ABSTRACT

The Sancho Reservoir (SW Spain) was built in 1962, about the time of maximum ^{137}Cs fallout, and it has been affected by acid mine drainage (AMD) particularly since the mining cease in 2001. This is a unique scenario for studying the radiogeochronological fingerprints in AMD-affected sediments deposited over the former flood plain. A sediment core sampled in 2011 was analysed for bulk density, ^{137}Cs, ^{239}Pu, ^{240}Pu, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{234}Th (^{238}U) and ^{40}K, and studied with various radiometric dating models. Bulk density revealed unsteady compaction and likely depositional events. The activity concentrations of ^{226}Ra, ^{228}Ra, ^{234}Th (^{238}U) and ^{40}K were uniform down-core, but declining overall in the upper 0-25 cm, revealing changes in provenance except for ^{238}U, which increased in the top 10 cm likely due to its supply by AMD. The AMD fingerprint was also found in the $^{239+240}$Pu/^{137}Cs activity ratio, which increased in the top sediment layers. The ^{137}Cs and $^{239+240}$Pu profiles show well defined peaks at the same depth, with inventories being about four times higher than the expected integrated atmospheric deposition in the area. The unsupported ^{210}Pb (^{210}Pb$_{exc}$) showed a complex non-monotonic profile interrupted at several sections, particularly around the ^{137}Cs peak. The whole dataset cannot be interpreted in terms of continuous sedimentation processes. Based upon correlated features in the bulk density and ^{210}Pb$_{exc}$ profiles, a series of depositional events (likely linked to peaks in the rainfall records) have been identified in the core. These events date back to the period comprised since the construction of the dam until its increase in height in 1972, which likely displaced upstream the main depositional area of riverine loads, as inferred from sediment trap

data. The CRS (with a reference date) and (a piecewise) CIC models have been used for complementing and discussing the chronology.

Keywords: • Sediment dating; • Reservoir; • Radiogeochronological fingerprints; •Depositional events; • Piecewise CIC

1. Introduction

Since the beginning of the Industrial Revolution, human influence on the environment has increased substantially. For this period of time, several radionuclides have proved useful tools for establishing chronologies in sediment cores and other reservoirs, which allows examination of the impact of the anthropogenic influence in the surrounding environment (Asmund and Nielsen, 2000; Audry et al., 2004).

The ^{210}Pb dating method is based on the particular cycle of this radionuclide in nature (Appleby and Oldfield, 1992; El-Daoushy, 1988; Robbins, 1978). After its production in the atmosphere (from radioactive decay of ^{222}Rn), ^{210}Pb is removed primarily by precipitation and dry deposition processes and it can follow a wide diversity of pathways to reach the sediment-water interface (SWI). This is known as the ^{210}Pb unsupported fraction or in excess ($^{210}Pb_{exc}$), while the supported fraction of ^{210}Pb is produced by the radioactive decay of ^{226}Ra present in the sediment, and is generally assumed to be in secular equilibrium with its parent radionuclide. The $^{210}Pb_{exc}$ decays with its own half-life (22.3 y) and is the basis of ^{210}Pb dating technique (Robbins, 1978). Since the first time it was used in glaciers from Greenland (Goldberg, 1963), the ^{210}Pb-based dating of freshwater and coastal sediments has been extensively applied over the past 50 years for studies of reconstruction of pollution records, sediment focusing, sediment accumulation rate (SAR), and mixing rate determination (e.g. Baskaran et al., 2014; Couillard et al., 2004; San Miguel et al., 2003).

There are many factors that can alter the ^{210}Pb versus depth profile in the sediment, such as sediment mixing, the biologically-mediated alteration of SARs, or chemical remobilisation (Putyrskaya et al., 2015; Robbins and Edgington, 1975).

Unfortunately, there is no universal model that can be applied to any case study. Moreover, for any given $^{210}Pb_{exc}$ profile, and in absence of restrictive assumptions, there is an infinite number of mathematically exact solutions for the chronology (Abril, 2015). The validation of the ^{210}Pb-based chronologies with some additional chronostratigraphic marks is then essential to be confident with results (Smith, 2001). This is commonly accomplished by the combination of ^{210}Pb with ^{137}Cs measurements in the sediments (e.g. Jha et al., 2003; San Miguel et al., 2003).

Caesium-137 ($T_{1/2}$=30.2 y) is an anthropogenic radionuclide originating from atmospheric nuclear weapons testing carried out from 1945 to 1972, and from some major nuclear accidents (e.g. Chernobyl in 1986 and Fukushima in 2011). If the ^{137}Cs vertical profile in a sediment core has not been significantly affected by post-depositional processes, it is expected to show a well-defined peak corresponding to the years of its maximum concentration in the atmosphere (1962-1963). The use of ^{137}Cs as an indicator of sedimentation processes is consistent as it binds almost irreversibly to clay and silt particles (Audry et al., 2004).

Plutonium isotopes have been also used as a complementary method of sediment dating (Putyrskaya et al., 2015). As in the case of ^{137}Cs, nuclear weapons testing also released significant amounts of Pu which were distributed into the atmosphere. In addition, the releases from nuclear fuel cycling facilities are regional sources of Pu contamination (Lindhal et al., 2011; MacKenzie et al., 2006). The activity ratios $^{239+240}$Pu/^{137}Cs in environmental samples can be useful as an indicator of contamination coming from a source other than radioactive fallout (Hodge et al., 1996; Wu et al., 2010).

Several constraints have been identified in the use of ^{137}Cs as a chronostratigraphic marker (Abril, 2003a). One of them is its potential mobility in the

sediment profile, especially in saline sediments (Hancock et al., 2011). Plutonium, on the other hand, remains particle-reactive in both fresh and saline waters. Nevertheless, some cases have been found in estuarine sediments in which Plutonium is significantly associated to exchangeable phases (Lucey et al., 2004).

Radiometric dating of sediments from reservoirs is particularly challenging, with complex non-monotonic $^{210}Pb_{exc}$ profiles (Anjum et al., 2017; Chen et al., 2014). Among other difficulties we can mention: i) in those systems affected by severe erosion, SARs can surpass one metre per year (in these cases the GIS-bathymetry is a more appropriate tool; Khaba and Griffiths, 2017); ii) the watershed-dominated inputs of matter and radiotracers onto the SWI may show high temporal variability (McCall et al., 1984); iii) in most cases reservoirs are not old enough for allowing steady-state inventories of $^{210}Pb_{exc}$ in sediments (as required for applying the standard CRS model); iv) depending on their age, the expected peaks in the profiles of artificial fallout radionuclides may be absent.

Because of the above issues, the radiometric dating of recent sediments has become a complex task in which all the available tracers (e.g., stable lead , Chi et al., 2009; pollen markers, Chen et al., 2014; etc..) and sources of evidence (e.g., bulk density profiles, Abril, 2011) must be analysed.

The effects on radionuclide concentrations in sediments due to progressive acidification of the aquatic system, and particularly their implications in the radiometric dating, have not been studied in detail. This study addresses this issue by analysing bulk density and the activity concentration profiles of ^{137}Cs, ^{239}Pu, ^{240}Pu, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{234}Th (^{238}U) and ^{40}K in a sediment core from the Sancho reservoir (SW Spain), which has been drastically impacted by acid mine drainage (AMD). The construction of the dam ended in late 1962, about the time of maximum ^{137}Cs and $^{239+240}Pu$ fallout. It was

heightened in 1972, almost doubling the capacity of the reservoir and displacing upstream the major depositional area of sediments transported by the Meca River. The reservoir is strongly affected by AMD, particularly since 2001, when the closure of the mining of Tharsis ceased the treatment of its waters (Cánovas et al., 2016). This scenario brings a unique opportunity to study the different radiogeochronological fingerprints in an AMD-affected sediment core, which records the transition from the former flood plain to aquatic sediments. The methodology (involving a multi-tracer approach and outstanding dating tools), results and discussion may be of general interest for environmental scientists.

2. Materials and methods

2.1. Site description

The Sancho Reservoir (Fig.1) has a capacity of 58 Mm^3. It was built in 1962 (works ended on December 31st) to supply water to a paper mill factory, and it has been used as a domestic water supply after treatment. The dam was enlarged in height to increase (almost doubling) the capacity of the reservoir, with works ending on January 1st 1972. This water body has a surface area of 4.27 km^2 and a maximum depth of 40 m. It is mainly fed by the Meca River, with a catchment area of 314 km^2 and an average stream flow of 61 Mm^3/year. The studied site has a Mediterranean-type climate with an average temperature of 19 ºC and an average annual rainfall of 614 mm. About 60% of rainfall occurs between October and January, although the precipitation is subject to great inter-annual and intra-annual variability (Galván et al., 2012).

In the headwaters of the Meca River, the huge mining complex of Tharsis is located (Cánovas et al., 2016). Due to the intense mining activities, there is an extensive

132 area of flooded open-pits, galleries, shafts and mining wastes that release metal and
133 acidity to the Meca watershed, finally reaching the Sancho Reservoir (Cánovas et al.,
134 2016; Galván et al., 2012; Torres et al., 2013). These pollutants are mainly transferred
135 to the bottom sediments where high metal concentrations are observed in pore waters
136 (Sarmiento et al., 2009; Torres et al., 2013). Torres et al. (2015) estimated a removal of
137 As, Fe and Mn of around 98, 80 and 70 %, respectively, and around 10% for Al and
138 other metals (i.e. Zn and Cu). However, this removal pattern of pollutants seems to have
139 changed due to increasing acidification of reservoir waters (up to pH ~3.0 nowadays) by
140 AMD after mine closure; and Fe seems to be replacing Al as a buffering agent (Cánovas
141 et al., 2016). In addition, the Meca River transports huge amount of Fe-rich particulate
142 matter during floods, which undergoes chemical transformations in the reservoir
143 sediments (Cánovas et al., 2016).
144
145 *2.2. Sampling and sample treatment*
146
147 In late 2011, an 80 cm long sediment core was collected by scuba diving using a
148 manual corer consisting of a 5.5 cm inner diameter cylindrical PVC tube. The sampling
149 site was at 6º58.972'W and 37º27.697'N, close to the dam (Fig. 1), and at the time of
150 sampling the water depth was 36 m. The sediment core was frozen in the laboratory and
151 sliced in horizontal sections with a plastic cutter at 2 cm resolution, except the top layer
152 that was sliced at 3 cm. Sediment samples were dried at 60 º C to constant weight and
153 then powdered and homogenised. From the wet and dry weights of the sediments, water
154 content was determined. Porosity (ϕ) for each section was determined using the
155 following equation:
156 $$\phi = \frac{1}{1+\frac{\rho_W m_D}{\rho_D m_W}} \quad (1)$$

where ρ_W is the water density (assumed to be 1 g cm^{-3}); ρ_D is the particulate matter density (assumed to be 2.45 g cm^{-3}); m_W is the water mass; and m_D is the mass of solids in each layer. The bulk density, ρ, can be then estimated as $\rho = \rho_D(1-\phi)$. The cumulative sediment mass per unit area (g cm^{-2}) from the SWI till depth z, $m(z)$, can be obtained as follows:

$$m(z) = \int_0^z \rho dz' \quad (2)$$

In practice, the integral is replaced by a discrete summation. Mass depth can be used instead of the true depth to compensate the effects of sediment compaction and the shortening in the coring and storage processes. Torres et al. (2013) sampled another sediment core in October 2010 at approximately the same location to support their study on trace metals cycling during sediment early diagenesis. The above study also reported results on sediment traps deployed for a whole hydrologic year (September 2009-2010), which will be useful for our present goals. The traps consisted of vertical tubes (6 cm diameter × 50 cm) placed at the centre of the reservoir (at around 30-33 m depth) and close to the river entrance (at around 5-7 m depth), more than 3 m from the bottom to prevent the recollection of re-suspended material (see locations in Fig. 1).

2.3. Gamma Spectrometry

An aliquot of about 4-5 g of each homogenised sample was sealed and stored in 5 ml cylindrical containers of polyethylene for at least one month before radionuclide determinations through gamma spectrometry. This allows for secular equilibrium between ^{226}Ra and its shorter half-life daughter ^{222}Rn. The sample containers were filled to avoid the differences in efficiency as a consequence of the diffusion of ^{222}Rn within it.

The activity concentrations were determined by gamma spectrometry for the following set of radionuclides (the selected gamma-emissions appear in brackets): ^{210}Pb (46.5 keV), ^{234}Th (63.3 keV), ^{137}Cs (661 keV), ^{228}Ac (911.3 keV), in secular equilibrium with ^{228}Ra, ^{40}K (1460 keV) and ^{214}Pb (352 keV), in secular equilibrium with ^{226}Ra. As measurements were done about one year after sampling, ^{234}Th is expected to be in secular equilibrium with ^{238}U.

Measurements were carried out in a Well Ge detector (Canberra), with a full-width at half-maximum (FWHM) of 1.33 keV at 122 keV (^{57}Co) and 2.04 keV at 1332 keV (^{60}Co), and a peak/Compton ratio of 56.2/1. The detector was coupled to a multichannel analyser and was shielded with 10 cm thickness lead shield. To avoid interferences from X-ray from the Pb of the shield, a 2 mm thick layer of Cu was placed between the Pb shield and the detector. Efficiency calibration with self-absorption corrections were determined according to procedures detailed elsewhere (Appleby and Piliposian, 2004). The minimum detectable activity (MDA) for each radionuclide was obtained following the method by Currie (2004). The MDA for counting times of two days was about 100 mBq for ^{210}Pb, 70 mBq for ^{234}Th, 30 mBq for ^{226}Ra (^{214}Pb), 20 mBq for ^{137}Cs, 50 mBq for ^{228}Ra (^{228}Ac) and 200 mBq for ^{40}K.

The quality assurance of gamma measurements was regularly ensured through participation in inter-comparison exercises organised by the International Atomic Energy Agency (IAEA) and the CSN (Nuclear Spanish Organism), as well as the periodic measurement of certified reference materials.

2.4. *^{239,240}Pu determination*

For plutonium determinations, dried and homogenised aliquots of each sediment slice were used, with masses ranging from ~0.3 to approximately 5.0 g. Samples were weighed into a 20-ml borosilicate glass vial and then dry-ashed at 600 °C for 16 h to remove organic matter. Then, 0.007 Bq (~50 pg) of ^{242}Pu (IRMM-085) was added as a spike for isotope dilution analysis. The native ^{242}Pu content in the sample is negligible compared to the amount of spike added. Ten ml of 16 M HNO_3 was added and the mixture was heated to 75–80 °C for 16 h. The mixture was then diluted with deionised water to 20 ml, filtered, and 0.4 g $NaNO_2$ solution was added to reduce Pu to Pu(IV). Next, 0.06 g of TEVA resin beads (EIChrom, Darien, IL) were used to retain Pu (Horwitz et al., 1995). The resin was collected in a pipet tip, and was rinsed using 5 ml of 2 M HNO_3 followed by 1.5 ml of 8 M HCl. After the rinse steps, Pu is eluted from the columns with a sequence of 1 ml of water, 1 ml of 0.05 M ammonium oxalate, and 1 ml of water. The 3.0 ml fraction is then used directly for ICP-MS analysis. Pu isotopes were analysed using a Thermo X Series II quadrupole ICP-MS equipped with an APEX sample introduction system and PFA Teflon nebuliser. The sample solution was introduced at 300 μl/min rate.

A potential problem in the determination of Pu by ICPMS is the presence of isobaric $^{238}U^{1}H^{+}$ at mass 239 (Ketterer et al., 2004). It has been shown that the yield of $^{238}U^{1}H^{+}$, expressed as $^{238}U^{1}H^{+}/^{238}U^{+}$, is on the order of 0.00001–0.00004. The resolution of $^{238}U^{1}H^{+}$ and $^{239}Pu^{+}$ is not possible, and therefore U must be sufficiently removed from the sample solutions prior to measurement. Small amounts of U are tolerable, provided that a subtractive correction is applied:

$$^{239}Pu^{+} \text{ signal} = \text{Raw } m/z \text{ 239 signal} - (^{238}U^{1}H^{+}/^{238}U^{+})\ (^{238}U^{+}\text{signal}) \quad (3)$$

The HNO_3 and HCl rinse steps remove >99.999% of U and ~99.99% of Th (Kuehl et al., 2012). The removal of these elements reduces the isobaric interferences to

manageable levels ($^{238}U^{1}H^{+}$ on $^{239}Pu^{+}$). The UH^{+} correction is applied using Eq. (3) and a $^{238}U^{1}H^{+}/^{238}U^{+}$ ratio of $(22 \pm 3)\cdot10^{-6}$ was measured in the U standard solution. The application of Eq. (3) is not encumbered by interference of ^{238}Pu upon ^{238}U. Global fallout Pu exhibits a $^{238}Pu/^{239+240}Pu$ activity ratio of 0.04–0.05 (Mietelski and Was, 1995), corresponding to a $^{238}Pu/^{239}Pu$ atom ratio of ~0.0003; even for samples exhibiting ^{239}Pu signals of 10^{4} ions/s, the ^{238}Pu signal would amount to a negligible level of 3 ions/s.

Signals were collected for $^{238}U^{+}$, $^{239}Pu^{+}$, $^{240}Pu^{+}$ and $^{242}Pu^{+}$. After application of Eq. (3), raw ratio data ($^{240}Pu/^{239}Pu$, $^{239}Pu/^{242}Pu$ and $^{240}Pu/^{242}Pu$) was computed and corrected for mass discrimination based upon the $^{238}U/^{235}U$ determined for a modern coral solution (true value=137.88). The analysis of each sample involved three replicate peak-jump integrations and approximately 10 min of acquisition time.

The analyses of three aliquots of a control sample, MAPEP 01 S8, were performed in order to assess the accuracy of the activity data. This material consists of a clay soil spiked with various radionuclide, including $^{239+240}Pu$. Small quantities (30–70 mg) of this material were mixed with an inactive sandstone material to prepare larger solid samples simulating the quantities of unknown samples actually analysed. The masses of ^{239}Pu and ^{240}Pu present in the sample, determined by isotope dilution calculations, were converted into the summed $^{239+240}Pu$ activity. A detection limit of 0.01 Bq/kg $^{239+240}Pu$ was estimated for a sample of average nominal mass of 5 g.

3. Results and Discussion

Table 1 summarises the results on the measurements of bulk density and radionuclide activity concentrations in the core from the Sancho Reservoir. They are discussed in the subsequent sections.

A key question for understanding the present dataset is whether the core length includes only sediments accumulated after the construction of the dam or whether it also incorporates a portion of the former soil. Measurements on settling fluxes reported by Torres et al. (2013) for the hydrological year 2009-2010 (see deployment stations in Fig. 1) were of 3.4 g cm^{-2} y^{-1} in the upper reaches of the reservoir (about 7 km upstream of the dam) and less than 0.05 g cm^{-2} y^{-1} in the stations closer to the dam (0.049 g cm^{-2} y^{-1} in the location of the studied core). Rainfall collected in the hydrological year 2009-2010 was about 50% higher than the 1950-2010 mean value (data from the Spanish AEMET https://datosclima.es/Aemethistorico). From the above settling fluxes, it can be concluded that most of the sediment load supplied by the Meca River is now settled in the upstream reaches of the reservoir. Accounting to the combined effect of settling velocities and travel distance, only the very small grain size particles could reach the dam area. In this area, the provenance of mass flows into the SWI is expected to be dominated by Aeolian transport, inputs from the closer shoreline and internal production.

As sediment traps were deployed at least 3 m above the SWI, any effects of sediment resuspension or near-bottom transport of sediments can be neglected. These effects are expected to be of minor importance in the reservoir in modern conditions. Assuming these settling fluxes to be a good proxy for SAR, if these conditions prevailed during the elapsed time of 49 years since the construction of the dam, the cumulated mass depth would be of ~2.4 g cm^{-2} (~17 cm in the core –see Table 1). It still remains unknown whether the SAR values would have been significantly higher in the

279 past, although it can be expected that the increase in height of the dam in 1972 would
280 have resulted in a noticeable upstream displacement of the main depositional area of
281 sediments transported by the Meca River, with the consequent reduction of SAR near
282 the dam.

283 Figure ESM-1 (in electronic supplementary material) shows an historical aerial
284 photograph of the sampling site dated between 1956-1957. The sampling site can be
285 approximately located taking into account the main topographic features. At that time
286 the Meca River remained almost dry most of the year, with waters flowing only during
287 the wet season. The original conditions at the sampling site would have consisted of
288 vegetated soil on a slope hill or in floodplain soils.

289

290 *3.1. Physic-chemical characterisation of the sediment core*

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292 The sediment core consisted of dark, anoxic (dissolved O_2 saturation < 0.5% at
293 the SWI) and unconsolidated sediment with grain sizes ranging from fine sand to clay.
294 Some burrows were observed near the SWI. Nevertheless, the impact of bioturbation on
295 the sedimentary record should be negligible because of low density and sizes.

296 In the nearby core studied by Torres et al. (2013), the TOC values in the surface
297 of the sediment were about 12%, with a steep decrease with depth indicating the
298 reactive nature of organic matter. The TOC/N ratios in the sediment ranged from 9 to
299 13, which suggest a phytoplanktonic origin of the organic matter rather than cellulosic
300 land-plant provenance (Torres et al., 2013).

301 Measurements of bulk densities are reported in Table 1 and plotted in Figure 2.
302 It is not clear whether the anomalous value found in the last sediment slice (75 cm
303 depth) can be attributable to sampling artifacts. The porosity was over 90 % for the

upper 15 cm of the sediment core. The dotted line in Figure 2 represents the fit to a typical steady-state asymptotic bulk density profile (Abril, 2011). It is worth noting how the data noticeably deviates from the expected trend-line for a steady state early compaction. Particularly for the interval 0-30 cm, data follows an atypical parabolic trend with a positive curvature. The region 30-75 cm shows large fluctuations which can be interpreted as a sequence of depositional events. Discussion will be readdressed in the scope of the global dataset.

3.2. *Naturally-occurring radionuclides*

Figure 3 plots the normalised activity concentrations of ^{226}Ra, ^{228}Ra, ^{40}K and ^{234}Th versus depth in the sediment core. Normalisation refers to mean value within the area with almost uniform values at the core base (56-75 cm interval). The measurements of ^{234}Th (in secular equilibrium with ^{238}U since measurements were carried out around 1 year after sampling) are affected by large counting uncertainties, but the four radionuclides show similar and uniform normalised values in the 25-75 cm interval, indicating that the isotopic ratios remain near constant. The ^{234}Th (^{238}U)/ ^{226}Ra ratio is of 1.1 ± 0.2, being compatible with secular equilibrium. The ^{228}Ra/ ^{226}Ra ratio is close to unity in the earth crust, but the value of 1.36 ± 0.13 found in this work compares well with other values reported for aquatic sediments (e.g., ^{228}Ra/ ^{226}Ra~1.6 for estuarine sediments in Equatorial Africa, Klubi et al., 2017).

From the above uniform values, the activity concentrations of ^{228}Ra, ^{226}Ra and ^{40}K gradually decreases towards the SWI following a similar trend (more properly, the above three magnitudes are linearly correlated in the 0-25 cm interval wit R >0.98 and p < 0.05). This fact unambiguously reveals a change in the environmental conditions

likely related to the amount, provenance and mineralogical composition of the mass flow onto the SWI and/or processes related to the progressive acidification of the reservoir after the mine closure. On the contrary, the concentrations of ^{234}Th (^{238}U) show an increase towards the SWI, at least in the 0-8.5 cm interval, similar to the trend reported by Torres et al. (2013) in their nearby core for Cu, Zn, Cd, Ni and Co. The acid mining drainage is the likely source for the supply of these elements to the sediments.

^{210}Pb activity concentrations (Table 1) are contributed by their supported (from the in situ radioactive decay of ^{226}Ra) and unsupported ($^{210}Pb_{exc}$, from the radioactive decay of ^{222}Rn in the atmosphere) fractions. The $^{210}Pb_{exc}$ fraction can be estimated by subtracting the ^{226}Ra activity (assumed to be in secular equilibrium with the supported fraction) from the total ^{210}Pb activity in a layer-by-layer basis. Results are shown in Figure 4.

It is worth noting that $^{210}Pb_{exc}$ decreases with depth from the SWI and it cannot be distinguished from zero (at a 95% confidence level) at z_L= 25 cm depth (Fig. 4). The $^{210}Pb_{exc}$ inventory from SWI until such a depth is $\Sigma_0^{z_L} = \int_0^{z_L} A(z)\rho dz$ = 1740 ± 120 Bq m^{-2} (with $A(z)$ the $^{210}Pb_{exc}$ activity concentration at depth z; linear interpolations have been used for the non-measured slices in Table 1). If this 0-25 cm interval is interpreted as the sediment accumulated over the former soil after the construction of the dam (t_L = 49 years before sampling), the equivalent constant flux onto the SWI during this period, F_{eq}, can be estimated as:

$$\int_0^{t_L} F_{eq}\, e^{-\lambda t} dt = \Sigma_0^{z_L}, \qquad (4)$$

with λ being the radioactive decay constant for ^{210}Pb. This leads to F_{eq} =69 ± 5 Bq m^{-2} y^{-1}. Bulk depositional fluxes of ^{210}Pb in precipitation measured over a period of 16 months (April 2009–July 2010) in Huelva (Spain) ranged from 0.8 to 8.1 Bq m^{-2} month^{-1} (annual mean: 59 Bq m^{-2} y^{-1}), with the lowest depositional fluxes occurring during dry

summer months (Lozano et al., 2011a). This value compares well with the above F_{eq} despite the several meteorological factors influencing the ^{210}Pb concentrations in surface air from the southwestern Iberian Peninsula (Lozano et al., 2012; 2013).

The CF-CS model (Robbins, 1978) applied to the uppermost layers (0-15 cm interval, including 4 measured slices, with $p < 0.05$ for the exponential fit -continuous line in Fig. 4) leads to a SAR value of 0.050 ± 0.011 g cm^{-2}y^{-1}, in agreement with settling fluxes obtained from sediment traps. The CF-CS derived initial activity concentration is 107 ± 14 Bq kg^{-1}, which corresponds to a constant flux of 54 ± 14 Bq m^{-2} y^{-1}, being of the order of the estimated F_{eq}.

It is worth noting that the elapsed time of 49 years since the construction of the dam is not enough to allow a $^{210}Pb_{exc}$ steady-state inventory. The conditions for the applicability of the CF-CS model are met, but the typical exponential profile does not reach zero value theoretically ending as a step-function. This is illustrated in Figure 4 (nested panel) with two numerical solutions for the general advection-diffusion equation for a particle-bound tracer in accreting sediments (see Abril, 2003b):

$$\frac{\partial}{\partial t}(\rho A) = -\lambda \rho A + \frac{\partial}{\partial z}\left(D\rho \frac{\partial A}{\partial z}\right) - \frac{\partial}{\partial z}(wA) \qquad (5)$$

The symbols in Eq. 5 have been previously defined but (physical dimensions in square brackets) D [LT^{-2}], the diffusion coefficient, and w [ML^{-2}T^{-1}] denote SAR. The equation has been solved under the assumption of steady-state bulk density and null diffusion, rewritten in terms of mass depth, and using the MSOU high-order numerical scheme. The numerical solution 1 (Fig. 4) assumes a constant $^{210}Pb_{exc}$ flux of 54 Bq m^{-2} y^{-1} over 49 years, and a constant SAR of 0.05 g cm^{-2}y^{-1} (the new sediment is formed over a $^{210}Pb_{exc}$-free media). The numerical solution 2 (Fig. 4) is as the previous one, but now with SAR values monotonically decreasing with time from 0.15 g cm^{-2}y^{-1} after the construction of the dam till 0.05 g cm^{-2}y^{-1} at present.

The above analysis and numerical exercises suggest that it is possible to interpret the 0-25 cm of the core as the new sediment formed after the construction of the dam, and the $^{210}Pb_{exc}$ profile the likely result of moderate changes in SAR and fluxes during the elapsed time. Nevertheless, under the 25 cm horizon, $^{210}Pb_{exc}$ is reencountered at low concentrations and in scattered groups of sediment slices (Fig. 4). Its total inventory in the whole core length (accounting for those slices where it is higher than zero at 95% of confidence level and by using linear interpolation for the non-measured ones) is 3820 ± 230 Bq m^{-2}. Interpreting the whole $^{210}Pb_{exc}$ profile is an open question which will be readdressed after the new insights provided by the measured artificial radionuclides.

3.3. *Man-made radionuclides*

Figure 5 plots the measured activity concentration versus mass depth profiles for ^{137}Cs and $^{239+240}Pu$. Both show a well-defined peak at 65.5 ± 1 cm depth (m = 31.6 ± 0.8 g cm^{-2}) likely related to the maximum atmospheric fallout occurring in 1963.

The geographically closest historical records of ^{137}Cs atmospheric fallout are those from the UK-monitoring site in Gibraltar (south of the Iberian Peninsula). They comprise the period 1955-1985 (Wright et al., 1999). The extension of this dataset to the period 1950-2011 was performed as follows: i) for 1950-1953 we used the recorded fallout at Denmark as a proxy (Aarkrog et al., 1992) with an interpolate value from 1954 for connecting with data from Gibraltar; ii) for the period 1975-1985 the fallout series (Wright et al., 1999) reasonably fits an exponential function (R^2=0.97), which has been extrapolated to the period 1986 to 2011 as a simplifying approach. This last approach is justified by the following arguments: i) the impacts on the studied area of the nuclear accidents of Chernobyl (1886) and Fukushima (2011) were negligible (De

Cort et al., 1998; Lozano et al., 2011b; Piñero and Ferro, 2012; Vargas et al., 2016), and particularly the Chernobyl impact was not recorded in lacustrine (San Miguel et al., 2003) nor in estuarine systems of this area (Morales et al., 2008; San Miguel et al., 2004); ii) because of its low values, fallout in this period has minor effects in the total inventory and in the depth-profiles of activity concentrations in the core; iii) the series ends with values of ~ 1 Bq $m^{-2}y^{-1}$, which are of the order of the values reported in the scientific literature (e.g., 0.89 Bq $m^{-2}y^{-1}$ measured in Monaco for the period 1998-2010 - Pham et al., 2013). Results are shown in Figure ESM-2.

Figure ESM-2 allows the estimating of the integrated atmospheric deposition decay-corrected to the date of sampling, with results of 1.86 kBq m^{-2}. For the Spanish peninsular territory, Legarda et al. (2011) found a linear relationship between the integrated ^{137}Cs atmospheric deposition and the mean annual rainfall for the period 1950-1980 (rainfall is 582 ± 231 mm y^{-1} for the studied area), leading to (1.5 ± 0.3) kBq m^{-2} (uncertainty at one sigma level). This result is in reasonable agreement with the previous estimate.

Measured records of $^{239+240}$Pu atmospheric deposition are scarce. They can be estimated from those of ^{90}Sr using a constant scaling factor (Nakano and Povinec, 2003). For our present goals, the above ^{137}Cs fallout records can also support an approximate estimate. Hodge et al. (1996) reported a ^{137}Cs/$^{239+240}$Pu activity ratio of 38 ± 4 in soils samples from USA in the latitude band 38-41º and collected on 1 July 1994. This ratio is in agreement with the estimated 29.3 ± 2.2 from Greenland on January 2007 (Everett et al., 2008) and is close to the average value of 29.4 ± 3.5, obtained in soils from Mongolia in the Latitude range 45-48º in October 2007 (Hirose et al., 2017). Therefore, the global fallout value for $^{239+240}$Pu/^{137}Cs of 0.026 on 1 July of 1994 can be assumed (Wu et al., 2010). Using the above ratio and the decay-corrected integrated

^{137}Cs atmospheric deposition at Gibraltar at 1994, the resulting (assumed constant) scaling factor for $^{239+240}$Pu is 0.013. According to Hancock et al. (2011), the $^{239+240}$Pu/^{137}Cs activity ratio was of 0.018 before 1959 and 0.012 since then. Thus, this last and more refined estimate has been the one adopted in this work.

The absence of any source of Pu isotopes other than the atmospheric fallout can be inferred from the ^{240}Pu/^{239}Pu ratio (Lindahl et al., 2011). The ^{240}Pu/^{239}Pu atom ratios in this sediment core ranged from ~0.14 in the deepest layers to 0.20 in the sub-surface layers with an average of 0.17 (Table 1), which is similar to the global fallout ratio of 0.18 (Kelley et al., 1999).

The activity vs depth profiles in Figure 5 are not mirror images of the atmospheric deposition records (Fig. ESM-2). Thus, taking into account corrections by radioactive decay (referred to the date of sampling), the peak-level to present (SWI) ratio is ~170 for fallout records and only ~17 in the core profile. As a consequence, some post or pre depositional redistribution processes must be invoked. The System Time Averaged (STA) model by Robbins et al. (2000) attempts to account for the latest. It is an updated and simpler formulation of the existing knowledge on the integration of atmospheric fluxes (Abril and García-León, 1994; McCall et al; 1984). The fluxes onto the SWI, F_S, are expressed as a function of the atmospheric deposition, F_a, involving the radioactive decay constant of the radionuclide, λ_R, and a system's constant, k_r, with physical dimension of T^{-1}:

$$\frac{dF_S}{dt} = k_r F_a - (\lambda_R + k_r) F_S \quad (6)$$

The aquatic sediments with watershed-dominated inputs of radionuclides (and matter) are the main target systems for the STA model, although it may be more general (Robbins et al., 2000). Intuitively, atmospheric fluxes are accumulated (integrated) in the catchment area from where they are transferred towards the sediment at a constant

rate k_r (for a long-lived radionuclide, $1/k_r$ is the residence time required for reducing the amount of any individual atmospheric input by a factor e). For large-surface catchments the fluxes onto the sediment (and the resulting inventory) are larger than the atmospheric deposition (and its integrated value), both being decay-corrected to the date of sampling. This is solved in the STA model by means of a normalisation factor, Z, defined as the ratio between the inventory and the integrated atmospheric deposition. The simplest version of the model assumes a constant (mean) value of SAR, which can be first-estimated from the peak position and then further refined (the position of the peak is only slightly displaced by the STA model). This allows the converting of the sequence of F_S into activity concentrations and ages into mass depths. Thus, STA only involves a free parameter, namely k_r, which is selected to provide the best agreement between the measured and the modelled profiles.

Results from the STA model are shown in Figure 5 for ^{137}Cs and $^{239+240}$Pu. For both cases, SAR is 0.68 g cm^{-2}y^{-1}. The ^{137}Cs inventory in the core is 8170 ± 35 Bq m^{-2}, a factor Z_{Cs}=4.6 higher than the estimated integrated atmospheric deposition at the site. For $^{239+240}$Pu, the inventory is 256.2 ± 1.4 Bq m^{-2}, and Z_{Pu}=3.6. Finally, k_r is 0.065 y^{-1} for ^{137}Cs and 0.095 y^{-1} for $^{239+240}$Pu. These values are comparable to those for ^{137}Cs reported by Abril and García-León (1994) of k_r =0.043 y^{-1}, Z_{Cs}=2.8 (Lake Krageholmsjön, Sweden) and k_r = 0.010 y^{-1}, Z_{Cs}=4.5 (the Palace Moat in Tokyo), and to the value of k_r =0.062±0.004 y^{-1} found by Robbins et al. (2000) for ^{137}Cs and $^{239+240}$Pu in sediments from Florida Bay.

Concerning the $^{239+240}$Pu/^{137}Cs ratio for the total inventory, a value of 0.031 was found in this work, which compares well with the ratios found in Doñana Park (SW Spain), in the narrow range 0.027-0.036 (Gascó et al., 2006).

The STA model seems to be able to capture the most basic features of the activity profiles: the position, size, and roughly the wide of the peaks, as well as the trend of decrease of activity towards the SWI. Activity concentrations are found deeper than expected from the STA model, which has often been attributed to diffusion processes (Crusius and Anderson, 1995) or to non-ideal deposition (Abril and Gharbi, 2012). Nevertheless, there are two major concerns: i) if the peak is dated 1963 then most of the core layers below this peak are not the result of the accretion of an aquatic sediment, however, they are the original soil; ii) $^{210}Pb_{exc}$ is absent around the peak area and its profile is not consistent with the interpretation of the pre-peak portion of the core as an aquatic sediment accreting with a constant SAR. Furthermore, this mean SAR value is not consistent with the settling fluxes measured by the sediment traps.

The activity ratio $^{239+240}Pu/^{137}Cs$, estimated on a layer-by-layer basis from data in Table 1 (as shown in Fig. ESM-3) provides additional insight. The ratio takes values of up to 0.04 around the peak, being almost uniform in the upper layers, with values ~0.024 except at the SWI where the ratio increases up to 0.042. These ratios are lower than the ones of 0.048-0.062 reported for soils in the latitude range 34.6º-38.9º N in China (Xu et al., 2017). A high range of spatial variability for the $^{239+240}Pu/^{137}Cs$ activity ratios (0.012-0.255) has been found in sediment samples from the southeastern Brazilian shelf–SW Atlantic margin (Figueira et al., 2006).

The activity ratios in the core from Sancho Reservoir can be compared against those expected from the annual atmospheric deposition (Fig. ESM-2) and the STA fluxes onto the SWI. At each particular date the ratio is captured and then corrected by radioactive decay to the date of sampling, allowing the comparison shown in Figure ESM-3. The ratios based upon the annual deposition consistently reflect the scaling factors used for building the $^{239+240}Pu$ deposition records from the ones for ^{137}Cs and

radioactive decay. They decline towards the SWI; however, this trend is not observed in the measured values. Because of the different residence times for both radionuclides (see k_r values above), the decline on STA-based ratios is smoothed (it is worth noting that k_r =0 would produce a constant value of 0.039 for this ratio). Nevertheless, the STA-based ratios are too high around the ^{137}Cs peak position. This suggests that in the core there are additional amounts of ^{137}Cs with respect to $^{239+240}$Pu. This is qualitatively consistent with the above ratio Z_{Cs}/Z_{Pu} ~1.3, and it may be linked to a higher ^{137}Cs supply from the watershed associated to the dissolved phase, particularly around the time of maximum deposition.

Another major feature in Figure ESM-3 is that none of the flux-based ratios predict the observed decline below the ^{137}Cs peak position, which is likely linked to a higher mobility of ^{137}Cs in the porous media (either in terms of diffusion or of non-ideal deposition). Finally, the unexpected increase of the $^{239+240}$Pu/^{137}Cs activity ratio near the SWI may reflect the effects of AMD on the k_d distribution coefficient, reducing the one for ^{137}Cs in a proportion larger than that for $^{239+240}$Pu (Abril and Fraga, 1996).

3.4. Depositional events and a piecewise chronology

Figure 6 summarises the bulk of the dataset and allows the distinguishing of up to six regions based upon correlated features of bulk density and $^{210}Pb_{exc}$. A key point for understanding data is the distinct behaviour of $^{210}Pb_{exc}$ in soils and aquatic sediments (unsaturated and saturated porous media, respectively), and its functioning in flood plains (Aalto and Nittrouer, 2012; Mabit et al., 2014).

Unperturbed soils exhale ^{222}Rn through their connected porous, and they receive $^{210}Pb_{exc}$ fallout which penetrates (roughly exponentially) in the uppermost layers. As a

result, a slight excess of ^{210}Pb in the upper most layers and a deficit in the inner ones (within the diffusion zone) can be expected. The values of $^{210}Pb_{exc}$ are not usually too high at about 15-25 Bq/kg (Benmansour et al., 2013; Gaspar et al., 2013).

In saturated sediments ^{222}Rn exhalation is inhibited and the media becomes a net accumulator of $^{210}Pb_{exc}$, with fluxes onto the SWI usually higher than atmospheric deposition and with a null or limited capacity of being distributed in depth (unless in some sediments with high porosities, as shown by Abril and Gharbi, 2012).

Floodplain soils develop from episodic supplies of materials forming more or less homogenous layers which, depending on their origin, can host significant concentrations of radionuclides (Aalto and Nittrouer, 2012). During normal (non-episodic) conditions $^{210}Pb_{exc}$ can increase in the uppermost layers, with an exponential decay distribution in depth like in a soil.

These three systems (i.e. saturated sediments, unperturbed and floodplain soils) are net accumulators of ^{137}Cs and $^{239+240}Pu$. In soils these artificial radionuclides are distributed at depth (up to few tens of cm – Bunzl et al., 1995) with typical profiles, which often are described in terms of convection-diffusion models (Szabó et al., 2012).

The exact details of the construction of the dam and the subsequent filling of the reservoir are not well documented, but the reported end of works was on December 31st 1962, very close to the maximum in ^{137}Cs atmospheric deposition (likely in early 1963). Thus, the observed peaks of ^{137}Cs and $^{239+240}Pu$ at 65.5 ±1 cm depth give a good proxy for the location of the soil surface at such a date. Consequently, most of the R6 region in Figure 6 must correspond to the former substrate before the filling of the dam.

This R6 region can be split into two different sub-regions. Within sub-region R6a, $^{210}Pb_{exc}$ is absent as expected from an unsaturated soil. It is worth noting that after 49 years any activity of $^{210}Pb_{exc}$ at the former surface would have decayed to 22% of its

553 initial value, so with the involved uncertainties it would not be detected if initially it
554 was lower than ~ 20-25 Bq kg^{-1}. But the 64.5-70.5 cm soil layer seems to lay over a
555 material which contains measurable amounts of $^{210}Pb_{exc.}$ The ^{137}Cs inventory obtained
556 from the bottom of the core to the ^{137}Cs peak layer is 3120 ± 20 Bq m^{-2}, noticeably
557 higher than the expected integrated atmospheric deposition during this time interval (~
558 1230 Bq m^{-2}, as estimated from data in Figure ESM-2 after corrections by radioactive
559 decay referred to the date of sampling). For $^{239+240}Pu$, the inventory is 122.4 ± 1.2 Bq m^{-2}
560 , and the integrated atmospheric deposition is ~ 53 Bq m^{-2}. This suggests a scenario
561 comparable to a flood plain in which the R6a layer was likely formed in a depositional
562 event over the R6b. To make a significant contribution to the inventories of artificial
563 radionuclides in R6b possible, the depositional event R6a should not have occurred
564 much earlier than 1962.

565 The historical records of annual rainfall in the studied area for the period 1960-
566 1989 (Fig. 7) show a very large variability, with a minimum value of 314 mm in 1974
567 and a maximum of 1229 mm in 1962. As mentioned above, 60% of rainfall occurs
568 between October and January and is characterised by its episodic character, with some
569 events of about 100 mm/day.

570 Region R6b can then be interpreted as the original soil or floodplain being likely
571 puddled with the on-course dam building works, and allowing the increase of $^{210}Pb_{exc}$
572 from the fluxes of mass and activity onto the SWI. The transition to region R6a is likely
573 related to one (or a series of) depositional event(s) associated with the extraordinary
574 high rainfall occurring in late 1962. The provenance of this mass flow would have been
575 the upslope unsaturated (and almost $^{210}Pb_{exc}$ free) soils. In terms of chronology this
576 would have happened almost simultaneously with the maximum fallout of artificial
577 radionuclides and the first fill up of the reservoir.

578 Regions R2 to R5 in Figure 6 may represent a sequence of depositional events
579 and periods of continuous sedimentation following the construction of the dam and the
580 first filling of the reservoir. Accounting for the episodic character of rainfall in the
581 region, the loading would have taken place with a sequence of floods likely
582 remobilising and transporting materials from the inundated regions and settling them in
583 the deepest areas around the dam. The process would have been repeated with
584 decreasing intensity in the (annual) cycles of loading and discharge of the reservoir.
585 After 1972, with the increase in height of the dam, the main depositional area of
586 sediments transported by the Meca River would have been noticeably displaced
587 upstream and the sedimentary conditions at the core site would have tended towards a
588 steady state.

589 The exact dating for these depositional events can hardly be inferred from the
590 $^{210}Pb_{exc}$ data, despite the above semi-quantitative arguments. Nevertheless, a tentative
591 correlation among relative maxima in rainfall (Fig. 7) and the depositional events
592 identified in Figure 6 can be established. This would provide a series of reference dates:

593 i) The transition from region R6b (71.5-76.5 cm) to R6a (64.5-71.5 cm) (denoted
594 as T6b in Fig. 6), can be ascribed to (late) 1962 as discussed above.

595 ii) The transition from region R6a to R5, denoted as T6a in Figure 6, would
596 correspond to (early) 1963, at the time of maximum atmospheric fallout of artificial
597 radionuclides.

598 iii) The deepest portion of region R5 (R5b, 58.5-64.5 cm) is $^{210}Pb_{exc}$ free, with a
599 marked downwards gradient in bulk density, and it can be interpreted as one (or a series
600 of) depositional event(s) following the first fill up of the reservoir under the also very
601 high rainfall of 1963 (Fig. 7), and with likely provenance in the unsaturated upslope
602 soils. This process should have finished with the current hydrological year (late 1963 or

603 early 1964, time mark T5b in Fig. 6) since global rainfall during 1964 was lower than
604 the average (Fig. 7).

605 iv) The upper portion of region R5 (R5a, 52.5-58.5 cm) has relatively uniform
606 values of bulk density and measurable amounts of $^{210}Pb_{exc}$, which increase upwards.
607 This can be interpreted as sediments accreting under relatively smoother environmental
608 conditions (likely during 1964) until the next large depositional event (region R4),
609 likely linked to the high rainfall of 1965 (Fig. 7). Thus, the transition T5a (Fig. 6) can be
610 ascribed to (late) 1965. The mass thickness of R5a is 3.78 g cm^{-2}, which, if accumulated
611 during ~2 years, allows the estimating of a mean SAR of ~1.9 g $cm^{-2}y^{-1}$.

612 v) The (single or multiple) depositional event of R4 (46.5-52-5 cm) shows the
613 same characteristics as R6a and the deepest portion of R5, and is likely dated (T4~T5a)
614 in (late) 1965. The overlaying region, R3 (32.5-46.5 cm) is comparable to the upper
615 layers of R5, and seems to have been formed under the smoother environmental
616 conditions likely prevailing up to T3 (Fig. 6), linked to the next noticeable peak in
617 rainfall at 1968-1969 (Fig. 7). Thus, the 7.87 g cm^{-2} of R3, if accumulated during ~3
618 years, gives an estimate of ~2.6 g $cm^{-2}y^{-1}$ for mean SAR in the period.

619 vi) Region R2 (26.5-32.5 cm) is the last large depositional event, similar to R4
620 which can be tentatively linked to the high rainfall in 1969 (Fig. 7); and thus, T2~T3
621 (Fig. 6).

622 vii) The next peak in rainfall occurs in 1972 (Fig. 7) after the increase in height
623 of the dam, what is supposed to have decreased the supply of matter in the region where
624 the core was sampled. The conditions for high depositional events would not have been
625 met afterwards. Thus, region R1 (0-26.5 cm) is characterised by a smooth change in the
626 concentration of crustal radionuclides (denoting a change in provenance – Fig. 3), a

627 monotonic upwards increase in $^{210}Pb_{exc}$ (Fig. 4) and the development of the fingerprint
628 of early compaction in bulk density (Fig. 2).

629 Adopting T2 (1969) as a suitable time mark, the CRS model with this reference
630 date (Appleby, 1998) can be applied to the R1 region of the $^{210}Pb_{exc}$ profile (by using
631 linear interpolations for the non-measured sediment slices). This, along with the above
632 set of time marks provides the tentative chronology for the core shown in Figure 8. The
633 large uncertainties do not allow a proper analysis of SARs, but the general trend is a
634 decrease towards the SWI, from values around 0.7 g cm^{-2} y^{-1} at the base of region R1 to
635 values of ~0.08 at the SWI. The last is in reasonable agreement with the recent
636 measurements of settling fluxes and the trend of decreasing SARs is also consistent with
637 the numerical simulations (scenario number 2) shown in Figure 4 (subsection 3.2).

638 It is worth noting that the CRS model applies to region R1 and its choice is to
639 some extent arbitrary. A chronology can be seen as the plot of a continuous monotonic
640 function in the age versus mass-depth (or true depth) space, starting from the origin of
641 coordinates. There are an infinite number of chronological lines being mathematically
642 exact solutions for any given $^{210}Pb_{exc}$ profile in absence of restrictive assumptions
643 (Abril, 2015). When using time marks (points in such space) we are forcing the
644 chronological line to match such points, but there are still an infinite number of possible
645 mathematically exact solutions. Selecting a CRS model with a reference date for
646 connecting the origin of coordinates with the time-mark is only an option, and the
647 resulting chronology has to be considered as tentative.

648 To better illuminate this point, we applied the methodology by Arnaud et al.
649 (2002) consisting in truncating the $^{210}Pb_{exc}$ profile by removing those regions identified
650 as depositional events (in this case, regions R2, R4, R5b and R6a in Fig. 6). Then we
651 applied the CRS model with the reference date T5b (Fig. 6 and discussion above). The

652 resulting chronology is depicted in Figure 8. The method may be sensitive to the
653 accurate determination of the regions of depositional events, but the present modelling
654 exercise shows a chronology which substantially differs from the previous estimates.
655 SAR values are overall larger, around 0.12 g cm^{-2} y^{-1} at SWI (higher, but still in the
656 order of the measured settling fluxes), with a clear pattern of deceleration since 1994
657 when they peaked at 0.6 g cm^{-2} y^{-1}. The corresponding CRS dates for the depositional
658 events R2 and R4 are 1992 and 1971, respectively. While the latest could be related to
659 the new works on the dam, there are no clear reasons for the former (R2) or for the peak
660 in SAR. It is worth noting that the period 1988-1992 was of drought, with rainfall in
661 1993-1995 still under the average and with extraordinary high annual rainfall in 1996
662 (1207 mm).

663 Application of the CIC model (see model description in Sánchez-Cabeza and
664 Ruíz-Fernández, 2012) leads to a non-monotonic chronology, with unacceptable ages of
665 about one century at the depth of the ^{137}Cs peak (results not shown).

666 The major difficulties in the above CRS and CIC chronologies are their
667 respective basic assumptions (a constant rate of supply or a constant initial activity
668 concentration). It has been shown that $^{210}Pb_{exc}$ fluxes correlate with SAR in most of the
669 cases (Abril and Brunskill, 2014), and particularly in those with watershed dominated
670 inputs (McCall et al., 1984). The above correlation arises from the independent
671 variability of SAR and initial activity concentrations. This may seriously limit the
672 applicability of CIC and CRS models.

673 McCall et al. (1984) found that the ^{210}Pb models based upon a constant initial
674 concentration were more reliable for describing sediments in reservoirs. Nevertheless, a
675 natural variability around their mean values has been found for initial concentrations in
676 a large variety of aquatic ecosystems (Abril and Brunskill, 2014). When randomly

677 distributed in time, the effect of such variability in the CIC model is that the CIC-ages
678 fluctuate around the trend-line which provides the meaningful chronology. When
679 permanent changes in environmental conditions shift the initial concentrations towards
680 a different mean value, a piecewise CIC model can then be applied, as shown in Figure
681 8. For region R1 (Fig. 6), the one with a distinct fingerprint in crustal radionuclides
682 (Fig. 3), the value 105 Bq/kg was used as (mean) initial concentration, while for the
683 deepest layers (in the truncated core), a value of 55 Bq/kg was adopted. The so derived
684 CIC ages fluctuate around a trend-line, but they are in reasonable agreement with the
685 reference dates inferred from the rainfall records (Fig. 8).

686 A gradual change from both values of initial activity concentration would be
687 more reliable, but the above modelling exercise is enough for providing a basic insight.
688 The upstream migration of the main depositional area after 1972 prevents the supply
689 onto the SWI of large particle sizes, which in turn results (see Abril and Fraga, 1996) in
690 an increase in the activity concentration for the surface-bound tracers ($^{210}Pb_{exc}$) and a
691 decrease in the matrix-associated and relatively soluble species (such as ^{226}Ra and
692 ^{228}Ra).

693 Although in this particular case radionuclides cannot provide a conclusive
694 accurate chronology for the sediment core, they can still provide valuable insight on the
695 functioning of the studied sedimentary system, as it follows from the present work.

696

697 **4. Summary and Conclusions**

698

699 A sediment core was sampled in 2011 from Sancho Reservoir (SW Spain) and analysed
700 for bulk density, ^{137}Cs, ^{239}Pu, ^{240}Pu, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{234}Th (^{238}U) and ^{40}K.

Bulk density revealed unsteady compaction and likely depositional events (Fig. 2).

The activity concentrations of ^{226}Ra, ^{228}Ra, ^{234}Th (^{238}U) and ^{40}K were uniform along the core, but declining overall in the upper 0-25 cm. This reveals changes in provenance with the exception of ^{238}U, which increases in the upper 10 cm likely due to its supply by AMD (Fig. 3).

The AMD fingerprint is also found in the $^{239+240}$Pu/^{137}Cs activity ratio which increases in the uppermost sediment layers, while the radioactive flux based ratios predicts a decline.

The ^{137}Cs and $^{239+240}$Pu profiles show well defined peaks at the same depth, with inventories being about four times higher than the expected integrated atmospheric deposition in the area. The STA model is able to capture some basic features of these profiles, but the whole dataset cannot be interpreted in terms of continuous sedimentation processes. Particularly, the unsupported ^{210}Pb (^{210}Pb$_{exc}$) showed a complex non-monotonic profile, interrupted at several sections particularly around the ^{137}Cs peak.

The upper 0-25 cm of the core shows a monotonic decline in ^{210}Pb$_{exc}$ with depth, which can be interpreted in terms of the CF-CS model, as well as with the numerical solutions of the continuity equation for a particle-associated tracer. The corresponding SAR values for recent dates are ~0.05 g cm^{-2}y^{-1}, in good agreement with the reported settling fluxes at the sampling site.

Based upon correlated features in the bulk density and ^{210}Pb$_{exc}$ profiles, a series of depositional events (likely linked to peaks in the rainfall records) have been identified in the core. They followed the construction of the dam until its increase in

height (1972), which likely displaced upstream the main depositional area of riverine loads, as inferred from sediment traps.

The CRS model with a reference date has been used for complementing and discussing the chronology, but its basic assumption of a constant rate of supply may not be attained in this scenario, so the so obtained chronology has to be considered as tentative.

A piecewise CIC model can be applied to the truncated core (i.e., after excluding the regions associated to episodic depositional events), which leads to a chronology consistent with the reference dates inferred from the rainfall records and with the above radiogeochronological fingerprints.

Although in this particular case radionuclides cannot provide a conclusive accurate chronology for the sediment core, they can still provide valuable insight on the functioning of the studied sedimentary system.

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FIGURE CAPTIONS

Figure 1. The sampling site: Sancho Reservoir in the Meca River (SW Spain). The core was sampled in 2011 close to the dam. Symbol ST refers to the sediment traps deployed in the work by Torres et al. (2013). Image from the Spanish Instituto Geográfico Nacional (IGN, http://www.ign.es).

Figure 2. Measured bulk density versus true depth in the sediment core from the Sancho Reservoir (Fig. 1). The dotted line plots the typical profile under steady-state early compaction (Abril, 2011): $\rho = \rho_\infty - \rho_1 e^{-\alpha z}$ (with parameter values ρ_∞ =0.76 g cm^{-3}, ρ_1=0.90 g cm^{-3} and α=0.04 cm^{-1}, corresponding to the best least-squared fit with R^2 = 0.81). The continuous line is a second order polynomial fit for the 0-30 cm region.

Figure 3. Normalised (to their mean values in the 56-75 cm interval) activity concentrations of ^{226}Ra, ^{228}Ra, ^{40}K and ^{234}Th versus depth in the sediment core. Error bars correspond to the 1σ counting uncertainties.

Figure 4. Unsupported $^{210}Pb_{exc}$ versus mass depth, estimated by subtracting the ^{226}Ra activity from the total ^{210}Pb activity (Table 1) on a layer-by-layer basis. Error bars correspond to 1σ propagated uncertainties, while horizontal bars depict the mass depth interval for the measured sediment slices. The subpanel shows a zoon for the 0-7 cm upper region, where $^{210}Pb_{exc}$ data is compared against the CF-CS model and two numerical solutions of the governing equation for the mass conservation for a particle-bound tracer in accreting sediments (see text for details).

Figure 5. Measured activity concentrations versus mass depth for ^{137}Cs and $^{239+240}Pu$ in the core from the Sancho Reservoir. The continuous and dotted lines correspond to the interpretation of these profiles in terms of the system time averaged (STA) model for radioactive inputs onto the SWI with a constant (mean) SAR value. See text for model description and parameter values.

Figure 6. Bulk density and activity concentrations of $^{210}Pb_{exc}$, ^{137}Cs and ^{228}Ra versus actual depths in the sediment core from Sancho Reservoir. Vertical dashed lines define time marks (T_i) and sub-regions (R_i) in the core based upon the distinct behaviour of $^{210}Pb_{exc}$ and bulk density.

Figure 7. Annual rainfall records in the studied area. Data from AEMET (Spanish Meteorological Agency). Vertical dashed lines indicate the date of construction of the dam (1962) and the works for increasing its height (1972).

Figure 8. Tentative chronology for the core from the Sancho Reservoir. Dots are time marks based upon ^{137}Cs and $^{239+240}Pu$ peaks, and the rainfall records (Fig. 7) along with the depositional events (Fig. 6, see text for details). Also plotted is the chronology from the CRS model with the reference date T2 applied to region R1 (Fig. 6) and to the truncated core (by excluding the depositional events according with the methodology by Arnaud et al., 2002) with the reference date T5b (Fig. 6). A piecewise CIC model is also applied to the truncated core, with initial concentration of 105 Bq/kg in region R1 and 55 Bq/kg for the rest, with 10% relative uncertainty.

ELECTRONIC SUPLEMENTARY MATERIAL

Figure ESM-1. Historical aerial photograph of the sampling site with the approximate location of the studied core before the construction of the dam. Image from the Spanish Instituto Geográfico Nacional (IGN; https://fototeca.cnig.es/), photogrammetric flight: 1956-1957.

Figure ESM-2. ^{137}Cs atmospheric fallout recorded at Gibraltar (south of Iberian Peninsula) for the period 1955-1985 (Wright et al., 1999), extended to 1950 (based upon recorded data in Denmark; Aarkrog et al., 1992) and to 2011 (by extrapolating the exponential declining observed for the period 1975-1985).

Figure ESM-3. Measured (dots) $^{239+240}Pu/^{137}Cs$ activity ratios vs depth in the sediment core from Sancho Reservoir along with those estimated from the annual atmospheric deposition (continuous line) and the STA-derived fluxes onto the SWI (open circles), both decay corrected to the date of sampling.