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

Chuxian Li, Gaël Le Roux, Jeroen E. Sonke, Pieter van Beek, Marc Souhaut, Nathalie van Der Putten, François de Vleeschouwer

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

Chuxian Li [a, *], Gaël Le Roux [a], Jeroen Sonke [b], Pieter van Beek [c], Marc Souhaut [c], Nathalie Van der Putten [d], François De Vleeschouwer [a]

[a] EcoLab, Université de Toulouse, CNRS, INPT, UPS, Tolosan, France
[b] Geoscience, Environment, Toulouse, Midi- Pyrénées Observatory, Toulouse, France
[c] LEGOS (CNRS/CNES/IRD/UPS), Midi- Pyrénées Observatory, Toulouse, France
[d] Department of Geology, Quaternary Sciences, Lund University, Lund, Sweden

Keywords:
Radionuclides
^{210}Pb
^{137}Cs
^{241}Am
Peat
Southern indian ocean

A B S T R A C T

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 radionu clides (^{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 ± 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 ± 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 (^{210}Pb, $T_{½}$ 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 environ ments (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

* Corresponding author. EcoLab (Laboratoire Ecologie Fonctionnelle et Environnement), ENSAT, Avenue de l'Agrobiopole, 31326 Castanet Tolosan, France.
E-mail address: chuxian.li@ensat.fr (C. Li).

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 ra dionuclides 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 (Pfitzner 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 radio chronology exist for the Southern Indian Ocean, although the area is an important part of the global atmospheric and oceanic circu lation 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 at 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 km^2 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°32.898′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 pro cedure 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 (^{214}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 ^{210}Pb supported by ^{226}Ra. Since the ^{226}Ra activities were below the detection limit in the core, the $^{210}Pb_{ex}$ activities are equivalent to the total ^{210}Pb activities (Supplementary Table S1). In the following, we report $^{210}Pb_{ex}$ activities. The detection limits

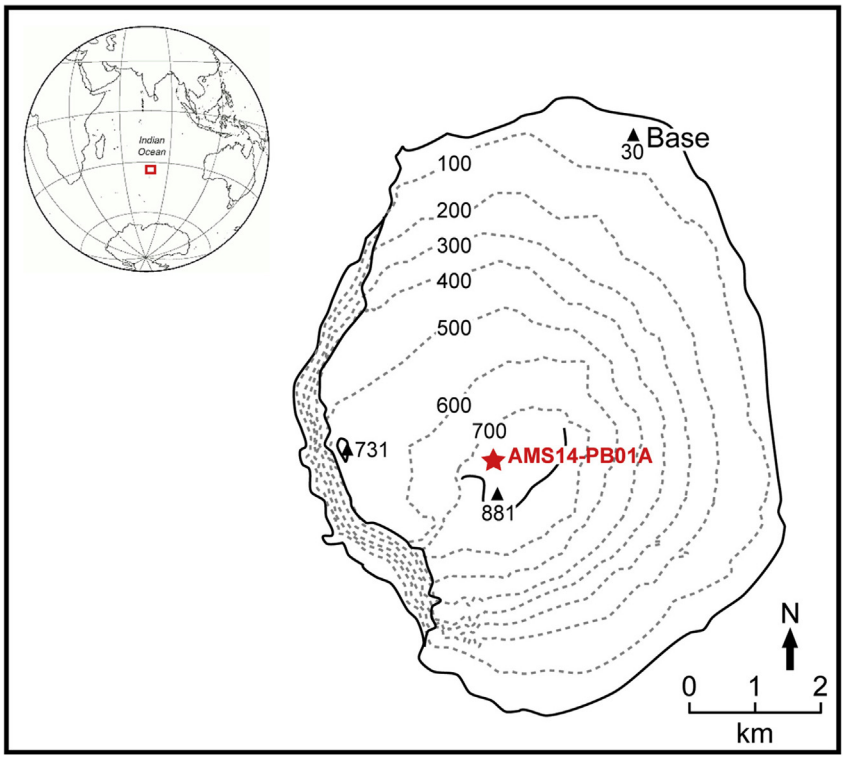

Fig. 1. Sampling site in Amsterdam Island.

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 Φ (Bq·m^{-2}·yr^{-1}) were calculated using:

$$\Phi \quad \lambda \; I_{210Pb} \tag{1}$$

where λ is the ^{210} Pb day constant (0.03114 yr^{-1}, λ ln2/T$_{1/2}$) and I is the ^{210}Pb$_{ex}$ inventory (Bq·m^{-2}) in peat calculated from (Appleby, 1997):

$$I \quad \sum_{i\ 0}^{i\ \infty} \rho(x) \cdot C(x) \cdot dx \tag{2}$$

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,

$$CD_{1969} \quad \sum_{\tau}^{1969} D_\tau e^{-\lambda(1969-\tau)} \tag{3}$$

CD_{1969}: cumulative decay corrected fallout at year 1969 (Bq·m^{-2}); λ: radioactive decay constant of ^{137}Cs (0.023 yr^{-1}, λ ln2/T$_{1/2}$); D_τ: annual deposition of ^{137}Cs at year τ (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}Pb$_{ex}$ 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 (4.4–5.8 cm), with values of 22 ± 2 Bq·kg^{-1} and 7 ± 1 Bq·kg^{-1}, respectively, indicating that these peaks can be related to nuclear bomb testing (Spalding et al., 2005).

The total ^{210}Pb$_{ex}$ inventory was 3160 ± 200 Bq·m^{-2} with a corresponding ^{210}Pb$_{ex}$ flux of 98 ± 6 Bq·m^{-2}·y^{-1}. According to equation (3), the corrected value for ^{137}Cs inventory on AMS in 1969 was 144 ± 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 radioelement 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 (Pourchet 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,

1997; Binford, 1990) 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)(Preiss et al., 1996). 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

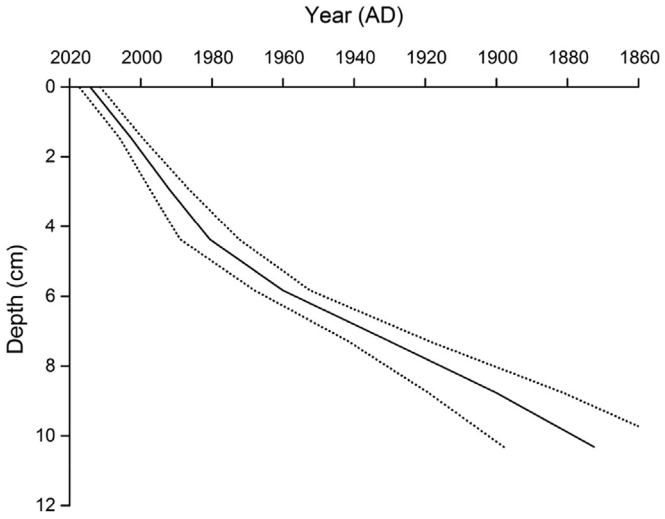

Fig. 3. ^{210}Pb$_{ex}$ inferred chronology based on CRS model.

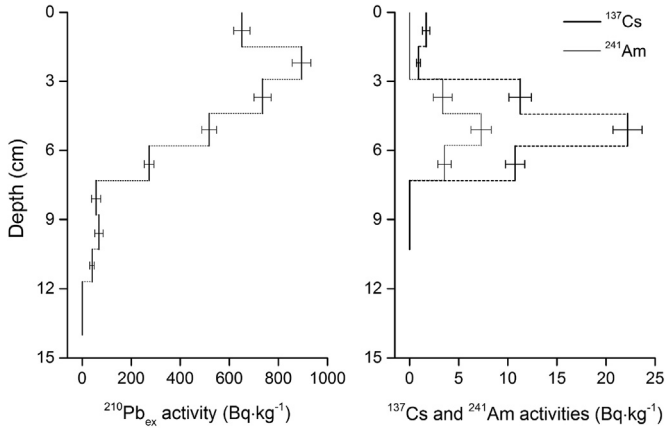

Fig. 2. Vertical profiles of ^{210}Pb$_{ex}$, ^{137}Cs and ^{241}Am activities in the peat core collected in Amsterdam Island.

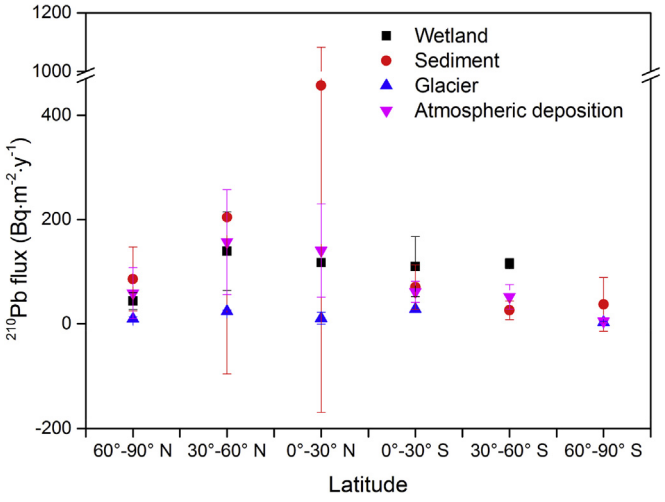

Fig. 4. Global atmospheric depositional fluxes of ^{210}Pb among different matrices at different latitude. (Data assembled from Supplementary Table S2).

deposition in Tasmania is 41.5 $Bq \cdot m^{-2} \cdot y^{-1}$ (42.5°S, 147.5°E) (Preiss, 1997). Rosen (1957) found a much lower value (1.24 $Bq \cdot m^{-2} \cdot y^{-1}$) in Wellington (41°17′S), New Zealand based on the calculation of the ^{222}Rn flux. The estimated unsupported ^{210}Pb fluxes values from the global model made by Turekian et al. (1977) and El Daoushy (1988) were ≤74 (in terrestrial settings) and 58.4 $Bq \cdot m^{-2} \cdot y^{-1}$, respec tively. Unsupported ^{210}Pb flux from lake sediments in Northern Patagonia (from 40°30′S to 41°10′S) showed very low values, ranging between 4 and 48 $Bq \cdot m^{-2} \cdot y^{-1}$ (Guevara et al., 2003). The average ^{210}Pb flux from lake sediment in central Chile (36°51′ S, 73°05′ W) was 23.6 $Bq \cdot m^{-2} \cdot y^{-1}$ (Cisternas et al., 2001). However, when compared with a soil profile from Madagascar, we find almost the same value (95.8 $Bq \cdot m^{-2} \cdot y^{-1}$, 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 $Bq \cdot m^{-2} \cdot y^{-1}$, n 4, Humphries et al., 2010; Kading et al., 2009; Ivanovich and Harmon, 1992). The ^{210}Pb flux in AMS is lower than some measurements conducted in wetlands between 30 and 60°N, e.g., Romania (between 133 and 277 $Bq \cdot m^{-2} \cdot y^{-1}$, Begy et al., 2016), China (254 and 421 $Bq \cdot m^{-2} \cdot y^{-1}$, Bao et al., 2010).

Depositional flux of ^{210} Pb at any given site depends on the local ^{222}Rn 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 ^{210}Pb 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 ^{210}Pb 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 ^{210}Pb from oceanic ^{222}Rn is negligible. Global ^{222}Rn flux from continents is estimated to be around 1300–1800 $Bq \cdot m^{-2} \cdot d^{-1}$, which is around 2 orders of magnitude higher than that from the oceans (Samuelsson et al., 1986; Nazaroff, 1992). The ^{210}Pb flux at AMS is relatively high (discussed above). Despite the remoteness of the island, Angot et al. (2014) found that air masses passing over the southern African continent and/or Madagascar could enhance ^{222}Rn activity in AMS based on the study of back trajectory. Con tinental air masses originating from South Africa and/or Madagascar are enriched in ^{210}Pb that is then scavenged to clouds over the Indian Ocean. The regular orographic heavy rainfalls in the caldera at the top of AMS promote the deposition of ^{210}Pb from the South Africa and/or Madagascar air masses into the peatland, resulting in a ^{210}Pb flux on the island similar to those in South Africa and Madagascar.

4. Conclusion

For the very first time, radionuclide (^{210}Pb, ^{137}Cs, ^{241}Am) mea surements conducted in a peat core taken from an ombrotrophic peatland on AMS are presented. The chronology based on ^{210}Pb using a CRS model is consistent with other chronomarkers (^{137}Cs and ^{241}Am), which allows the reconstruction of a peat mass accu mulation rate of 0.75 mm yr^{-1} for a period of 157 years. The ^{210}Pb flux of 98 ± 6 $Bq \cdot m^{-2} \cdot y^{-1}$ measured in the peat core is relatively high compared to many observations at around 40°S. This pattern might be explained by the fact that, most studies conducted in the mid latitudes in Southern Hemisphere are located on the west side of the continents, close to the coast, and are highly influenced by westerly winds bringing air masses with low ^{222}Rn activities. However, the ^{210}Pb flux at AMS is comparable to the ones in South Africa and Madagascar. This may be explained by the continental air mass enriched in ^{222}Rn 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 ^{137}Cs inventory was 144 ± 13 $Bq \cdot m^{-2}$ (corrected to 1969). Since no terrestrial studies were conducted in the south Indian Ocean, the data reported here from AMS (^{210}Pb, ^{137}Cs and ^{241}Am) are of value for the global database.

Acknowledgements

We are grateful to Svante Bjorck, Bart Klink and Elisabeth Michel for their help during fieldwork. These results would never have been obtained if we had not had the incredible support of the Mission 66 of Amsterdam Island. Very special thanks to Alain Quivoron and Hubert Launay. The field expedition of this project was supported by IPEV project 1066 PARAD to FDV. We are very grateful to Nina Marchand (IPEV) for the incredible logistical sup port and Cédric Marteau for making the sampling possible in the protected areas of the TAAF Nature Reserve. Chuxian Li's PhD is supported by a scholarship from the Chinese Scholarship Council (No. 201506990002). We are grateful to EDF (Electricité De France) for allowing us to run our germanium detectors in the tunnel of Ferrières. We thank the European Union and Région Occitanie Pyrénées Méditerranée for supporting the LAFARA underground laboratory through a FEDER funding (SELECT project). We finally thank the 4 anonymous reviewers for their useful comments that greatly improved the quality of this manuscript.

Appendix A. Supplementary data

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

References

Angot, H., Barret, M., Magand, O., Ramonet, M., Dommergue, A., 2014. A 2-year record of atmospheric mercury species at a background Southern Hemisphere station on Amsterdam Island. Atmos. Chem. Phys. 14 (20), 11461 11473.

Aoyama, M., Hirose, K., Igarashi, Y., 2006. Re-construction and updating our understanding on the global weapons tests 137 Cs fallout. J. Environ. Monit. 8 (4), 431 438.

Appleby, P., 1997. In Dating recent sediments by 210Pb: problems and solutions. Helsinki. In: Proc. 2nd NKS/EKO-1 Seminar.

Appleby, P., Birks, H.H., Flower, R.J., Rose, N., Peglar, S.M., Ramdani, M., Kraïem, M.M., Fathi, A.A., 2001. Radiometrically determined dates and sedimentation rates for recent sediments in nine North African wetland lakes (the CASSARINA Project). Aquat. Ecol. 35 (3 4), 347 367 (a).

Appleby, P., 2008. Three decades of dating recent sediments by fallout radionuclides: a review. Holocene 18 (1), 83 93 (b).

Bao, K., Xia, W., Lu, X., Wang, G., 2010. Recent atmospheric lead deposition recorded in an ombrotrophic peat bog of Great Hinggan Mountains, Northeast China, from 210 Pb and 137 Cs dating. J. Environ. Radioact. 101 (9), 773 779.

Baskaran, M., 2011. Po-210 and Pb-210 as atmospheric tracers and global atmospheric Pb-210 fallout: a review. J. Environ. Radioact. 102 (5), 500 513.

Begy, R.C., Kovacs, T., Veres, D., Simon, H., 2016. Atmospheric flux, transport and mass balance of 210 Pb and 137 Cs radiotracers in different regions of Romania. Appl. Radiat. Isotopes 111, 31 39.

Belokopytov, I., Beresnevich, V., 1955. Giktorf's peat borers. Torfyanaya Promyshlennost 8, 9 10.

Benoit, G., Rozan, T.F., 2001. 210Pb and 137Cs dating methods in lakes: a retrospective study. J. Paleolimnol. 25 (4), 455 465.

Binford, M.W., 1990. Calculation and uncertainty analysis of 210Pb dates for PIRLA project lake sediment cores. J. Paleolimnol. 3 (3), 253 267.

Bonotto, D.M., De Lima, J., 2006. 210Pb-derived chronology in sediment cores evidencing the anthropogenic occupation history at Corumbataí River basin, Brazil. Environ. Geol. 50 (4), 595 611.

Cisternas, M., Araneda, A., Martinez, P., Perez, S., 2001. Effects of historical land use on sediment yield from a lacustrine watershed in central Chile. Earth Surf. Process. Landforms 26 (1), 63 76.

De Vleeschouwer, F., Chambers, F.M., Swindles, G.T., 2010. Coring and sub-sampling of peatlands for palaeoenvironmental research. Mires Peat 7.

De Vleeschouwer, F., Vanneste, H., Mauquoy, D., Piotrowska, N., Torrejón, F., Roland, T., Stein, A., Le Roux, G., 2014. Emissions from pre-Hispanic metallurgy in the South American atmosphere. PLoS One 9 (10), e111315.

El-Daoushy, F., 1988. A summary on the lead-210 cycle in nature and related applications in Scandinavia. Environ. Int. 14 (4), 305 319.

Gaudichet, A., Lefevre, R., Gaudry, A., Ardouin, B., Lambert, G., Miller, J., 1989. Mineralogical composition of aerosols at Amsterdam island. Tellus B 41 (3), 344 352.

Givelet, N., Le Roux, G., Cheburkin, A., Chen, B., Frank, J., Goodsite, M.E., Kempter, H., Krachler, M., Noernberg, T., Rausch, N., 2004. Suggested protocol for collecting, handling and preparing peat cores and peat samples for physical, chemical, mineralogical and isotopic analyses. J. Environ. Monit. 6 (5), 481 492.
Graustein, W.C., Turekian, K.K., 1990. Radon fluxes from soils to the atmosphere measured by 210Pb 226Ra disequilibrium in soils. Geophys. Res. Lett. 17 (6), 841 844.
Guelle, W., Balkanski, Y., Dibb, J.E., Schulz, M., Dulac, F., 1998. Wet deposition in a global size-dependent aerosol transport model: 2. Influence of the scavenging scheme on 210Pb vertical profiles, surface concentrations, and deposition. J. Geophys. Res. Atmos. 103 (D22), 28875 28891.
Guevara, S.R., Rizzo, A., Sánchez, R., Arribére, M., 2003. 210Pb fluxes in sediment layers sampled from Northern Patagonia lakes. J. Radioanalytical Nucl. Chem. 258 (3), 583 595.
Hancock, G., Leslie, C., Everett, S., Tims, S., Brunskill, G., Haese, R., 2011. Plutonium as a chronomarker in Australian and New Zealand sediments: a comparison with 137 Cs. J. Environ. Radioact. 102 (10), 919 929.
Humphries, M.S., Kindness, A., Ellery, W.N., Hughes, J.C., Benitez-Nelson, C.R., 2010. 137 Cs and 210 Pb derived sediment accumulation rates and their role in the long-term development of the Mkuze River floodplain, South Africa. Geomorphology 119 (1), 88 96.
Ivanovich, M., Harmon, R.S., 1992. Uranium-series Disequilibrium: Applications to Earth, Marine, and Environmental Sciences, vol. 2.
Kading, T., Mason, R., Leaner, J., 2009. Mercury contamination history of an estuarine floodplain reconstructed from a 210 Pb-dated sediment core (Berg River, South Africa). Mar. Pollut. Bull. 59 (4), 116 122.
Knuth, R.H., et al., 1983. "Size distribution of atmospheric Pb and 210 Pb in rural New Jersey: implications for wet and dry deposition." Precipitation scavenging, dry deposition, and resuspension. In: Proceedings, vol. 2.
Lamy, F., Gersonde, R., Winckler, G., Esper, O., Jaeschke, A., Kuhn, G., Ullermann, J., Martínez-Garcia, A., Lambert, F., Kilian, R., 2014. Increased dust deposition in the Pacific Southern Ocean during glacial periods. Science 343 (6169), 403 407.
Le Roux, Gael, et al., 2008. Aerosol deposition and origin in French mountains estimated with soil inventories of 210 Pb and artificial radionuclides. Atmos. Environ. 42.7, 1517 1524.
Le Roux, G., Marshall, W.A., 2011. Constructing recent peat accumulation chronologies using atmospheric fall-out radionuclides. Mires Peat 7.1, e14.
Li, F., Ginoux, P., Ramaswamy, V., 2008. Distribution, transport, and deposition of mineral dust in the Southern Ocean and Antarctica: contribution of major sources. J. Geophys. Res. Atmos. 113 (D10).
Miller, J.M., et al., 1993. A 10-year trajectory flow climatology for Amsterdam island, 1980 1989. Atmos. Environ. Part A. Gen. topics 27 (12), 1909 1916.
Nazaroff, W.W., 1992. Radon transport from soil to air. Rev. Geophys. 30 (2), 137 160.
Oldfield, F., Richardson, N., Appleby, P., 1995. Radiometric dating (210Pb, 137Cs, 241Am) of recent ombrotrophic peat accumulation and evidence for changes in mass balance. Holocene 5 (2), 141 148.
Orsi, Alejandro H., Whitworth, Thomas, Nowlin, Worth D., 1995. On the meridional extent and fronts of the Antarctic Circumpolar Current. Deep Sea Res. Part I Oceanogr. Res. Pap. 42.5, 641 673.
Owens, P.N., Walling, D.E., 1996. Spatial variability of caesium-137 inventories at reference sites: an example from two contrasting sites in England and Zimbabwe. Appl. Radiat. isotopes 47 (7), 699 707.
Pfitzner, J., Brunskill, G., Zagorskis, I., 2004. 137 Cs and excess 210 Pb deposition patterns in estuarine and marine sediment in the central region of the Great Barrier Reef Lagoon, north-eastern Australia. J. Environ. Radioact. 76 (1), 81 102.
Pourchet, M., et al., 2003. Radionuclides deposition over Antarctica. J. Environ. Radioact. 68.2, 137 158.
Preiss, N., 1997. Etude du 210 Pb d'origine atmosphérique dans l'air, la neige, les sols et les sédiments: Mesures, inventaires et interprétation a l'échelle globale.
Preiss, N., Mélieres, M.A., Pourchet, M., 1996. A compilation of data on lead 210 concentration in surface air and fluxes at the air-surface and water-sediment interfaces. J. Geophys. Res. Atmos. 101 (D22), 28847 28862.
Prell, Warren L., William, H. Hutson, Williams, Douglas F., 1979. The subtropical convergence and late Quaternary circulation in the southern Indian Ocean. Mar. Micropaleontol. 4, 225 234.
Rabesiranana, N., Rasolonirina, M., Solonjara, A., Ravoson, H., Andriambololona, R., Mabit, L., 2016. Assessment of soil redistribution rates by 137 Cs and 210 Pb ex in a typical Malagasy agricultural field. J. Environ. Radioact. 152, 112 118.
Robbins, J.A., Edgington, D., 1975. Determination of recent sedimentation rates in Lake Michigan using Pb-210 and Cs-137. Geochimica Cosmochimica Acta 39 (3), 285 304.
Rodway-Dyer, S., Walling, D., 2010. The use of 137 Cs to establish longer-term soil erosion rates on footpaths in the UK. J. Environ. Manag. 91 (10), 1952 1962.
Roe, Gerard H., 2005. Orographic precipitation. Annu. Rev. Earth Planet. Sci. 33, 645 671.
Roos, P., et al., 1994. Deposition of 210Pb, 137Cs, 239+ 240Pu, 238Pu, and 241Am in the antarctic peninsula area. J. Environ. Radioact. 24.3, 235 251.
Rosen, R., 1957. Note on some observations of radon and thoron exhalation from the ground. NZJ Sci. Technol. 38, 644 654.
Samuelsson, C., Hallstadius, L., Persson, B., Hedvall, R., Holm, E., Forkman, B., 1986. 222Rn and 210Pb in the Arctic summer air. J. Environ. Radioact. 3 (1), 35 54.
Sanchez-Cabeza, J.-A., Garcia-Talavera, M., Costa, E., Pena, V., Garcia-Orellana, J., Masqué, P., Nalda, C., 2007. Regional calibration of erosion radiotracers (210Pb and 137Cs): atmospheric fluxes to soils (northern Spain). Environ. Sci. Technol. 41 (4), 1324 1330.
Sanders, C.J., Santos, I.R., Silva-Filho, E.V., Patchineelam, S.R., 2006. Mercury flux to estuarine sediments, derived from Pb-210 and Cs-137 geochronologies (Guaratuba Bay, Brazil). Mar. Pollut. Bull. 52 (9), 1085 1089.
Schettler, G., Mingram, J., Negendank, J.F., Jiaqi, L., 2006. Palaeovariations in the East-Asian monsoon regime geochemically recorded in varved sediments of Lake Sihailongwan (Northeast China, Jilin Province). Part 2: a 200-year record of atmospheric lead-210 flux variations and its palaeoclimatic implications. J. Paleolimnol. 35 (2), 271 288.
Spalding, Kirsty L., et al., 2005. Forensics: age written in teeth by nuclear tests. Nature 437.7057, 333 334.
Turekian, K.Y., Nozaki, Y., Benninger, L.K., 1977. Geochemistry of atmospheric radon and radon products. Annu. Rev. Earth Planet. Sci. 5, 227.
Van Beek, P., Souhaut, M., Lansard, B., Bourquin, M., Reyss, J.-L., Von Ballmoos, P., Jean, P., 2013. LAFARA: a new underground laboratory in the French Pyrenees for ultra low-level gamma-ray spectrometry. J. Environ. Radioact. 116, 152 158.
Vanneste, H., De Vleeschouwer, F., Bertrand, S., Martínez-Cortizas, A., Vanderstraeten, A., Mattielli, N., Coronato, A., Piotrowska, N., Jeandel, C., Roux, G.L., 2016. Elevated dust deposition in Tierra del Fuego (Chile) resulting from Neoglacial Darwin Cordillera glacier fluctuations. J. Quat. Sci. 31 (7), 713 722.