extent observations in the Southern Indian Ocean could define Southern Hemisphere mid latitude $^{210}$Pb, $^{137}$Cs and $^{241}$Am back ground conditions for the last 100 years and (2) how $^{210}$Pb levels are comparable between different matrices (e.g. wetland, sediment, glacier and atmospheric deposition) at different latitudes of the world, which could allow us to draw a new global sketch of $^{210}$Pb flux.

In this paper, for the very first time the inventories and fluxes of $^{210}$Pb and $^{137}$Cs together with a $^{210}$Pb based peat accumulation rate for AMS are reported. This island is located in Southern Indian Ocean at 37° S and 3400 km from the nearest land mass. The atmospheric conditions at this location offer the possibility to potentially define $^{210}$Pb background concentrations, in a place with minimal perturbation from anthropogenic influences (Gaudichet et al., 1989; Angot et al., 2014). AMS is therefore an ideal site to investigate the background levels of $^{210}$Pb, $^{137}$Cs and $^{241}$Am in the Southern Hemisphere, as well as to detect long range transport of anthropogenic radionuclides.

2. Materials and methods

2.1. Study area

AMS (37°50’S, 77°32’E) is a small volcanic island with a surface of 55 km² and a maximum elevation of 881 m above sea level (a.s.l.). The center of the island is formed by a volcanic caldera in which an ombrotrophic peatland develops. The island is located at the northern edge of the westerly wind belt in the South Indian Ocean at a minimum distance of 3400 km and 5000 km upwind from the nearest land masses, respectively, Madagascar and South Africa. The climate in AMS is mild oceanic, with frequent clouds (Angot et al., 2014). The annual precipitation is 1124 mm y⁻¹ based on 40 year annual average data from 1951 to 1990 (Meteo France data reported in Miller et al., 1993). The weather station is located at 29 m above sea level. However, although no record exists, the cloud accumulation at the top of the island makes the precipitation level much higher in the caldera than that recorded at the weather station. The orographic effect (Roe, 2005; Le Roux et al., 2008) would enhance precipitation and therefore, $^{210}$Pb bearing aerosols scavenging. Relatively higher volume of rainfall above 600 m a.s.l. also favors moss growth and peat accumulation in the caldera of AMS. The annual temperature is 13.8 °C at the weather station, while the average annual humidity is 80% with little seasonal var iations. Located in the middle of the Indian Ocean, AMS is also at the crossroads of African, Australian and Southern American dust tra jectories (Li et al., 2008; Lamy et al., 2014).

2.2. Core sampling and sub sampling

One 5 m long peat sequence (AMS14 PB01A, 37°50.742’S, 77°32.858’E) was collected from the center of a raised bog at 738 m a.s.l. in December 2014 using a stainless steel Russian D corer of 10 cm internal diameter and 50 cm length (Belokopytov and Beresnevich, 1955; De Vleeschouwer et al., 2014; Vanneste et al., 2016) (Fig. 1). A second (AMS14 PB01B, 4.15 m length) and a third core (AMS14 PB01C, top 1 m length) at the same site were collected and stored as archives. All cores were photographed, described and wrapped in plastic film and PVC tubes before being shipped by boat to France in -4 °C fridges. Cores were frozen and subsequently sliced at roughly 1 cm resolution using a clean sub sampling procedure described in De Vleeschouwer et al. (2010). After being cleaned with MilliQ water and the edges removed (Givelet et al., 2004), the subsamples were dried using an ALPHA 1–4 LD plus freeze dryer. Prior to freeze drying, the dimensions of each sub sample were measured using a vernier caliper in order to i) obtain the volume for calculating the dry bulk density and ii) to estimate the cut loss between each slice by comparing the cumulative slices thickness and the total length of the core. In this paper, we focus on last 100–150 years of peat accumulation representing the top 12 cm of the master core.

2.3. Radiometric measurements

Fifteen freeze dried sample aliquots (approximately 0.3 g) were analyzed at the LAFARA underground laboratory located in Ferrières in the French Pyrénées (Van Beek et al., 2013). Prior to analysis, samples were sealed to prevent any $^{222}$Rn loss and stored for a period of 3 weeks to ensure radioactive equilibrium between $^{226}$Ra, $^{214}$Pb and $^{214}$Bi. The $^{210}$Pb, $^{226}$Ra, $^{137}$Cs and $^{241}$Am activities were determined using a well type germanium detector that was protected from cosmic rays by 85 m of rock, thus yielding a very low background (Van Beek et al., 2013). The $^{210}$Pb, $^{137}$Cs and $^{241}$Am were measured using the gamma lines at 46.5 keV, 661.7 keV and 59.5 keV, respectively. The $^{226}$Ra was determined using the 295 keV, 351.9 keV and 609.3 keV gamma emissions of its decay chain descendants ($^{224}$Pb and $^{214}$Bi). Because of the very low ac tivities present in the peat samples and because the volume of material to be analyzed was small, each sample was analyzed for at least four days. We used RGU1, RGTH1 and IAEA 375 standards provided by IAEA to calibrate the detector.

Excess $^{210}$Pb activities are calculated by correcting the total $^{210}$Pb for the $^{226}$Ra supported by $^{226}$Ra. Since the $^{226}$Ra activities were below the detection limit in the core, the $^{210}$Pbex activities are equivalent to the total $^{210}$Pb activities (Supplementary Table S1). In the following, we report $^{210}$Pbex activities. The detection limits
achieved in this study (considering the low amount of material that was analyzed) were 20 Bq kg\(^{-1}\) for \(^{210}\)Pb, 0.4 Bq kg\(^{-1}\) for \(^{137}\)Cs and 0.6 Bq kg\(^{-1}\) for \(^{241}\)Am.

2.4. Calculating decays, fluxes and inventories

Atmospheric \(^{210}\)Pb fluxes \(\Phi\) (Bq m\(^{-2}\) yr\(^{-1}\)) were calculated using:

\[
\Phi = \lambda I_{210\text{Pb}}
\]

where \(\lambda\) is the \(^{210}\)Pb decay constant (0.03114 yr\(^{-1}\)), \(\ln 2/T_{1/2}\) and \(I\) is the \(^{210}\)Pbex inventory (Bq m\(^{-2}\)) in peat calculated from (Appleby, 1997):

\[
I = \sum_{i=0}^{\infty} \rho(x) \cdot C(x) \cdot dx
\]

where \(\rho(x)\) (g cm\(^{-3}\)) is the dry bulk density, \(dx\) is the soil thickness (cm) and \(C(x)\) is the excess \(^{210}\)Pb activity (Bq kg\(^{-1}\)). (Sanchez Cabeza et al., 2007) The \(^{137}\)Cs inventory was also calculated using equation (2).

The formula for calculating \(^{137}\)Cs cumulative decay corrected fallout at year 1969 (maximum fallout, according to Aoyama et al., 2006) is shown below,

\[
CD_{1969} = \sum_{\tau=1969}^{1969} D_{\tau} e^{-\lambda(1969-\tau)}
\]

\(CD_{1969}\): cumulative decay corrected fallout at year 1969 (Bq m\(^{-2}\)); \(\lambda\): radioactive decay constant of \(^{137}\)Cs (0.023 yr\(^{-1}\)), \(\ln 2/T_{1/2}\); \(D_{\tau}\): annual deposition of \(^{137}\)Cs at year \(\tau\) (Bq m\(^{-2}\)).

3. Results and discussion

3.1. Downcore distribution of \(^{210}\)Pb, \(^{137}\)Cs and \(^{241}\)Am activities

Given that ombrotrophic peatlands are only depending on precipitation for their water balance, \(^{210}\)Pb flux is assumed to be exclusively of atmospheric origin at AMS. This is confirmed by the absence of supported \(^{210}\)Pb because no \(^{226}\)Ra was detected. Except for the first sample that contained living Sphagnum moss, the \(^{210}\)Pbex activities decrease with increasing depth down to approximately 12 cm. (Fig. 2 and Supplementary Table S1). As shown in Fig. 2, the activities of \(^{137}\)Cs and \(^{241}\)Am displayed peaks at the same depth (4.4 \text{e} 5.8 cm), with values of 22 \pm 2 Bq kg\(^{-1}\) and 7 \pm 1 Bq kg\(^{-1}\), respectively, indicating that these peaks can be related to nuclear bomb testing (Spalding et al., 2005).

The total \(^{210}\)Pbex inventory was 3160 \pm 200 Bq m\(^{-2}\) with a corresponding \(^{210}\)Pbex flux of 98 \pm 6 Bq m\(^{-2}\) yr\(^{-1}\). According to equation (3), the corrected value for \(^{137}\)Cs inventory on AMS in 1969 was 144 \pm 13 Bq m\(^{-2}\), which is two times lower than the modeled average inventory of 580 Bq m\(^{-2}\) (range from 150 to 1430 Bq m\(^{-2}\)) at 35°S (Aoyama et al., 2006), but higher than the activity reported at 45°S in the same paper. Our observed \(^{137}\)Cs inventory for AMS is thus close to the lower limit of these estimates at 35°S. \(^{241}\)Am activity is relatively high showing potentially the immobility of this radionuclide in the peat column compared to cesium. Despite this, the activity inventory ratio \(^{137}\)Cs/\(^{241}\)Pu\(1969\) = 1.2, derived from \(^{137}\)Cs and \(^{241}\)Am activities respectively, is on the same order of magnitude than what can be found in Antarctica and sub Antarctica islands (Pournch et al., 2003; Roos et al., 1994).

3.2. \(^{210}\)Pb chronology

Well dated peat profiles are valuable archives of past environmental changes. The Constant Rate of Supply (CRS) model (Appleby,
was applied to $^{210}$Pb inventories calculated from the $^{210}$Pb$_{ex}$ data to generate ages. The peat core presented here spans a period of about 157 years, indicating a peat accumulation rate of about 0.75 mm yr$^{-1}$.

Oldfield et al. (1995) suggested that $^{210}$Pb measurements alone cannot result in an accurate chronology of peat accumulation. However, when constrained by $^{241}$Am and $^{137}$Cs activity profiles in the upper part of a sequence, $^{210}$Pb can provide good chronologies of peat accumulation. The $^{210}$Pb$_{ex}$ dates from the peat profile in AMS calculated using the CRS model were independently validated by $^{137}$Cs and $^{241}$Am that displayed highest activities between 1960 and 1981 (Fig. 3), corresponding to the period of nuclear weapon tests (i.e. in the 1960s). Moreover the detection of $^{241}$Am excludes the possibility that $^{137}$Cs post depositional mobility processes would have shifted the $^{137}$Cs activity maximum (Schettler et al., 2006 Part B).

### 3.3. $^{210}$Pb flux in AMS compared with global $^{210}$Pb depositional flux

We updated the database from Turekian et al. (1977) and Preiss (1997) by compiling 47 new entries from the last 20 years. Global estimates of $^{210}$Pb flux from the literature, integrated over 30° latitudinal belts within different matrices, are shown in Fig. 4 and Supplementary Table S2. Only terrestrial data, i.e. from wetlands (peatlands, salt marshes and swamps), sediments (lakes, estuarine and soil profiles), glaciers (ice core and firn) as well as atmospheric deposition (snow, precipitation plus dry fallout) are included.

Globally, the $^{210}$Pb flux measured in wetland sequences show smaller uncertainties compared to fluxes measured from atmospheric deposition and sediment sequences (Fig. 4). The dominant $^{210}$Pb input to wetlands is through the atmosphere in contrast to lake basins and the data are integrated over several years (1 cm layer represents a period of more than 10 years). The relatively larger uncertainty observed in atmospheric deposition could be explained by short term measurements (i.e. annual) that can enhance different tropospheric contributions. Many factors (e.g., adjoining drainage areas) could alter the $^{210}$Pb flux in sediment sequences.

In general, the $^{210}$Pb flux is lower at high latitudes (>60° latitude) (Fig. 4), especially when measured in glaciers and atmospheric deposition, where the low density of land masses results in lower $^{222}$Rn emissions. According to the existing data, the $^{210}$Pb fluxes measured in sediments are higher than those measured in the other three categories in the Northern Hemisphere, while in the Southern Hemisphere the values detected in wetlands are the highest (except between 60 and 90°S where no measurement exists). The $^{210}$Pb fluxes from atmospheric deposition are similar to those from wetlands in the Northern Hemisphere (Fig. 4). Wetlands are found to be the appropriate archives for a good estimation of atmospheric flux, due to the absence of in wash of sediments, and the low energy budget (absence of streams). However, in the Southern Hemisphere, $^{210}$Pb flux from wetlands and atmospheric deposition are not similar, which might be partially due to the scarcity of data (only 7 for wetlands) resulting in a non representative dataset. Compared to the Northern Hemisphere, fewer data are available for the Southern Hemisphere, and thus more effort should be made in the future to fill this gap.

In the peat core from AMS, the $^{210}$Pb flux was 98 ± 6 Bq m$^{-2}$ y$^{-1}$, which is higher than most values reported at around 40°S. For example, Baskaran (2011) found an average $^{210}$Pb flux of 61 ± 2 Bq m$^{-2}$ y$^{-1}$ between 30 and 40°S and 42 Bq m$^{-2}$ y$^{-1}$ between 40 and 50°S based on terrestrial or marine settings. Guelle et al. (1998) used three kinds of wet scavenging schemes to simulate the $^{210}$Pb distribution and found a flux of 28–34 Bq m$^{-2}$ y$^{-1}$ between 30 and 60°S. The $^{210}$Pb atmospheric deposition in AMS collected in Amsterdam Island.

![Fig. 2. Vertical profiles of $^{210}$Pb$_{ex}$, $^{137}$Cs and $^{241}$Am activities in the peat core collected in Amsterdam Island.](image)

![Fig. 3. $^{210}$Pb$_{ex}$ inferred chronology based on CRS model.](image)

![Fig. 4. Global atmospheric depositional fluxes of $^{210}$Pb among different matrices at different latitude. (Data assembled from Supplementary Table S2).](image)
deposition in Tasmania is 41.5 Bq m$^{-2}$ yr$^{-1}$ (42.5°S, 147.5°E) (Preiss, 1997). Rosen (1957) found a much lower value (1.24 Bq m$^{-2}$ yr$^{-1}$) in Wellington (41°17′S), New Zealand based on the calculation of the 222Rn flux. The estimated unsupported 210Pb fluxes values from the global model made by Turekian et al. (1977) and El Douasy (1988) were <74 (in terrestrial settings) and 58.4 Bq m$^{-2}$ yr$^{-1}$, respectively. Unsupported 210Pb flux from lake sediments in Northern Patagonia (from 40°30′S to 41°10′S) showed very low values, ranging between 4 and 48 Bq m$^{-2}$ yr$^{-1}$ (Guevara et al., 2003). The average 210Pb flux from lake sediment in central Chile (36°51′S, 73°05′W) was 23.6 Bq m$^{-2}$ yr$^{-1}$ (Cisternas et al., 2001). However, when compared with a soil profile from Madagascar, we find almost the same value (95.8 Bq m$^{-2}$ yr$^{-1}$, Rabesiranana et al., 2016) as in AMS. Our value is also quite similar to other values obtained from swamps or salt marshes found in South Africa (139 ± 37 Bq m$^{-2}$ yr$^{-1}$, n = 4, Humphries et al., 2010; Kading et al., 2009; Ivanovich and Harmon, 1992). The 210Pb flux in AMS is lower than some measurements conducted in wetlands between 30° and 60° N, e.g., Romania (between 133 and 277 Bq m$^{-2}$ yr$^{-1}$, Begy et al., 2016), China (254 and 421 Bq m$^{-2}$ yr$^{-1}$, Bao et al., 2010).

Depositional flux of 210Pb at any given site depends on the local 222Rn emanation rates and the relative proportion of maritime and continental air masses along with the differences in the amount and frequency of precipitation (Baskaran, 2011). The highly variable 210Pb depositional fluxes give insight into the sources and sinks of aerosols. The study sites in Argentina and Chile (Cisternas et al., 2001; Guevara et al., 2003), showing much lower 210Pb fluxes, are located at the coast near the Pacific Ocean, and are influenced by westerly winds bringing oceanic air masses. The contribution of sea salt for 210Pb from oceanic 222Rn is negligible. Global 222Rn flux from continents is estimated to be around 1300 m$^{-2}$ yr$^{-1}$, and Madagascar. This may be explained by the continental air mass enriched in 222Rn originating from South Africa and/or Madagascar influencing AMS together with the frequent and heavy rainfalls at the top of the island which would enhance deposition. The 137Cs inventory was 144 ± 13 Bq m$^{-2}$ (corrected to 1969). Since no terrestrial studies were conducted in the south Indian Ocean, the data reported here from AMS (210Pb, 137Cs and 241Am) are of value for the global database.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenrad.2017.05.004.

References
