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Atmospheric fluxes of ^{210}Pb to the western Mediterranean Sea and the Saharan dust influence

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[1] Pb 210 is a well known tracer of particle dynamics in the marine environment. Geochemical models partially rely on the knowledge of its atmospheric input. Unfortunately, this is poorly known in the western Mediterranean Sea, especially regarding long-term records. In this work we have evaluated the ^{210}Pb annual atmospheric flux to this region from the analysis of 12 soil cores collected from coastal and island sites and total atmospheric deposition collected in Corsica during 1 year. The ^{210}Pb fluxes ranged from 34 ± 3 to 121 ± 12 Bq m^{-2} yr^{-1}, with an average of 75 Bq m^{-2} yr^{-1}, and were strongly correlated (R^2 = 0.95) with mean annual rainfall. This provides the possibility to determine ^{210}Pb fluxes in a given location if the mean annual rainfall is well known, a useful outcome for both marine biogeochemical and soil erosion studies in this region. We have also estimated the mean annual atmospheric flux of ^{210}Pb due to Saharan dust events registered in total deposition north of Barcelona during the last 17 years, yielding a value of 20 Bq m^{-2} yr^{-1}. This flux represents a fraction of about 16% of the total atmospheric deposition of ^{210}Pb in this area, but it could be up to 50% in sites with lower rainfall.

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1. Introduction

[2] Pb 210 ($T_{1/2}$ = 22.3 years) is continuously produced in the atmosphere by decay of gaseous ^{222}Rn ($T_{1/2}$ = 3.8 d) exhaled from the continental crust after decay from ^{226}Ra. Pb 210, commonly named excess, atmospheric or unsupported ^{210}Pb, is scavenged to the Earth's surface by wet and dry deposition. Pb 210 is one of the most widely used radiotracers to study environmental processes. It has been commonly used to study biogeochemical processes in the oceans [*Nozaki et al.*, 1991; *Wei and Murray*, 1994], atmospheric deposition and anthropogenic contamination [*Shotyk et al.*, 2002; *Kaste et al.*, 2003], sedimentary processes [*Robbins and Edgington*, 1975; *DeMaster et al.*, 1991; *Roberts et al.*, 1997] and sediment geochronology [*Koide et al.*, 1972; *Benninger et al.*, 1976; *Sanchez-Cabeza et al.*, 1999a; *Benoit and Rozan*, 2001].

[3] Pb 210 has previously been applied as a tracer of various marine processes in the western Mediterranean basin [i.e., *Zuo et al.*, 1997; *Palanques et al.*, 1998; *Radakovitch et al.*, 1999; *Sanchez-Cabeza et al.*, 1999b; *Masqué et al.*, 2003], and other oceanic regions.

[4] One of the key parameters of the ^{210}Pb cycle in the environment is its atmospheric flux, knowledge of which is required for dating recent reservoirs, and assessment of soil erosion, sediment processes and biogeochemical processes in the oceanic water column. It is well known that the ^{210}Pb deposition to the Earth's surface is higher over continents than oceans and varies with season, longitude and local meteorological conditions [*Preiss et al.*, 1996]. The most common procedure to estimate the ^{210}Pb flux is by collecting wet and dry deposition during periods long enough to accommodate seasonal and episodic variations. The ^{210}Pb flux can also be determined in natural repositories such as ice cores, lake sediments and soils that integrate long-term deposition [*Turekian et al.*, 1977; *Preiss et al.*, 1996]. The difficulty of installing a network of collection stations over the sea during large periods of time has revealed the requirement to estimate the basin-wide deposition of ^{210}Pb by using data obtained from undisturbed soils collected in surrounding coastal areas [*Moore et al.*, 1973; *Nozaki et al.*, 1978; *Olsen et al.*, 1985; *Graustein and Turekian*, 1986; *Monaghan*, 1989].

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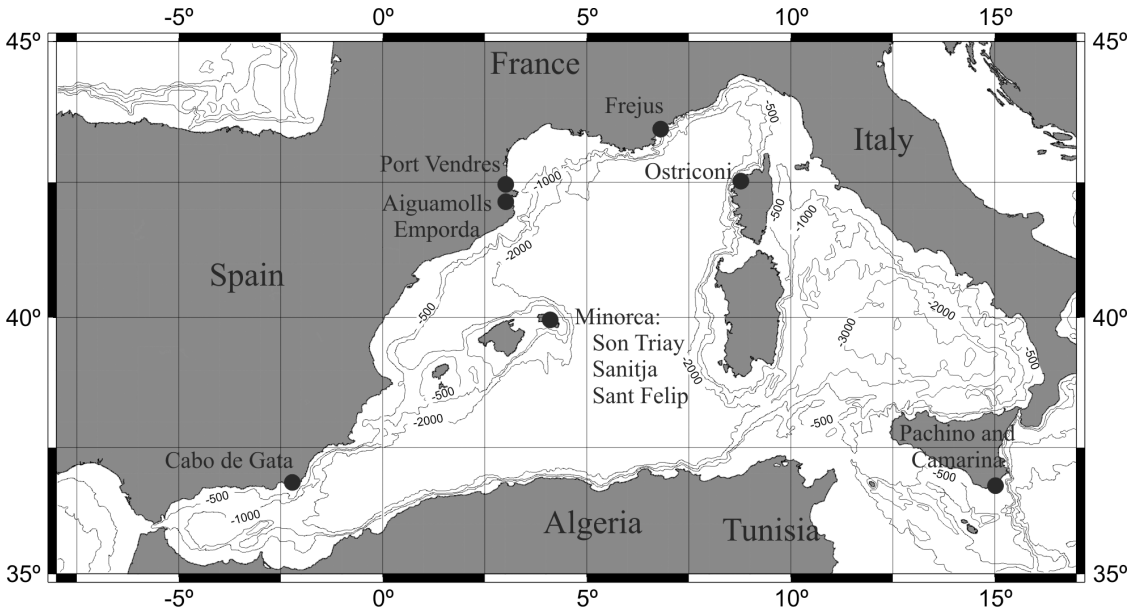

Figure 1. Location of soil samples collected along the western Mediterranean coasts.

[5] The western Mediterranean is a semiclosed basin which is affected by a variety of climates, demographic trends and corresponding environmental changes [*Martin and Milliman*, 1997]. North Africa (Morocco, Algeria and Tunisia) represents an area where rainfall is concentrated in Northern areas and mostly averages less than 500 mm yr^{-1}. On the other hand, rainfall is more variable in the north side of the basin (Spain, France and Italy), ranging from 200 mm yr^{-1} in the Almeria area (Spain) to more that 1000 mm yr^{-1} in some areas near the Rhône River (France) and north of Italy.

[6] In the Mediterranean Sea there is a scarcity of data on atmospheric ^{210}Pb fluxes, especially regarding long-term records, and those available correspond mainly to the northwestern Mediterranean. In the Ebro River Delta (558 mm yr^{-1} rainfall) the atmospheric ^{210}Pb flux determined from a microbial mat was 81.2 Bq m^{-2} yr^{-1} [*Sanchez-Cabeza et al.*, 1999b]. The annual ^{210}Pb fluxes measured from total atmospheric deposition were 110 Bq m^{-2} yr^{-1} [*Heyraud*, 1982] and 102 Bq m^{-2} yr^{-1} [*Tateda et al.*, 2003] in Monaco (883 mm^{-1}) and 82 Bq m^{-2} yr^{-1} [*Hussain et al.*, 1990] in the Camargue, where *Miralles et al.* [2004] estimated 132 Bq m^{-2} yr^{-1} in a wetland soil. Only one study has been reported in Northern Africa [*Appleby et al.*, 2001], where atmospheric ^{210}Pb fluxes were found to range from 116 to 134 Bq m^{-2} yr^{-1} in wetland lakes of Tunisia. However, these data are not sufficient to provide a realistic estimation of ^{210}Pb fluxes to the Mediterranean basin.

[7] On the other hand, it is well known that the transport and deposition of Saharan dust affects extensive areas of the world, such as the North Atlantic, America and Europe [*Prospero and Carlson*, 1972; *Loÿe-Pilot et al.*, 1986; *Chester et al.*, 1997; *Avila et al.*, 1997; *Guerzoni et al.*, 1999]. The Mediterranean area is also strongly affected by events that inject to the atmosphere large amounts of dust emitted from North Africa. The Saharan dust inputs may also significantly contribute to the atmospheric ^{210}Pb flux by scavenging ^{210}Pb from the atmosphere during its route from Africa to the Mediterranean Sea and also during its deposition [*Appleby et al.*, 2002]. These fallout events (''red rain'') are relatively frequent in the Mediterranean region. Saharan dust deposition has been estimated at 5 g m^{-2} yr^{-1} in northeastern Spain, 4 g m^{-2} yr^{-1} in southeastern France, 8 g m^{-2} yr^{-1} in Sardinia and 12 g m^{-2} yr^{-1} in Corsica, while in the eastern Mediterranean basin the deposition was larger, ranging from 20 to 50 g m^{-2} yr^{-1} (compiled by *Guerzoni et al.* [1999]).

[8] In this work we have evaluated the atmospheric flux of ^{210}Pb over the western Mediterranean Sea by determining its inventory in 12 undisturbed soils from coastal areas and islands: Cabo Gata and Aiguamolls Empordà (southern and northeastern Iberian Peninsula); Son Triay, Sant Felip and Sanitja in Minorca (Balearic Islands); Fréjus and Port Vendres (southern mainland France); three soils in Ostriconi (Island of Corsica); Pachino and Camarina (Island of Sicily) (Figure 1). Data obtained from soils in Ostriconi were compared with total atmospheric deposition collected at the same site during one year. We have also evaluated the fraction of ^{210}Pb flux due to Saharan dust events from the particulate fraction of total deposition sampled at La Castanya–Montseny biological station (41°46′N, 2°21′E), 40 km to the northeast of Barcelona, and 25 km west of the Mediterranean Sea, during the period 1983–2000, where an average of 7 Saharan wet dust events per year has been reported [*Escudero et al.*, 2005].

2. Materials and Methods

[9] Soil samples were collected from sites bearing low vegetation and without clear signals of anthropogenic impact (i.e., agriculture, civil engineering, etc.). Soil cores were collected with a stainless steel cylindrical sampler (4.5 cm diameter and 50 cm length); after description samples were cut into 5-cm slices and stored in sealed plastic bags at 4°C until analysis. Once in the laboratory, samples were dried to constant weight at 50°C and were homogenized to calculate the dry bulk density. A fraction

Table 1. The ^{210}Pb Surface Activities, Inventories and Atmospheric Fluxes From Soil Cores Collected in Coastal Areas of the Western Mediterranean Sea

	Coordinates			^{210}Pb		
Sampling Site	Latitude, N	Longitude, E	Rainfall, mm yr^{-1}	Surface Activity, Bq kg^{-1}	Inventory, Bq m^{-2}	Flux, Bq m^{-2} yr^{-1}
Pachino	36°41′	15°06′	417[a]	80 ± 5	1849 ± 422	56 ± 13
Camarina	36°52′	14°26′	435[b]	51 ± 3	1888 ± 160	58 ± 5
Cabo Gata	36°44′	02°11′	196[c]	82 ± 3	1129 ± 112	34 ± 3
Sant Felip (Minorca)	39°52′	04°18′	561[d]	57 ± 3	2770 ± 284	85 ± 9
Sanitja (Minorca)	40°04′	04°05′	556[d]	47 ± 3	2764 ± 376	84 ± 11
Son Triay (Minorca)	40°02′	03°51′	512[d]	85 ± 6	2685 ± 446	82 ± 14
Ostriconi 1 (Corsica)	42°40′	09°04′	464[e]	73 ± 3	2008 ± 233	61 ± 7
Ostriconi 2 (Corsica)	42°39′	09°05′	464[e]	82 ± 3	1917 ± 307	59 ± 9
Ostriconi 3 (Corsica)	42°39′	09°05′	464[e]	68 ± 3	2037 ± 260	62 ± 8
Port Vendres	42°31′	03°07′	382[f]	102 ± 5	1999 ± 182	61 ± 6
Fréjus	43°24′	06°44′	833[g]	100 ± 7	4317 ± 357	121 ± 12
Aiguamolls Empordà	43°14′	03°06′	628[h]	91 ± 4	2703 ± 209	83 ± 6
Mean			493 ± 46	77 ± 17	2286 ± 769	71 ± 27
Range			196–833	47–102	1129–4317	34–121

[a]Cozzo Spadaro meteorological station (http://www.meteosicilia.it) (1951–2002).
[b]Camarina meteorological station (http://www.meteosicilia.it) (2001–2002).
[c]Almeria – airport meteorological station, Instituto Nacional de Meteorologia) (1971–2000).
[d]Seguiment dels recursos hídrics a Menorca (1984–2002) (http://www.obsam.org).
[e]Ostriconi meteorological station (1999–2003).
[f]Port Bou (Catalunya) meteorological station (1997–2002).
[g]http://meteodumuy.free.fr/.
[h]Parc Natural dels Aiguamolls de l'Empordà meteorological station (1984–2002).

of each sample was ground for radiometric analysis. Aliquots of 100–200 mg were totally digested by using an analytical microwave, after addition of ^{209}Po as internal tracer.

[10] Total atmospheric deposition was collected at Ostriconi using a Plexiglas collector composed of a bottom hemisphere with a cylindrical component above [*Lambert*, 1963]. The cylindrical part, 10 cm high, defines a collecting surface of 0.1 m^2 and the collecting aperture was 1.50 m above the ground. A nylon mesh (80 μm mesh size) was placed between the cylinder and the hemisphere to prevent coarse local debris from falling into the collector. Deposition was collected in a polyethylene tank containing about 50 mL of HCl 1M, placed on the ground below the hemisphere. At the end of the collection period the volume of the sample was reduced by subboiling evaporation and the collector tank was rinsed with diluted HCl to desorb radionuclides from the walls. When the volume was lower than 1 L, a known amount of ^{209}Po was added as internal tracer and the samples were digested in the presence of HCl (32%) and HNO_3 (35%).

[11] Total atmospheric deposition at Montseny was sampled with 4 open bulk deposition collectors placed at 1.5 m above the ground. The collectors, designed like those of *Likens et al.* [1977], consisted of a polyethylene funnel of 19 cm diameter connected by tygon tubing to a 10 L polyethylene bottle. The site was visited weekly and the collected rainwater during the week was taken to the laboratory for analysis. Also with the same periodicity, collectors were replaced by clean funnels and bottles. The dust fraction had sedimented gravitationally during the period of exposure, and both the wet and dust fractions were always sampled because the collectors were continuously open to the atmosphere. In a previous work it was showed that bulk precipitation collected at the Montseny site retained the principal features of wet precipitation [*Avila and Alarcón*, 1999]. Sample treatment is detailed by *Avila and Rodà* [2002]. Samples were filtered through 0.45 μm pore size filters (Millipore). In order to digest the insoluble fraction of the atmospheric deposition, a procedure developed at the Jaume Almera–CSIC Institute was used [*Querol et al.*, 2001]. A half of each filter was digested in closed PFA reactors with HNO_3:$HClO_4$:HF at 90°C. Subsequently, the acidic solution was cooled and dried on a hot plate at 200°C. The dry residue was redissolved in 2 mL of NHO_3 and subsequently made to volume to 25 mL; 12.5 mL of this solution was used for determination of ^{210}Pb activity. In order to determine the organic and other nondust components in the dust samples, a measure was made in May 1999 to differentiate the weight contribution of organic particulate material with respect to the mineral dust in the filtrate: the organic material was not detectable in terms of mass. Therefore, although there might be some contribution of organic material to the filtered weight, owing to the low density of the pollen compared to mineral dust, it is not likely that it produces an important bias.

[12] The ^{210}Pb activities were determined by measuring its daughter nuclide ^{210}Po in radioactive equilibrium following the methodology described by *Sanchez-Cabeza et al.* [1998]. Polonium isotopes from each sampling process were plated onto high-purity silver discs and counted with α spectrometers equipped with low background silicon surface barrier (SSB) detectors (EG&G Ortec). Excess ^{210}Pb was calculated as the difference between total (measured) ^{210}Pb and ^{210}Pb supported by ^{226}Ra in the soils; by this, excess ^{210}Pb equals the atmospheric supply of ^{210}Pb. The ^{226}Ra was determined by γ spectrometry using calibrated geometries on a coaxial HPGe detector (Canberra) at the Physics Department of the Universitat Autònoma de Barcelona. The ^{226}Ra was quantified from ^{214}Pb (351 keV line), after secular equilibrium of the radioactive chain. Uncertainties (1 sigma) were calculated by quadratic propagation of all uncertainties, notably counting errors of samples and blanks.

[13] Atmospheric ^{210}Pb fluxes (F) were calculated using $F = I \cdot \lambda$, where λ is the ^{210}Pb decay constant (0.0311 yr^{-1})

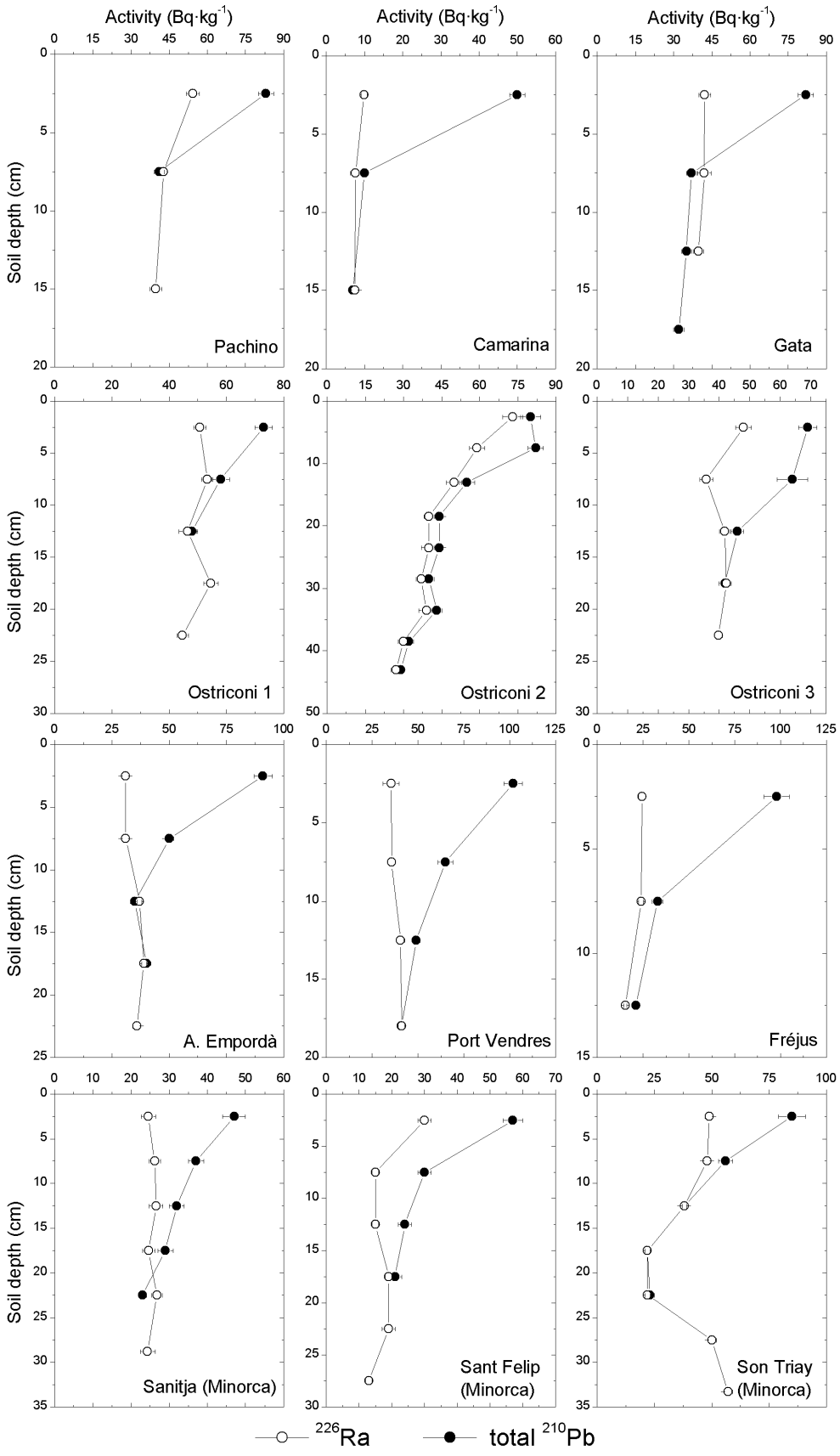

Figure 2. The $^{210}Pb_{total}$ (solid circles) and ^{226}Ra (open circles) activity profiles in soil cores collected from coastal areas in the western Mediterranean.

Table 2. Published ^{210}Pb Atmospheric Fluxes in the Western Mediterranean Sea Area

Site	Environment	Latitude (N), Longitude (E)	Rainfall, mm yr^{-1}	^{210}Pb Atmospheric Flux, Bq m^{-2} yr^{-1}
Ebro Delta[a]	microbial mat	40°40′, 00°40′	558	81 ± 5
Monaco[b]	total atmospheric deposition (2 years)	43°42′, 07°25′	870	102
Radó Lake[c]	soil	42°38′, 00°46′W	1500	255 ± 8
Catalan Pyrenees[c]	soil	-	1250	186 ± 10
Lac de Korba[d]	shallow lake sediment	36°37′, 10°53′	450	116
Gaeret Ichkeul[d]	shallow lake sediment	37°10′, 09°37′	920	134
Pobla de Segur[e]	soil	38°11′, 02°27′	900	154 ± 8
Lleida[e]	soil	37°42′, 02°11′	395	63 ± 5
Camargue[f]	soil	43°30′, 04°30′	600	132 ± 7
Camargue[g]	bulk precipitation	-	600	82
Monaco[h]	-	43°42′, 7°25′	880	110

[a] *Sanchez-Cabeza et al.* [1999b].
[b] *Tateda et al.* [2003].
[c] *Appleby et al.* [2002].
[d] *Appleby et al.* [2001].
[e] *Costa* [2005].
[f] *Miralles et al.* [2004].
[g] *Hussain et al.* [1990].
[h] *Heyraud* [1982].

and I is the excess ^{210}Pb inventory in soils calculated from $I = \sum_{z=0}^{z=\infty} \rho(x) \cdot C(x) \cdot dx$, where $\rho(x)$ (kg m^{-3}) is the dry bulk density, dx is the soil thickness and $C(x)$ is the excess ^{210}Pb concentration (Bq kg^{-1}).

[14] Rainfall data were obtained from meteorological stations located close to sampling sites (Table 1). It is important to point out that the temporal series are not long for all stations, as the installation of meteorological stations has began only recently in most of the study area. The rainfall range of our sampling sites, from 196 mm yr^{-1} at Cabo Gata to 833 mm yr^{-1} at Fréjus is wide enough to represent different rainfall regimes in the basin.

3. Results and Discussion

3.1. Estimation of Atmospheric ^{210}Pb Flux From Soils

[15] The profiles of ^{210}Pb and ^{226}Ra activities are shown in Figure 2. Activities of ^{210}Pb decreased with depth, reaching equilibrium with ^{226}Ra. Excess ^{210}Pb extended down to a depth between 5 and 20 cm in the soil profiles, because of adsorption in the soil matrix [*He and Walling*, 1997; *Huh and Su*, 2004]. Surface total ^{210}Pb activities ranged from 46 ± 3 Bq kg^{-1} in Sanitja to 102 ± 5 Bq kg^{-1} in Port Vendres (Table 1). The ^{226}Ra activity was constant along most of the cores, although some variability was observed in some instances, possibly related to changes in soil composition. In the Son Triay soil core, the ^{226}Ra activity profile reflected this situation: ^{226}Ra activities were 50 Bq kg^{-1} in the top (clay) and decreased to 20 Bq kg^{-1} at 15–20 cm depth (limestone). Overall, ^{226}Ra activities ranged from 8.7 ± 1.0 Bq kg^{-1} in a beach sedimentary sandy soil in Camarina to 50 ± 4 Bq kg^{-1} in a clayey soil in Ostriconi.

[16] Atmospheric ^{210}Pb fluxes ranged from 34 ± 3 at Cabo Gata (196 mm^{-1}) to 121 ± 12 Bq m^{-2} yr^{-1} at Fréjus (883 mm^{-1}), with a mean value of 71 ± 27 Bq m^{-2} yr^{-1}. The mean value is slightly lower than the published data for the northwestern Mediterranean, which ranges from 81 Bq m^{-2} yr^{-1} obtained from a microbial mat in the Ebro River Delta (558 mm^{-1} rainfall) [*Sanchez-Cabeza et al.*, 1999b] to 132 Bq m^{-2} yr^{-1} estimated by *Miralles et al.* [2004] in Camargue (France) and 134 Bq m^{-2} yr^{-1} estimated by *Appleby et al.* [2001] in Gaeret Ichkeul Lake (Tunisia) (see Table 2). However, most previously published data on ^{210}Pb fluxes correspond to areas where precipitation is higher than 500 mm yr^{-1}, while extensive areas of the western Mediterranean are characterized by rainfall levels of 400 mm yr^{-1} or lower (coastal Magreb, southeastern Spain and South Sicily and Sardinia) [*Martin and Milliman*, 1997]. The results obtained from sites where more than one sample was collected were internally consistent. In Minorca Island (45 km long) the three cores were collected in the north coast of the island with a distance of 15 km between them; the atmospheric ^{210}Pb fluxes calculated for these cores ranged from 82 ± 14 to 85 ± 9 Bq m^{-2} yr^{-1}. In the case of the Ostriconi cores, they were collected within a distance of only several meters from each other; calculated atmospheric ^{210}Pb fluxes ranged from 59 ± 9 to 62 ± 8 Bq m^{-2} yr^{-1}.

[17] Indeed, atmospheric ^{210}Pb fluxes are strongly correlated with rainfall ($R^2 = 0.95$) (Figure 3a). Such a relationship

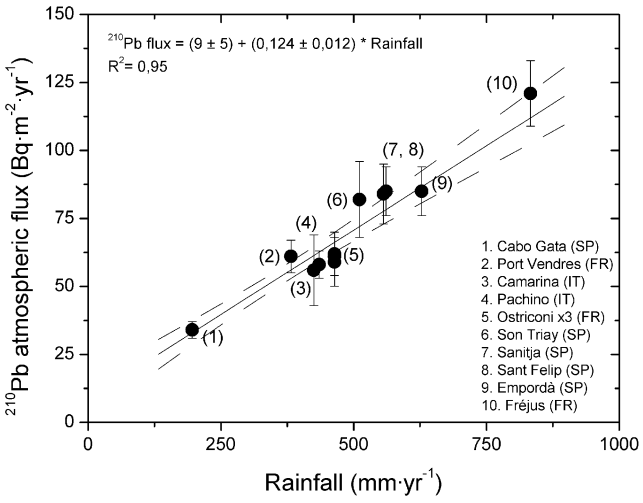

Figure 3a. Atmospheric ^{210}Pb fluxes from soil cores collected in this study versus rainfall in the western Mediterranean.

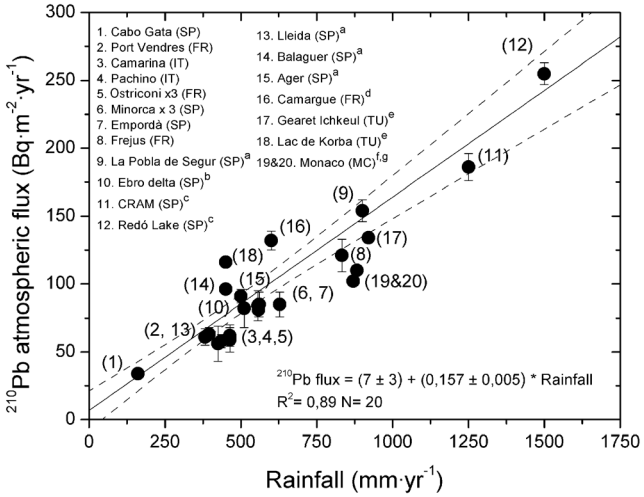

Figure 3b. Atmospheric ^{210}Pb fluxes versus rainfall taking into account data from other published works including Mediterranean noncoastal areas. Data are from *Costa* [2005] (labeled a), *Sanchez-Cabeza et al.* [1999] (labeled b), *Appleby et al.* [2002] (labeled c), *Miralles et al.* [2004] (labeled d), *Appleby et al.* [2001] (labeled e), (f) *Tateda et al.* [2003], and *Heyraud* [1982] (labeled g).

has been commonly observed, and explained on the basis that rainfall is the major depositional pathway for this radionuclide, as an efficient scavenger of the atmosphere [*Turekian et al.*, 1983; *Olsen et al.*, 1985; *Baskaran et al.*, 1993; *Caillet et al.*, 2001]. The distribution of ^{210}Pb fluxes against rainfall, considering data from other published works including Mediterranean noncoastal sites, is shown in Figure 3b. The rainfall varies from 196 mm^{-1} at Cabo Gata to 1500 mm^{-1} in Redó Lake (Pyrenees), corresponding to a ^{210}Pb flux range of 34 ± 3 to 255 ± 8 Bq m^{-2} yr^{-1}. The strong correlation (R^2 = 0.89) confirms that the model explains most of the atmospheric ^{210}Pb fluxes in the western Mediterranean. Then, the atmospheric flux of ^{210}Pb in a given location can be estimated if the average precipitation is well known. Taking into account that the mean precipitation in the western Mediterranean area ranges from 500 to 750 mm yr^{-1}, the predicted atmospheric ^{210}Pb flux from Figure 3b would fall between 80 and 120 Bq m^{-2} yr^{-1}, in agreement with most published works (Table 2).

3.2. Atmospheric ^{210}Pb Flux From Total Atmospheric Deposition

[18] The annual flux of ^{210}Pb in Ostriconi was measured during 1 year (16 September 2001 to 2 October 2002) by collecting total atmospheric deposition. The ^{210}Pb activities, rainfall and Saharan dust deposition during the sampling period are shown in Table 3. Each sampling period covers between 8 and 50 days with rainfall ranging from 0.2 to 102 L m^{-2}. The rain ^{210}Pb activities ranged from 57 to 1500 Bq m^{-3}, with a mean value of 372 ± 6 Bq m^{-3}, omitting from this analysis an event corresponding to very low rainfall (<1 L m^{-2}) that led to an anomalously high ^{210}Pb concentration (40 kBq m^{-3}). Pb 210 activities in rain fluctuated widely and a seasonal behavior is not recognized, unlike in other studies [*Lee et al.*, 2002, 2003]. The maximum rain ^{210}Pb concentration was observed in a precipitation sample of only 0.2 L m^{-2} (6 March 2002 to 2 April 2002), while minimum activities generally corresponded to heavy rain periods (13 November 2001 to 2 January 2002). This has been previously reported by other authors [*Turekian et al.*, 1977; *Caillet et al.*, 2001; *McNeary and Baskaran*, 2003] and is due to the fact that, during dry periods, aerosols tend to accumulate in the lower troposphere, being efficiently washed down by the first rain, and subsequently diluted as rain continues. However, ^{210}Pb activities are relatively high for some collecting periods (i.e., 5–11 November 2001) with relatively high precipitation rates.

[19] Atmospheric ^{210}Pb fluxes calculated from total atmospheric deposition ranged from 134 ± 6 to 4507 ± 219 mBq m^{-2} d^{-1}, with a mean of 419 ± 7 mBq m^{-2} d^{-1} (153 ± 2 Bq m^{-2} yr^{-1}). This value decreases to 330 ± 5 mBq m^{-2} d^{-1} (121 ± 2 Bq m^{-2} yr^{-1}) when excluding the period 5–13 November 2002 with a large dust event (21.9 g m^{-2}). The atmospheric ^{210}Pb flux measured during 1 year is larger, by a factor of 2, than that estimated from soils collected nearby (61 ± 2 Bq m^{-2} yr^{-1}, Table 1), suggesting that the measurements at Ostriconi are influenced by the presence of another source of ^{210}Pb.

Table 3. The ^{210}Pb Activities and Fluxes, Rainfall and Saharan Dust Fallout at Ostriconi Site (Corsica) for Sampling Periods Covering 1 Year (19 September 2001 to 2 October 2002)

	Period		Sampling, days	Rainfall, L m^{-2}	Dust deposition, mg m^{-2}	^{210}Pb Activity Bq m^{-3}	^{210}Pb Flux, Bq m^{-2} d^{-1}
1	19/09/2001	01/10/2001	12	32.4	20	57 ± 2	156 ± 7
2	01/10/2001	05/11/2001	35	13.8	550	340 ± 15	134 ± 6
3	05/11/2001	13/11/2001	8	49.8	21925	726 ± 35	4507 ± 219
4	13/11/2001	02/01/2002	50	101.8	240	87 ± 3	177 ± 7
5	02/01/2002	04/02/2002	33	7.6	0	1503 ± 58	346 ± 13
6	04/02/2002	06/03/2002	30	18.0	1235	994 ± 51	596 ± 31
7	06/03/2002	02/04/2002	27	0.2	178	40244 ± 2048	299 ± 15
8	02/04/2002	01/05/2002	29	29.6	33	325 ± 17	331 ± 18
9	01/05/2002	02/06/2002	32	18.3	2068	686 ± 32	391 ± 18
10	02/06/2002	01/07/2002	29	23.8	740	427 ± 17	352 ± 14
11	01/07/2002	04/08/2002	34	17.5	1918	491 ± 22	254 ± 11
12	04/08/2002	31/08/2002	27	65.3	370	209 ± 10	500 ± 23
13	31/08/2002	19/09/2002	19	38.0	28	255 ± 11	513 ± 22
14	19/09/2002	02/10/2002	13	9.5	105	535 ± 23	389 ± 17
Range						57–40244	134–4507
Total				426	29410		
Mean						372 ± 6[a]	330 ± 5[a]

[a]Mean excluding period 7.

Table 4. The ^{210}Pb Activities in Particulate Fraction of Weekly Total Atmospheric Deposition Corresponding to Saharan Dust Events Collected at the La Castanya–Montseny Station

Sampling Date	Rainfall, L m^{-2}	Dust Deposition, mg m^{-2}	^{210}Pb, kBq kg^{-1}	^{210}Pb, Bq m^{-2}
04/11/1987	43	1013	3.7 ± 0.4	3.8 ± 0.4
05/12/1987	71	3989	2.57 ± 0.16	10.3 ± 0.6
06/05/1988	5	1402	4.7 ± 0.3	6.6 ± 0.4
27/06/1988	14	975	1.14 ± 0.07	1.1 ± 0.04
15/10/1990	105	776	6.9 ± 0.4	5.4 ± 0.3
23/10/1990	26	620	2.9 ± 0.3	1.8 ± 0.2
24/03/1991	169	19435	0.39 ± 0.09	7.7 ± 1.7
11/03/1992	23	2739	4.9 ± 0.3	13.6 ± 0.7
22/01/1996	31	218	9.7 ± 0.5	2.1 ± 0.1
29/01/1996	92	518	9.6 ± 0.6	5.0 ± 0.3
21/11/1996	270	6440	7.7 ± 0.4	50 ± 8
27/01/1997	15	623	8.4 ± 0.4	5.2 ± 0.3
20/08/1997	60	421	0.77 ± 0.07	0.32 ± 0.03
10/05/1999	10	168	2.4 ± 0.4	0.40 ± 0.05
17/05/1999	25	280	2.09 ± 0.14	0.59 ± 0.03
25/05/1999	25	249	1.30 ± 0.09	0.32 ± 0.02
17/08/1999	60	817	2.8 ± 0.2	2.3 ± 0.2
15/11/1999	80	5553	7.9 ± 0.5	43.9 ± 2
17/05/2000	16	338	2.64 ± 0.18	0.89 ± 0.05
23/08/2000	41	238	2.96 ± 0.17	0.70 ± 0.04
07/09/2000	20	75	1.09 ± 0.07	0.08 ± 0.04
03/04/2002	36	50	5.5 ± 0.4	0.28 ± 0.03
Range		50–19435	0.39–9.7	
Mean		2134	4.2 ± 1.5	

[20] During the study period, a total of 29 g m^{-2} of Saharan dust were sampled, including a large event (21.9 g m^{-2}) in the period 5–13 November 2001, 3 events ranging from 1.2–2.1 g m^{-2} and other smaller events during the rest of the year (see Table 3). This annual flux is much larger than the average 12 g m^{-2} yr^{-1} reported for Corsica [*Loÿe-Pilot and Martin*, 1996] and the 4.6 g m^{-2} yr^{-1} at La Castanya–Montseny station [*Avila et al.*, 1997], and reflects the high variability both in space and time of the Saharan dust transport to the Mediterranean [*Loÿe-Pilot and Martin*, 1996; *Guerzoni et al.*, 1999].

3.3. Saharan Dust Enhancement of the Atmospheric ^{210}Pb Flux

[21] The fraction of the atmospheric ^{210}Pb flux associated with Saharan dust was estimated by analyzing the particulate fraction of weekly total atmospheric deposition corresponding to 22 dust events collected at La Castanya–Montseny during the period 1983–2002 (Table 4). Dust deposition was highly variable from year to year, with four events accounting for 70% of total dust deposition in the 17-year record (Figure 4). The annual dust deposition (4.6 g m^{-2} yr^{-1}) is low when compared with areas where dust storms are more frequent, such as the central and eastern Mediterranean Sea, where the average deposited annual mass fluxes have been estimated to be 12 and 35 g m^{-2} yr^{-1}, respectively [*Guerzoni et al.*, 1999].

[22] The ^{210}Pb activities ranged from 0.39 ± 0.09 to 9.7 ± 0.5 kBq kg^{-1}, with a mean value of 4.2 ± 1.5 kBq kg^{-1} (see Table 4). Fine dust aerosols transported by air currents from the Sahara desert to the Mediterranean area scavenge ^{210}Pb from the atmosphere, increasing significantly their ^{210}Pb concentration and therefore the atmospheric ^{210}Pb flux where precipitation occurs. This high ^{210}Pb concentration in aerosols increase the atmospheric ^{210}Pb flux. In order to estimate the supported ^{210}Pb in dust, one sample corresponding to a high deposition event (19.4 g m^{-2}, 24 March 1991) was analyzed by gamma spectrometry, showing a ^{226}Ra activity of 16 ± 8 Bq kg^{-1}. Therefore,

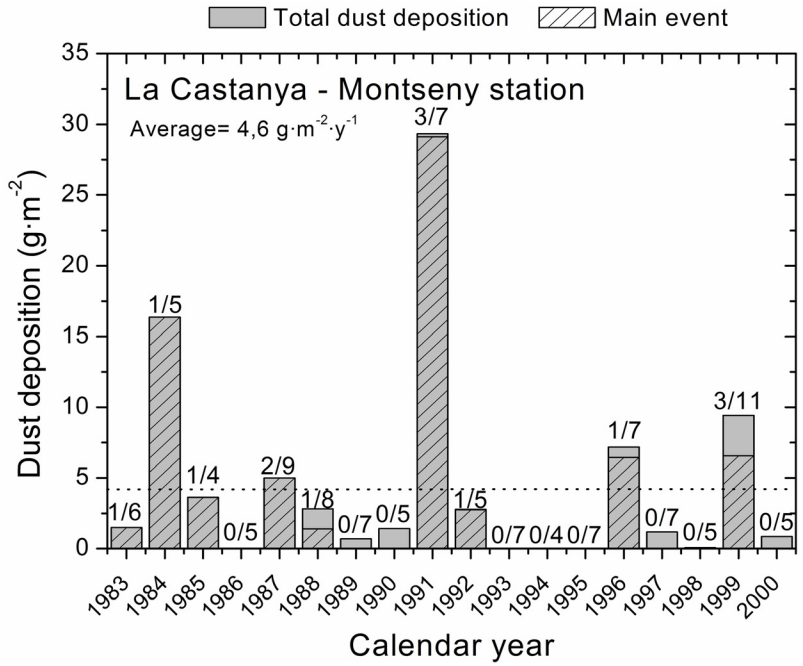

Figure 4. Annual dust deposition during the period 1983–2000 at the La Castanya–Montseny biological station. The relation between Saharan dust events and the contribution of main events (dust deposition > 1 g m^{-2}) shows that few main events account for the majority of Saharan dust deposition in the 17-year record.

although there is some contribution of ^{210}Pb from the grain dust, it represent less than 1% of the average ^{210}Pb activity scavenged in the samples. The ^{210}Pb fluxes ranged from 0.08 to 50 Bq m^{-2} per event, leading to an average annual atmospheric flux of ^{210}Pb associated with Saharan dust inputs of 20 Bq m^{-2} yr^{-1}. The ^{210}Pb flux from a soil collected in the Montseny area was estimated at 120 ± 9 Bq m^{-2} yr^{-1}, close to the regression estimation from rainfall (140 ± 5 Bq m^{-2} yr^{-1} for 850 mm yr^{-1}, Figure 3b). Therefore the ^{210}Pb flux associated with Saharan dust deposition would account for about 16% of the total atmospheric deposition. This value is lower than the 39% estimated by *Appleby et al.* [2002], who compared the total ^{210}Pb flux obtained in a soil close to the Redó Lake in the Pyrenees (255 Bq m^{-2} yr^{-1}) with the ^{210}Pb activity of 3.2 kBq kg^{-1} measured in a single dust sample collected in "red" snow precipitation, and using a rather large Saharan dust deposition (30 g m^{-2} yr^{-1}) in the area.

[23] On the basis of the data obtained and discussed above from this and other studies, an estimation of the ^{210}Pb deposition fluxes can be obtained for the western Mediterranean area by making some simplifications. If we assume a medium-term impact of Saharan dust events in the western Mediterranean area, ranging from 3 to 12 g m^{-2} yr^{-1} [*Guerzoni et al.*, 1999] and a mean ^{210}Pb activity in Saharan dust of 4.2 kBq kg^{-1}, then the ^{210}Pb flux associated with Saharan dust deposition would range from 13 to 50 Bq m^{-2} yr^{-1}. These fluxes could account for a Saharan dust ^{210}Pb flux greater than 50% in sites with low precipitation, and therefore low annual ^{210}Pb flux.

4. Conclusions

[24] Undisturbed soils, which are excellent environmental archives of integrated atmospheric fluxes, were used to estimate atmospheric ^{210}Pb fluxes to the western Mediterranean region. These ^{210}Pb fluxes were strongly correlated with mean annual rainfall, indicating that wet deposition is the most important transfer mechanism from the lower troposphere to the terrestrial and aquatic ecosystems in this area. Therefore atmospheric ^{210}Pb flux in a given area of the western Mediterranean basin can be estimated by using this correlation if the mean annual rainfall is known, thus facilitating the use of ^{210}Pb as a tracer of biogeochemical processes in the Mediterranean Sea and in soil erosion studies.

[25] The atmospheric ^{210}Pb flux is enhanced by Saharan dust events. Saharan dust is enriched in ^{210}Pb because atmospheric aerosols scavenge, amongst other substances, ^{210}Pb present in the atmosphere from decay of ^{222}Rn, thus significantly increasing its deposition. In the Montseny area this represents 16% of the annual flux but this value is obviously larger in areas with more Saharan dust deposition. Therefore the estimation of atmospheric ^{210}Pb fluxes to the Mediterranean area requires either long time series records, which integrate the interannual variability and especially the influence of irregular Saharan dust events, or the use of integrated archives such as undisturbed soils. Furthermore, the irregular input of ^{210}Pb associated with Saharan dust events to the Mediterranean Sea should be seriously considered in biogeochemical studies carried out using this marine radiotracer.

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References

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

Appleby, P. G., A. O. Koulokov, L. Camarero, and M. Ventura (2002), The input and transmission of fallout radionuclides through Redó, a high mountain lake in the Spanish Pyrenees, *Water Air Soil Pollut.*, *2*, 19–31.

Avila, A., and M. Alarcón (1999), Relationship between precipitation chemistry and meteorological situations at a rural site in NE Spain, *Atmos. Environ.*, *33*, 1663–1677.

Avila, A., and F. Rodà (2002), Assessing decadal changes in rainwater alkalinity at a rural Mediterranean site in the Montseny Mountains (NE Spain), *Atmos. Environ.*, *36*, 2881–2890.

Avila, A., I. Queralt-Mitjans, and M. Alarcón (1997), Mineralogical composition of African dust delivered by red rains over northeastern Spain, *J. Geophys. Res.*, *102*, 21,977–21,996.

Baskaran, M., C. H. Coleman, and P. H. Santschi (1993), Atmospheric depositional fluxes of ^{7}Be and ^{210}Pb at Galveston and College-Station, Texas, *J. Geophys. Res.*, *98*, 20,555–20,571.

Benninger, L. K., R. C. Aller, J. K. Cochran, and K. K. Turekian (1976), Lead-210 Geochronology of contemporary near-shore sediments: Status and problems transactions (abstract), *Eos Trans. AGU*, *57*(12), 931–932.

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

Caillet, S., P. Arpagaus, F. Monna, and J. Dominik (2001), Factors controlling ^{7}Be and ^{210}Pb atmospheric deposition as revealed by sampling individual rain events in the region of Geneva, Switzerland, *J. Environ. Radioact.*, *53*, 241–256.

Chester, R., M. Nimmo, and P. A. Corcoran (1997), Rain water aerosol trace metal relationships at Cap Ferrat: A coastal site in the western Mediterranean, *Mar. Chem.*, *58*, 293–312.

Costa, E. (2005), Processos de sedimentació i distribució de radioactivitat natural i artificial en sistemes aquàtics continentals de Catalunya (Embassaments de Flix i tram català del riu Ebre, Camarasa i llacs pirinencs), Ph.D. thesis, 262 pp, Univ. Autòn. de Barcelona, Barcelona, Spain.

DeMaster, D. J., D. C. Brewster, B. A. McKee, and C. A. Nittrouer (1991), Rates of particle scavenging, sediment reworking and longitudinal ripple formation at the Hebble site based on measurements of the Th-234 and Pb-210, *Mar. Geol.*, *99*, 423–444.

Escudero, M., S. Castillo, X. Querol, A. Avila, M. Alarcón, M. M. Viana, A. Alastuey, E. Cuevas, and S. Rodriguez (2005), Wet and dry African dust episodes over eastern Spain, *J. Geophys. Res.*, *110*(D18), D18S08, doi:10.1029/2004JD004731.

Graustein, W. C., and K. K. Turekian (1986), ^{210}Pb and ^{137}Cs in air and soils measure the rate and vertical profile of aerosol scavenging, *J. Geophys. Res.*, *91*, 14,355–14,366.

Guerzoni, S., et al. (1999), The role of atmospheric deposition in the biogeochemistry of the Mediterranean Sea, *Prog. Oceanogr.*, *44*, 147–190.

He, Q., and D. E. Walling (1997), The distribution of fallout Cs-137 and Pb-210 in undisturbed and cultivated soils, *Appl. Radiat. Isotopes*, *48*, 677–690.

Heyraud, M. (1982), Contribution a l'étude du Po-210 et Pb-210 dans les organismes marins et leur environment, Ph.D. thesis, 144 pp., Univ. é Pierre et Marie Curie, Paris.

Huh, C. A., and C. C. Su (2004), Distribution of fallout radionuclides (^{7}Be, ^{137}Cs, ^{210}Pb and ^{239,240}Pu) in soils of Taiwan, *J. Environ. Radioact.*, *77*, 87–100.

Hussain, N., T. Church, M. Heyraud, S. Fowler, S. Heussner, A. Monaco, P. Biscaye, and F. Anderson (1990), Systematics of radon daughters (^{210}Pb–^{210}Po): Atmospheric fallout to coastal regions (abstract), *Eos Trans. AGU*, *71*(2), 72.

Kaste, J. M., A. J. Friedland, and S. Sturup (2003), Using stable and radioactive isotopes to trace atmospherically deposited Pb in montane forest soils, *Environ. Sci. Technol.*, *37*, 3560–3567.

Koide, M., A. Soutar, and E. D. Goldberg (1972), Marine geochronolgy with Pb-210, *Earth Planet. Sci. Lett.*, *14*, 442–446.

Lambert, G. (1963), Etude du comportement des aérosols radioactifs artificiels atmosphérique, Thèse d'Etat, Fac. des Sci. de l'Univ. é de Paris, Paris.

Lee, S. H., M. K. Pham, and P. P. Povinec (2002), Radionuclide variations in the air over Monaco, *J. Radioanal. Nucl. Chem.*, *254*, 445–453.

Lee, S. H., J. J. La Rosa, I. Levy-Palomo, B. Oregioni, M. K. Pham, P. P. Povinec, and E. Wyse (2003), Recent inputs and budgets of ^{90}Sr, ^{137}Cs, ^{23,9240}Pu and ^{241}Am in the northwest Mediterranean Sea, *Deep Sea Res., Part II*, *50*, 2817–2834.

Likens, G. E., F. H. Bormann, R. S. Pierce, J. S. Eaton, and N. M. Johnson (1977), *Biogeochemistry of a Forested Ecosystem*, Springer, New York.

Loÿe-Pilot, M. D., J. M. Martin, and J. Morelli (1986), Influence of Saharan dust on the rain acidity and atmospheric input to the Mediterranean, *Nature*, *321*, 427–428.

Loÿe-Pilot, M. D., and J. M. Martin. (1996), Saharan dust input to the western Mediterranean: An eleven years record in Corsica, in *The Impact of Desert Dust From Northeastern Africa Across the Mediterranean*, edited by S. Guerzoni and R. Chester. pp. 191–199, Springer, New York.

Martin, J. M., and J. D. Milliman (1997), EROS 2000 (European River Ocean System). The western Mediterranean: An introduction, *Deep Sea Res., Part II*, *44*, 521–529.

Masqué, P., J. Fabres, M. Canals, J. A. Sanchez-Cabeza, A. Sanchez-Vidal, I. Cacho, A. M. Calafat, and J. M. Bruach (2003), Accumulation rates of major constituents of hemipelagic sediments in the deep Alboran Sea: A centennial perspective of sedimentary dynamics, *Mar. Geol.*, *193*, 207–233.

McNeary, D., and M. Baskaran (2003), Depositional characteristics of ^{7}Be and ^{210}Pb in southeastern Michigan, *J. Geophys. Res.*, *108*(D7), 4210, doi:10.1029/2002JD003021.

Miralles, J., O. Radakovitch, J. K. Cochran, V. Véron, and P. Masqué (2004), Multitracer study of anthropogenic contamination records in the Camargue, southern France, *Sci. Total Environ.*, *320*, 63–72.

Monaghan, M. C. (1989), Lead 210 in surface air and soils from California: Implications for behavior of trace constituents in planetary boundary layer, *J. Geophys. Res.*, *94*, 6449–6456.

Moore, H. E., S. E. Poet, and E. A. Martell (1973), ^{222}Rn, ^{210}Pb, ^{210}Bi, and ^{210}Po profiles and aerosol residence times versus altitude, *J. Geophys. Res.*, *78*, 7065–7075.

Nozaki, Y., H. Tsubota, V. Kasemsupaya, M. Yashima, and N. Ikuta (1991), Residence times of surface-water and particle-reactive Pb-210 and Po-210 in the East China and Yellow seas, *Geochim. Cosmochim. Acta*, *55*, 1265–1272.

Nozaki, Y., D. J. Demaster, D. M. Lewis, and K. K. Turekian (1978), Atmospheric ^{210}Pb fluxes determined from soil profiles, *J. Geophys. Res.*, *83*, 4047–4051.

Olsen, C. R., I. L. Larsen, P. D. Lowry, N. H. Cutshall, J. F. Todd, G. T. F. Wong, and W. H. Casey (1985), Atmospheric fluxes and marsh-soil inventories of ^{7}Be and ^{210}Pb, *J. Geophys. Res.*, *90*, 10,487–10,495.

Palanques, A., J. A. Sanchez-Cabeza, P. Masque, and L. Leon (1998), Historical record of heavy metals in a highly contaminated Mediterranean deposit: The Besos prodelta, *Mar. Chem.*, *61*, 209–217.

Preiss, N., M. A. Melieres, and M. Pourchet (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.*, *101*, 28,847–28,862.

Prospero, J. M., and T. N. Carlson (1972), Vertical and areal distribution of Saharan dust over the western equatorial North Atlantic Ocean, *J. Geophys. Res.*, *77*, 5255–5265.

Querol, X., A. Alastuey, S. Rodríguez, F. Plana, C. R. Ruiz, N. Cots, G. Massagué, and O. Puig (2001), PM10 and PM2.5 source apportionment in the Barcelona metropolitan area, Catalonia, Spain, *Atmos. Environ.*, *35*, 6407–6419.

Radakovitch, O., R. D. Cherry, and S. Heussner (1999), Particulate fluxes on the Rhône continental margin (NW Mediterranean). Part III: ^{210}Po and ^{210}Pb data and the particle transfer scenario, *Deep Sea Res., Part I*, *46*, 1539–1563.

Robbins, J. A., and D. N. Edgington (1975), Determination of recent sedimentation rates in Lake Michigan using Pb-210 and Cs-137, *Geochim. Cosmochim. Acta*, *39*, 285–304.

Roberts, K. A., J. K. Cochran, and C. Barnes (1997), ^{210}Pb and ^{239,240}Pu in the northeast water polynya, Greenland: Particle dynamics and sediment mixing rates, *J. Mar. Syst.*, *10*, 401–413.

Sanchez-Cabeza, J. A., P. Masqué, and I. Ani-Ragolta (1998), ^{210}Pb and ^{210}Po analysis in sediments and soils by microwave acid digestion, *J. Radioanal. Nucl. Chem.*, *227*, 19–22.

Sanchez-Cabeza, J. A., P. Masqué, I. Ani-Ragolta, J. Merino, M. Frignani, F. Alvisi, A. Palanques, and P. Puig (1999a), Sediment accumulation rates in the southern Barcelona continental margin (NW Mediterranean Sea) derived from ^{210}Pb and ^{137}Cs chronology, *Prog. Oceanogr.*, *44*, 313–332.

Sanchez-Cabeza, J. A., P. Masqué, M. Martínez-Alonso, J. Mir, and I. Esteve (1999b), ^{210}Pb atmospheric flux and growth rates of a microbial mat from the northwestern Mediterranean Sea (Ebro River Delta), *Environ. Sci. Technol.*, *33*, 3711–3715.

Shotyk, W., D. Weiss, M. Heisterkamp, A. K. Cheburkin, P. G. Appleby, and F. C. Adams (2002), New peat bog record of atmospheric lead pollution in Switzerland: Pb concentrations, enrichment factors, isotopic composition, and organolead species, *Environ. Sci. Technol.*, *33*, 3893–3900.

Tateda, Y., F. P. Carvalho, S. Fowler, and J. C. Miquel (2003), Fractionation of ^{210}Po and ^{210}Pb in coastal waters of the NW Mediterranean continental margin, *Cont. Self Res.*, *23*, 295–316.

Turekian, K. K., L. K. Benninger, and E. P. Dion (1983), ^{7}Be and ^{210}Pb total deposition fluxes at New Haven, Connecticut and at Bermuda, *J. Geophys. Res.*, *88*, 5411–5415.

Turekian, K. K., Y. Nozaki, and L. K. Benninger (1977), Geochemistry of atmospheric radon and radon products, *Earth Planet. Sci. Lett.*, *5*, 227–255.

Wei, C. L., and J. W. Murray (1994), The behavior of scavenged isotopes in marine anoxic environments: ^{210}Pb and ^{210}Po In the water column of the Black Sea, *Geochim. Cosmochim. Acta*, *58*, 1795–1811.

Zuo, Z., D. Eisma, R. Gieles, and J. Beks (1997), Accumulation rates and sediment deposition in the northwestern Mediterranean, *Deep Sea Res., Part II*, *44*, 597–609.

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