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Fallout ^{210}Pb as a soil and sediment tracer in catchment sediment budget investigations: A review

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A B S T R A C T

Increasing anthropogenic pressures coupled with climate change impacts on natural resources have promoted a quest for innovative tracing techniques for understanding soil redistribution processes and assessing the environmental status of soil resources. Among the different existing tracers, the fallout component of the radioisotope lead-210, also termed unsupported or excess lead-210 ($^{210}Pb_{ex}$) when referring to its presence in soil or sediment, arguably offers the broadest potential for environmental applications, due to its origin and relatively long half-life. For more than five decades, $^{210}Pb_{ex}$ has been widely used for dating sediments, to investigate sedimentation processes and, since the 1990s, to provide information on the magnitude of soil and sediment redistribution.

The aim of this review is to provide a comprehensive evaluation and discussion of the various applications of $^{210}Pb_{ex}$ as a tracer in terrestrial and aquatic environments, with particular emphasis on catchment sediment budget investigations. This paper summarizes the state-of-the-art related to the use of this tracer, the main assumptions, the requirements (including the need for accurate analytical measurements and for parallel validation), and the limitations which must be recognised when using this fallout radionuclide as a soil and sediment tracer. Lessons learned and current and future research needs in the environmental and radiochronological application of $^{210}Pb_{ex}$ are also presented and discussed.

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1. Introduction

Lead-210 (^{210}Pb) is a valuable radioactive isotope for environmental investigations and it has been widely used in the natural and physical sciences for dating, tracing, and modelling biogeochemical cycling in ecosystems. Since the 1970s, ^{210}Pb measurements have been used extensively for dating sediment deposits in a range of sedimentary environments, including lakes, reservoirs, flood plains, wetlands, estuaries, and coastal marine environments, thereby permitting sedimentation rates over the last 100–150 years to be determined (e.g. Appleby and Oldfield, 1978; Robbins, 1978; Wise, 1980; Benninger and Krishnaswami, 1981; Oldfield and Appleby, 1984; Robbins et al., 1990; Abril et al., 1992; Walling and He, 1994; He and Walling, 1996a; Walling and He, 1999a; Wallbrink et al., 2002a; Benmansour et al., 2006; Laissaoui et al., 2008; Simms et al., 2008; Navas et al., 2011).

While ^{210}Pb was also used concurrently as an atmospheric and aerosol tracer (Burton and Stewart, 1960), it is only since the mid-1990s that studies have used ^{210}Pb for assessing rates and patterns of soil redistribution and for tracing sediment movement in terrestrial and aquatic environments (e.g. He and Owens, 1995; Walling et al., 1995; Wallbrink and Murray, 1996; He and Walling, 1997; Walling and He, 1999b; Walling et al., 2003a,b; Owens et al., 2012). Although several review papers and books have been published in recent years dealing with the environmental impact, distribution and behaviour of lead (e.g. Selim, 2011, 2012) and with the use of its stable (e.g. ^{206}Pb/^{207}Pb ratio) and radioactive isotopes as geochemical tracers to investigate pollution pathways and anthropogenic impacts in different environmental systems (e.g. Robbins, 1978; Renberg et al., 2002; Komarek et al., 2008; Cheng and Hu, 2010; Bird, 2011), reviews focussing on ^{210}Pb are scarce. Existing overviews have focused on the radioecology and environmental behaviour of ^{210}Pb (e.g. Persson and Holm, 2011), on its atmospheric applications, on its method of use for dating recent biogenic and inorganic carbonates (see Baskaran, 2011a,b), and on its use as a sediment geochronometer (see Appleby, 2008). Other reviews have focused on other fallout radionuclide (FRN) soil tracers or such tracers more generally (e.g. Zapata, 2002; Mabit et al., 2008; Kaste and Baskaran, 2011; Matisoff and Whiting, 2011; Walling, 2012; Taylor et al., 2013; Walling, 2013), but to date there is little available literature focusing in depth on ^{210}Pb and its terrestrial environmental applications (i.e. as a soil and sediment tracer).

The focus of this review is therefore on the use of ^{210}Pb as a tracer of soil and sediment in the terrestrial environment, including both the land surface and surface waters. Marine and coastal environments are explicitly excluded and the reader is referred to DeMaster et al. (1985), Du et al. (2011), Nozaki (1977) and Santos et al. (2008) and other publications for information on these environments.

In this contribution, current understanding of the environmental behaviour of ^{210}Pb and the basis of its use as a tracer will be summarised, followed by a critical review of the assumptions involved and the advantages, as well as the limitations, associated with its use as a soil and sediment tracer and as a sediment geochronometer. Because the measurement facilities and the technical expertise required for its effective use as an environmental tracer can provide major challenges, this topic will also be covered and will be followed by a review of recent advances and future research needs.

2. Origin and characteristics

With an atomic number of 82, lead (Pb) belongs to the heavy metals group and has four natural stable isotopes: ^{206}Pb, ^{207}Pb, ^{208}Pb and ^{204}Pb, with natural abundances of 23.6%, 22.6%, 52.3% and 1.4%, respectively. ^{210}Pb is a natural geogenic radioisotope occurring as one of the decay products of the ^{238}U series (i.e. – ^{226}Ra–^{222}Rn–^{218}Po–^{214}Pb–^{214}Bi–^{214}Po–^{210}Pb). Due to its relatively short half-life ($t_{1/2} = 22.23$ years), ^{210}Pb is the only radioisotope of Pb that offers potential for applied environmental investigations.

Theoretically, ^{226}Ra and ^{210}Pb should be in secular equilibrium but owing to the diffusional behaviour of the noble gas daughter ^{222}Rn, disequilibrium can occur in natural materials. A fraction of ^{222}Rn ($t_{1/2} =$ 3.8 days) diffuses from geological and soil materials into the overlying atmosphere or upper soil layers, leading to disequilibrium between the parent and daughter isotopes. When ^{222}Rn escapes into the air its daughter products (including ^{210}Pb) are attached to the surfaces of aerosols and dust particles. These daughter isotopes reach the soil surface by wet and dry fallout and accumulate in the surface layers of the soil (see Fig. 1). As a result of this process, the surface soil layer contains a higher ^{210}Pb concentration than that expected from equilibrium with ^{226}Ra. The part of the ^{210}Pb loading, which is in equilibrium with the ^{226}Ra activity, is called 'supported ^{210}Pb'. Its activity will decrease with the half-life of ^{226}Ra (1600 years). The ^{210}Pb in excess of the equilibrium concentration is termed 'unsupported' or 'excess' ($^{210}Pb_{ex}$). $^{210}Pb_{ex}$ decays by the physical half-life of ^{210}Pb. It is this 'excess' component, often also referred to as fallout ^{210}Pb because of its input to the land surface as fallout, that provides the basis of most tracer applications.

^{210}Pb is a beta decay radionuclide and 19.8% of its decay occurs at the ground level of the ^{210}Bi isotope. However 80.2% results in an excited nucleus which stabilises by emission of low energy gamma rays (46.53 keV), and internal conversion of electrons to the ground state of ^{210}Bi ($t_{1/2} = 5.012$ days) which in turn decays by emission of beta particles to a pure alpha emitter ^{210}Po ($t_{1/2} = 138.37$ days) with an energy of 5.407 MeV (Laboratoire National Henri Becquerel, 2012).

The ^{210}Pb fallout flux is essentially continuous through time because of its natural origin. However, weekly, monthly, seasonal and longer-term variations of the ^{210}Pb concentration in the air and in atmospheric fallout (rain, snow and dry deposition) have been documented by long-term measurements (e.g. Baskaran et al., 1993; Preiss et al., 1996; Winkler and Rosner, 2000). The global pattern of ^{210}Pb fallout is characterized by a high spatial variability due to the predominant west to east movement of air masses. This commonly results in low ^{210}Pb fallout in the western areas of the continents, where the air masses have travelled over the oceans and have had little opportunity to collect ^{210}Pb, and much higher ^{210}Pb fallout in the eastern areas of continents, where the air masses will have passed over the continental interiors.

At the global scale, the annual fallout flux of ^{210}Pb is reported to vary from 0.1 Bq m^{-2} yr^{-1} to 360 Bq m^{-2} yr^{-1} (Turekian et al., 1977; Robbins, 1978; Appleby and Oldfield, 1992; Preiss and Genthon, 1997; Baskaran, 2011a) with higher values (i) over continental areas than

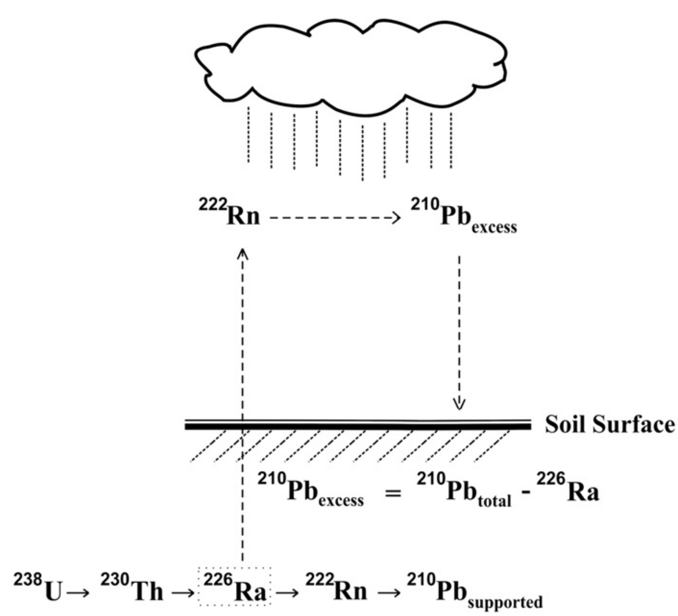

Fig. 1. Origin of geogenic ^{210}Pb and fallout ^{210}Pb (i.e. $^{210}Pb_{ex}$).

over oceanic or polar areas and (ii) in the Northern rather than in the Southern Hemisphere, due to the higher land to sea ratio.

Significant correlations have been observed between mean annual rainfall and/or number of heavy rain events/thunderstorms and the mean annual deposition flux of ^{210}Pb (e.g. Baskaran, 1995; Beks et al., 1998; Winkler and Rosner, 2000; Caillet et al., 2001; Baskaran, 2011a), suggesting that wet precipitation is mainly responsible for the scavenging of this fallout radionuclide from the atmosphere. In south Germany, for example, the mean annual ^{210}Pb deposition flux for the period 1981–1999 was 180 Bq m^{-2} yr^{-1} for a mean annual precipitation of 855 mm (Winkler and Rosner, 2000). The minimum and maximum values for the same location were 120 and 250 Bq m^{-2} yr^{-1}. In Hungary (Budapest), the mean annual ^{210}Pb deposition flux for the period 1999–2006 was 81 Bq m^{-2} yr^{-1} for a mean annual precipitation of 476 mm, with minimum and maximum values for the same location of 44 and 100 Bq m^{-2} yr^{-1}, respectively (Central Radioanalytical Laboratory of Food and Feed Safety Directorate, 2006).

Seasonal variations of ^{210}Pb concentrations in the atmosphere have been reported in many parts of the world. For example, in Budapest (Hungary), the temporal variation of the ^{210}Pb concentration (mBq m^{-3}) in the air monitored by the Central Radioanalytical Laboratory of Food and Feed Safety Directorate (2006) shows that in the autumn and winter seasons, the mean activity concentrations of ^{210}Pb were slightly higher than those of the spring and summer seasons (Fig. 2). Similar findings concerning long-term variation of ^{210}Pb concentrations in air have been reported from Egypt (Ahmed et al., 2004) and from south Germany (Winkler and Rosner, 2000). This seasonal variability can be related to the higher temperatures in spring and summer, which are usually accompanied by higher turbulence.

Taking into account the seasonal variability and the time-scales involved in the studies of soil erosion and in the radiometric dating of recent sediments, meaningful definitions of fluxes and mass accumulation rates require averaging over time intervals of the order of one to several years. For most of the applications, the assumption of constant fluxes therefore remains useful.

3. Environmental behaviour of ^{210}Pb

One major requirement for using an element as a potential tracer of soil redistribution is to confirm its reduced mobility in the topsoil layer and that it is predominantly moved by soil/sediment mobilisation and transport processes (see Guzmán et al., 2013; Koiter et al., 2013). The key feature of Pb behaviour in the soil, on which tracer applications are founded, is its strong binding to soil particles (both mineral particles

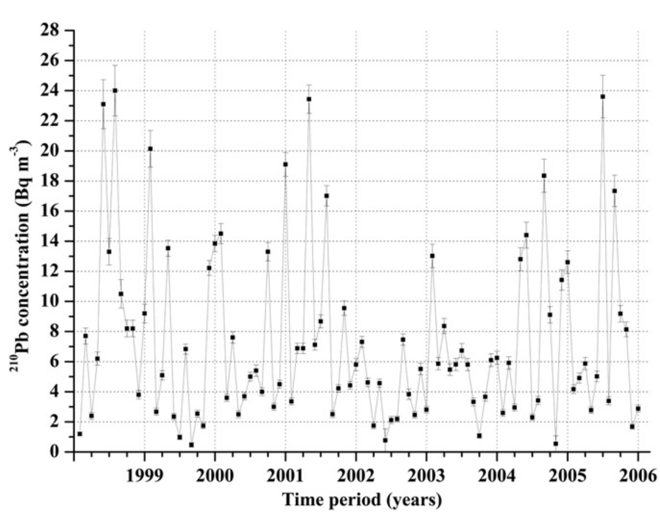

Fig. 2. Variation of ^{210}Pb activity concentrations in air (mBq m^{-3}) measured in Hungary. Data presented as monthly mean ± SE.

and soil organic matter) and its chemical stability in soil environments, making possible the fundamental assumption that Pb moves only with soil particles and that the major processes causing its redistribution in the landscape are mechanical processes such as water, wind and tillage erosion.

Prediction of the fate of metals in soil requires knowledge of their solid–liquid partitioning. Sorption and desorption are key processes involved in the interaction between soil and heavy metals. Several studies of the environmental fate of heavy metals have highlighted the strong sorption of Pb in soils (Alloway, 1995). Results reported by Welp and Brümmer (1999), based on several sorption and solubility tests, involving nine metals within A horizons of Hapic Podzol, Fimic Anthrosol, Eutric Cambisol and Calcaric Regosol soils and experimental conditions ranging from strongly acidic to slightly alkaline, showed that lead was the metal most strongly adsorbed by the soils tested, as compared to Mg, Sr, Co, Zn, Ni, Cd, Cr(III) and Cu (elements classified by increasing adsorption based on Freundlich K values). This behaviour was further confirmed for a loessic soil (hapic luvisol) by Welp (1999), with Pb exhibiting the highest sorption constant of the studied metals.

For most metals, pH and organic matter are the key parameters that control the partitioning between the solid and the liquid phase and therefore the adsorption and solubility of metals in soil (Welp and Brümmer, 1999; Degryse et al., 2009). Lead sorption in soil increases with pH and vice versa (see Adriano, 1986; Davies, 1995; Appel and Ma, 2002; Anagu et al., 2009). Some investigations have demonstrated that Pb is more strongly retained in soil than Cs (e.g. Klaminder et al., 2006). Also, Pb binds more strongly on Fe and Mn oxides than many other metals, such as Cu, Zn, and Cd (see Selim, 2011, 2012) and Pb is also preferentially associated with organic matter (see Dörr, 1995; Gustafsson et al., 2011), especially at acidic pH (Degryse et al., 2009).

Lead forms solid inner-sphere complexes with clay minerals (Strawn and Sparks, 1999), organic matter (e.g. Ferraz and Lourenco, 2000; Strawn and Sparks, 2000) and oxides (Trivedi et al., 2003). In addition, Pb forms precipitates with a moderate to low solubility in soil, such as hydroxides and phosphates in calcareous soils (Evans., 1989; Bradl, 2004).

Based on the available evidence (e.g. Selim, 2011, 2012), the bioavailability of Pb depends mostly on soil physico-chemical characteristics (e.g. texture, pH, soil organic matter (SOM), cation exchange capacity (CEC)) and the living phase of soil and plant types. Usually, Pb accumulates in the upper soil layers and is highly immobile. As highlighted by Davies (1995), except for a Ultisol with a low CEC and a loamy sand texture (i.e. Korte et al., 1976), there is little evidence of leaching of Pb in soil profiles. In addition, strong fixing to transported soil and sediment particles has been reported (Van Hoof and Andren, 1989; He and Walling, 1996b).

When reaching the soil surface as wet or dry fallout from the atmosphere, ^{210}Pb is therefore rapidly and strongly adsorbed by organic matter and clay-sized mineral particles and its subsequent redistribution across the landscape is mainly controlled by soil/sediment erosion and transport processes. The limited root absorption and limited uptake of lead by plants (Kabata Pendias and Pendia, 1992; Alloway, 1995; Davies, 1995; Klaminder et al., 2005), in combination with a strong affinity between the Pb atom and the surface of soil particles, means that the post-depositional redistribution of ^{210}Pb in the soil primarily reflects the redistribution of soil and sediment particles plus associated organic and mineral complexes over time (Klaminder and Yoo, 2008). However it should be pointed out, as highlighted by Davies (1995), Selim (2011, 2012) and Tamponnet (2009), that transfer of ^{210}Pb in the environment (especially soil–plant transfers) has been relatively poorly studied when compared to other radionuclides such as ^{137}Cs and ^{90}Sr.

Despite some high concentrations in kale, ryegrass or celery the relative Pb accumulation in plant species is limited (Alloway, 1995). Normally, Pb is neither biologically accumulated by the vegetation, nor leached by the water percolating through the soil profile. Moreover, even though some discrepancies were registered between stable and radioactive isotopes of Pb in freshwater environments, their partition coefficients and soil–plant transfer factors are in the same magnitude range (Tamponnet, 2009).

The amount of Pb absorbed by plants is affected by the pH, organic matter, and the phosphorus content of the soil. That is why a remediation strategy aimed at reducing the bioavailability and uptake of Pb by plants involves adjusting the pH of the soil with lime to a level of 6.5 to 7.0 (Tangahu et al., 2011). Some of the assumptions related to ^{210}Pb soil–plant interaction should undoubtedly be confirmed under controlled and/or natural conditions to clearly quantify the transfer factor and the adsorption/absorption of this isotope by plants under various environmental conditions. The influence of the vegetation cover on ^{210}Pb deposition should be verified for different soils and cultivation practices. Al-Masri et al. (2008) found transfer factors greater than 1.0 for soil–plant leaves for tomato, cucumber, broad bean and chickpea. However, the transfer of ^{210}Pb to plants is mainly due to the direct atmospheric deposition of ^{210}Pb to the above-ground portion of the plant, rather than transfer of supported ^{210}Pb via the root system (Bunzl and Trautmannsheimer, 1999; IAEA, 2009). This could influence the total ^{210}Pb inventory of soil profiles, especially in long-term undisturbed areas. However, it is debateable whether trapping of atmospheric dust containing ^{210}Pb by plants should be seen as representing a component of the local natural fallout input. The dust could represent fine particulate material resuspended from local or more distant sources. Furthermore, if the plants are absent in some locations, this input might be lacking. Loss of ^{210}Pb through biomass export must also be considered in a similar context. There is currently no consensus as to whether the ^{210}Pb activity concentration in plants should be included in the total inventory. The decision should arguably depend on careful consideration of the assumptions and requirements of the tracing application involved. In conclusion, more studies are needed to investigate and verify issues related to ^{210}Pb–plant interactions.

4. Technical aspects regarding the determination of $^{210}Pb_{ex}$

The accurate determination of $^{210}Pb_{ex}$ activity is a fundamental requirement for using this nuclear tool. In-depth knowledge and experience of gamma spectroscopy is required for its reliable determination in soil and sediment matrices. The determination of $^{210}Pb_{ex}$ activity requires the measurement of total ^{210}Pb activity and ^{226}Ra activity. The latter is equivalent to supported ^{210}Pb and so $^{210}Pb_{ex} = {}^{210}Pb_{total} - {}^{226}Ra$.

Several analytical techniques are available for the measurement of both ^{226}Ra and ^{210}Pb, including gamma spectrometry, alpha spectrometry and beta counting. The differences between them reflect achievable detection limit, laboratory workloads, reproducibility and analytical times and costs (Ebaid and Khater, 2006; IAEA, 2011). Gamma spectrometry has the advantage of being a non-destructive technique, allowing the simultaneous measurement of ^{210}Pb and ^{226}Ra and also other fallout radionuclides used as soil and sediment tracers, such as ^{137}Cs and ^{7}Be. This analytical method provides reliable results, generally with reduced time, effort and costs when compared with other techniques. Both gamma and alpha spectrometry can be employed for ^{226}Ra analysis in soil and/or sediment samples. ^{226}Ra can be measured by gamma spectrometry via the gamma ray energies of its daughters ^{214}Bi and ^{214}Pb, at 609.3 keV and 351.9 keV, respectively, or directly through its gamma line at 186.2 keV. The second method involves subtracting the contribution of ^{235}U from the overlapping peak, which can be difficult if the ^{235}U activity in the soil sample is very low. When ^{226}Ra is measured using its daughters ^{214}Pb or ^{214}Bi by gamma spectrometry, it is essential to ensure equilibrium with its direct gaseous daughter ^{222}Rn. To achieve equilibrium between ^{226}Ra and ^{222}Rn, soil samples should be properly sealed, to avoid radon escape, in high density polyethylene or aluminium containers for at least 21 days prior to performing gamma analysis. Zhang et al. (2009) demonstrated that the use of the doublet deconvolution method – to resolve the overlapping ^{235}U and ^{226}Ra gamma peaks at 185.7 keV and 186.2 keV – can provide a more accurate ^{226}Ra determination in soil samples, since it is independent of the error caused by the equilibrium assumption between the ^{222}Ra progenies and ^{226}Ra. The amount of radon gas escaping through the sample container walls should be also considered and quantified, even if sealing was performed. However, the ^{214}Pb and/or the ^{214}Bi approaches are preferred by most laboratories, due to the fact that they are easier.

In contrast, alpha-spectrometry offers the direct measurement of radium through its alpha-emission, which is independent of equilibrium or gaseous loss considerations. This method offers several advantages over other analytical techniques, including: (i) high sensitivity resulting from the high-yield alpha decay process, low intrinsic detector background and peak selectivity; (ii) high reliability associated with the use of an alpha-emitting isotope tracer; (iii) the wide range of radioisotopes that can be determined. Disadvantages are related to the need to use a tracer isotope solution and to the long and laborious chemical separation process (Hou and Roos, 2008).

^{210}Pb can be measured by gamma spectrometry using a high-purity germanium (HPGe) detector. This non-destructive measurement permits subsequent physico-chemical tests on the samples collected. However, the γ-rays are emitted at the low energy of 46.5 keV with a low emission probability of 4.25%. Therefore, the analysis of soils and sediments for ^{210}Pb by gamma spectrometry requires a low energy HPGe detector, with a thin front and side contact and little dead Ge beyond the active region, resulting in improved count rate performance and peak-to-background ratio (see Wallbrink et al., 2002b). 'N-type' germanium crystals with beryllium or carbon windows are commonly used for measuring both high and low energy gamma rays. Also, a stabilised ambient air temperature and low background environment must be assured by using conventional passive shielding (e.g. a thick layer of aged lead, coupled with copper and cadmium sheet) or even additional active shielding (e.g. gas or scintillation detectors surrounding the HPGe detector or NaI(Tl) anti-Compton shielding) when low detection limits of e.g. 1 mBq are required (Kozak et al., 2001; Povinec, 2008). Particular attention should also be devoted to self-absorption, which varies as a function of the chemical composition and density of the matrix. It is necessary to apply a self-attenuation correction in order to obtain a precise determination of the activity concentration. Several protocols and approaches have been proposed to take into account and to assess this factor (e.g. Cutshall et al., 1983; Boshkova and Minev, 2001; San Miguel et al., 2002; Jodlowski, 2006; Quindos et al., 2006; Hult, 2007; Jodlowski, 2007; Saïdou et al., 2007; Khater and Ebaid, 2008; Robu and Giovani, 2009; Jodlowski et al., 2014). A recent International Atomic Energy Agency (IAEA) proficiency test on gamma spectrometry measurement of total ^{210}Pb in spiked soil samples highlighted a clear need for improvement with less than 40% of the 14 participating laboratories able to provide an accurate measurement of this isotope (see Shakhashiro and Mabit, 2009). Furthermore, Albrecht (1999) reported that measurement of the ^{210}Pb and ^{137}Cs activity of sediment samples collected from an alpine catchment by gamma spectroscopy, using a well detector shielded with an anticoincidence proportional chamber provided an overall error of measurement for ^{210}Pb at least two times higher than that for ^{137}Cs. Therefore, quality control procedures, proficiency tests and inter-comparison exercises should be encouraged, to assess the validity and reliability of the results of ^{210}Pb analysis by gamma spectrometry.

Another important consideration associated with obtaining accurate values of $^{210}Pb_{ex}$ activity relates to the need to apportion the measurement of total ^{210}Pb between its supported and unsupported (or excess) components. As indicated above, this is usually achieved by assuming that the supported ^{210}Pb is in secular equilibrium with ^{226}Ra, which is also measured, and subtracting this value from the measurement of total ^{210}Pb. However, if additional input or removal of any of the parents of ^{210}Pb occurs, secular equilibrium is not established (Ravichandran et al., 1995). In most environments, the supported ^{210}Pb will not be in equilibrium with ^{226}Ra, since some ^{222}Rn will diffuse upwards through the soil or rock and escape to the overlying atmosphere. This loss is commonly quantified by the emanation coefficient (the proportion of the ^{222}Rn that escapes from the soil or rock), which typically has a value of ca. 0.2–0.3 (Du and Walling, 2012). Failure to take account of this loss of ^{222}Rn will result in overestimation of the supported ^{210}Pb and therefore underestimation of the $^{210}Pb_{ex}$, including the generation of negative values of $^{210}Pb_{ex}$ (see Du and Walling, 2012). This problem can be addressed by applying a correction or reduction factor, based on the emanation coefficient, to the estimated supported ^{210}Pb activity. This reduction factor will be influenced by the texture, bulk density and moisture content of the parent material and can prove difficult to quantify, particularly when determining the $^{210}Pb_{ex}$ activity of fluvial sediment mobilised from its original in situ source. However, when calculating the $^{210}Pb_{ex}$ activity of soils, the reduction factor can be estimated from the average ratio of the measured total ^{210}Pb and ^{226}Ra concentrations for samples collected from the lower part of the soil profile (e.g. 40–50 cm in an eroding soil), where $^{210}Pb_{ex}$ can be assumed to be absent (e.g. Wallbrink and Murray, 1996). This ratio is frequently of the order of 0.8. This measurement problem is of more limited significance for submerged lake sediment deposits, which are characterized by very low emanation coefficients. Ravichandran et al. (1995) calculate an Rn escape probability of less than 0.5 for the sediments of some stations in the Sabine–Neches Estuary, Texas (USA) and concluded that diffusion of Rn did not significantly affect the $^{210}Pb_{ex}$ profile. As indicated by Ravichandran et al. (1995), this is a very small correction which could be due to the significantly higher sedimentation rates in the study area (for more information see also Imboden and Stiller, 1982; Stiller and Imboden, 1986).

Despite recent reports of the applicability of portable gamma spectrometry for detecting ^{210}Pb in soil (Li et al., 2010), in-situ measurement of ^{210}Pb is almost impossible due to the self-absorption of the low energy 46.5 keV gamma ray in the soil. For example, assuming a soil density of 1.65 g cm^{-3} (soil composition: 68% SiO_2, 10% H_2O, 1% C, 3% Fe, 7% Al and 11% O_2), based on the gamma attenuation law, 97% of the emitted radiation will be absorbed by the first 5 cm of the soil, with an additional 3% absorption in dry air if the detector is placed 1 m above ground level.

The indirect approach to determining $^{210}Pb_{ex}$ activity from measurements of total ^{210}Pb and ^{226}Ra induces potential errors associated with the measurement of both ^{210}Pb and ^{226}Ra and results in a low precision for the final estimate of $^{210}Pb_{ex}$. Therefore, other methods of quantifying total ^{210}Pb, such as alpha spectrometry and beta or liquid scintillation counting, have been suggested and tested (e.g. Ebaid and Khater, 2006; Jia and Torri, 2007; Zaborska et al., 2007; Baskaran, 2011b).

^{210}Pb concentration can be indirectly determined by alpha-ray spectrometry through its decay progeny ^{210}Po. This radiochemical method is based on a total digestion of the sample and the separation of ^{210}Pb from other radionuclides present in the sample, followed by auto-deposition of its daughter nuclide, ^{210}Po on a silver, nickel or stainless steel disc and further measurement with an alpha spectrometer generally using a Silicon Surface Barrier Detector (SSBD) or Passivated Implanted Planar Silicon (PIPS) detectors (see Vesterbacka and Ikäheimonen, 2005; Ebaid and Khater, 2006). This method is very sensitive with an excellent detection limit and assumes that secular equilibrium is reached between ^{210}Pb and ^{210}Po. This condition is usually fulfilled in the case of undisturbed lake or marine sediments with low sedimentation rates. In most cases, equilibrium between ^{210}Pb and ^{210}Po is not established. Ideally, soil samples should be kept for a minimum of 2 years (around 7 times the half-life of ^{210}Po) to reach secular equilibrium. Otherwise, to reduce the time delay before obtaining reliable measurements of ^{210}Pb activity, the remaining solution with no ^{210}Po should be stored for a period of 3–6 months and ^{210}Po should again be analysed after its significant growing from ^{210}Pb (Ebaid and Khater, 2006). Also, liquid scintillation counting (LSC) after radiochemical separation of ^{210}Pb, based on a final precipitation of $PbSO_4$, represents a suitable alternative to other spectrometric techniques for direct determination of ^{210}Pb activity through its beta energy line at 63.5 keV (see Villa et al., 2007). The method requires a suitable chemical procedure, careful calibration of the low-background scintillation spectrometer and spectrum correction for the contribution of ingrowing ^{210}Bi to the total ^{210}Pb region. The radiochemical yield can be gravimetrically determined. The main technical advantage of both methods (i.e. alpha spectrometry and LSC) is the ability to provide measurements with low detection limits. However, they are usually more labour intensive and expensive than gamma spectroscopy and it should be kept in mind that these analytical methods are 'destructive'.

Mabit et al. (2008) provide a comparison of alpha and gamma spectrometry measurements. The authors demonstrated under experimental conditions that for low $^{210}Pb_{ex}$ activities in the soil matrix, the difference between alpha and gamma spectrometry measurements of total ^{210}Pb is high and that alpha spectrometry performs much better than gamma analysis. The relative uncertainty associated with gamma spectrometry determinations for a $^{210}Pb_{ex}$ activity of 10 Bq kg^{-1} can reach 40%, although for higher activities (>30 Bq kg^{-1}) uncertainties become acceptable (<15%) and when activities exceed 120 Bq kg^{-1}, uncertainty levels are close to those associated with alpha spectrometry (Mabit et al., 2008). Readers are also referred to an intercomparison of the three main analytical techniques used to measure ^{210}Pb reported by Villa et al. (2007). These authors compared the accuracy of the results obtained by LSC for quantifying ^{210}Pb in sediment samples from the estuary of the Odiel and Tinto Rivers (SW Spain) with two other alternative methods (i.e. alpha- and gamma-spectrometry). They found the three methods to give identical results within the uncertainty of the measurements, with the exception of a few samples. These exceptions were thought likely to be related to incomplete homogenization of the associated samples (see Villa et al., 2007).

However, even if chemical separation methods can provide more accurate results for total ^{210}Pb activity, there is still a need to measure the ^{226}Ra content of a sample using gamma spectrometry, in order to determine $^{210}Pb_{ex}$. The measurement of ^{210}Pb and ^{226}Ra using radiochemical separation methods (alpha spectrometry and/or LSC) could prove appropriate when using a limited number of samples with low activities to determine sedimentation or soil erosion rates.

To summarise, if a sample is expected to have a high ^{210}Pb activity (e.g. >5 Bq kg^{-1}), gamma-spectrometry is recommended. However, if the expected activity is <5 Bq kg^{-1}, either alpha-spectrometry or LSC could be more appropriate. Alpha spectrometry can achieve low detection limits. The choice between alpha spectrometry and LSC should be guided by the accuracy and detection limit required and the time constraints of the study. For a more in-depth assessment of the comparative advantages and limitations of ^{210}Pb measurement by radiochemical separation methods and gamma spectrometry, the reader is referred to Desideri et al. (2011), Ebaid and Khater (2006), Jia and Jia (2012), Saïdou et al. (2008), Villa et al. (2007) and Zaborska et al. (2007).

5. Using $^{210}Pb_{ex}$ for documenting soil redistribution and sediment sources

The principles associated with the use of $^{210}Pb_{ex}$ for the assessment of soil redistribution have been described in several publications (e.g. Walling and He, 1999b; Walling et al., 2003a; Zhang et al., 2006; Mabit et al., 2008; Matisoff and Whiting, 2011). The basic principles underlying the method are simple, but there are a number of methodological issues which require careful consideration and some pitfalls which can prove serious obstacles to the successful application of the approach.

The use of FRNs to investigate soil redistribution rates has several important advantages over traditional erosion measurement or assessment methods (see Walling and Quine, 1991; Mabit et al., 2008; Zupanc and Mabit, 2010). The use of $^{210}Pb_{ex}$ for documenting soil redistribution rates involves the same advantages (e.g. global distribution, retrospective assessment, spatially distributed data, only one sampling campaign required for assessing the magnitude of both erosion and deposition) and limitations (e.g. choice of reliable reference site, specific detection system) as other FRNs (see Mabit et al., 2008; Matisoff and Whiting, 2011). In addition, it has the following additional advantages: (i) the use of $^{210}Pb_{ex}$ can extend the timescale associated with other FRN measurements to provide a retrospective assessment of longer-term soil redistribution rates over a period of up to 100 years; and (ii) since the fallout of ^{210}Pb is continuous, its use does not face the problems associated with short- and medium-lived anthropogenic radioactive soil tracers such as ^{137}Cs.

The general principle underpinning the use of $^{210}Pb_{ex}$ as a soil tracer is the comparison of the areal activity or inventory of $^{210}Pb_{ex}$ (Bq m^{-2}) of the sampling point with the inventory of an undisturbed site or 'reference site'. If the inventory at the sampling point is significantly less than the reference inventory, erosion can be assumed to have occurred, whereas, if it exceeds the reference inventory, deposition can be assumed to have occurred. The use of appropriate conversion models allows the quantification of soil erosion and deposition rates (t ha^{-1} yr^{-1}) associated with a period of ca. 100 years, which corresponds to about five half-lives of ^{210}Pb.

The vertical distributions of $^{210}Pb_{ex}$ associated with undisturbed and cultivated soils are very similar to those documented for ^{137}Cs (see Fig. 3), however the continuous input of ^{210}Pb results in a maximum activity occurring at the surface (Walling et al., 1995).

Conversion of $^{210}Pb_{ex}$ inventories to soil erosion rates follows the same principles as for the ^{137}Cs technique (Walling et al., 2011). In order to derive estimates of soil redistribution rates, two mass balance models have been developed for use with the $^{210}Pb_{ex}$ inventories of samples collected from cultivated sites. They represent adaptations of mass balance models (MBM) 2 and 3 developed for the more widely used ^{137}Cs (see Walling and He, 1999c; Walling et al., 2002, 2003a, 2011). The MBM 3 includes a tillage component, whereas MBM 2 does not. An adaptation of the diffusion and migration model developed for the use of ^{137}Cs in uncultivated areas has also been successfully applied to convert $^{210}Pb_{ex}$ inventories obtained from non-cultivated (e.g. pasture and rangeland) areas (Walling et al., 2003a). In addition to the need to change the $^{210}Pb_{ex}$ decay constant, which differs from that for ^{137}Cs, Walling et al. (2011) made two key modifications to the ^{137}Cs conversion models upon which they were based. Firstly, the modelling period extends back 100 years from the sampling date. It is assumed that the influence of any ^{210}Pb deposition prior to this point in time is insignificant due to decay. In theory, only around 4% of the original deposition will remain after 100 years. The time of sampling is not used in the calculation. It is only relevant for interpreting the results obtained from the model. Secondly, it is assumed that the annual ^{210}Pb fallout remains effectively constant through time. The reference inventory will represent a steady state balance between input and decay, and thus will also remain constant over time. The annual deposition flux (I) can be determined from the local reference inventory (A_{ref}) and the decay constant of ^{210}Pb (λ_{Pb}) as:

$$I = A_{ref}\ \lambda_{Pb}. \tag{1}$$

Most of the parameters used in the conversion models for $^{210}Pb_{ex}$ inventories are very similar to those used for the ^{137}Cs models. However, a degree of caution is needed in their specification, since the behaviour of ^{210}Pb in soils may differ from that of ^{137}Cs. One key difference is the greater preference of ^{210}Pb for organic matter, with the result that ^{210}Pb is a better tracer of organic and minerogenic particles in combination.

There is a need for a wider testing of the $^{210}Pb_{ex}$ conversion models to generate an improved understanding of their advantages and limitations and thus assist the further development and improvement of such models and such work is already being reported (see Porto and Walling, 2012; Porto et al., 2013). Attention should also be given to the careful interpretation of the results provided by $^{210}Pb_{ex}$ (alone or in combination with other radiotracers), because its inventories in soils will be influenced by soil erosion rates over the last ca. 100 years, but are also likely to be more sensitive to the recent soil redistribution rates when compared to ^{137}Cs (Porto et al., 2009, 2013). Therefore $^{210}Pb_{ex}$ has been promoted for assessing the effectiveness of soil conservation measures and for investigating climate change impact on agricultural landscapes (e.g. Dercon et al., 2012).

The main limitation of the $^{210}Pb_{ex}$ approach is the difficulty of accurately determining $^{210}Pb_{ex}$ by laboratory gamma spectrometry. Additional sources of ^{222}Rn to the atmosphere may also create problems. ^{222}Rn and its short-lived decay products are commonly present in the atmosphere at much lower levels than in the soil. However, increased activities can occur in the vicinity of volcanoes, thermal spas, and other sources of gas emanating from zones of the Earth's surface where rocks or minerals with high radium content are found. In some locations, the $^{210}Pb_{ex}$ content in soil has been found to be very low and even below detection limits. This could reflect either very high soil erosion rates or a low ^{210}Pb fallout deposition flux, and clearly constitutes a major limitation to the use of $^{210}Pb_{ex}$ in some areas. Where the total ^{210}Pb content of a soil is very close to the content of supported ^{210}Pb indicated by the ^{226}Ra content, the uncertainty associated with determining $^{210}Pb_{ex}$ may be too high. Limitations of the use of this isotope in some environmental applications such as sediment dating in New Zealand (Graham et al., 2004) and as a soil tracer under certain conditions, including frozen soil (Golosov et al., 2011) have already been reported. Consequently, like ^{137}Cs (Parsons and Foster, 2011; Mabit et al., 2013), $^{210}Pb_{ex}$ is not applicable everywhere. For example, very low levels of $^{210}Pb_{ex}$ activity (due to either high erosion process

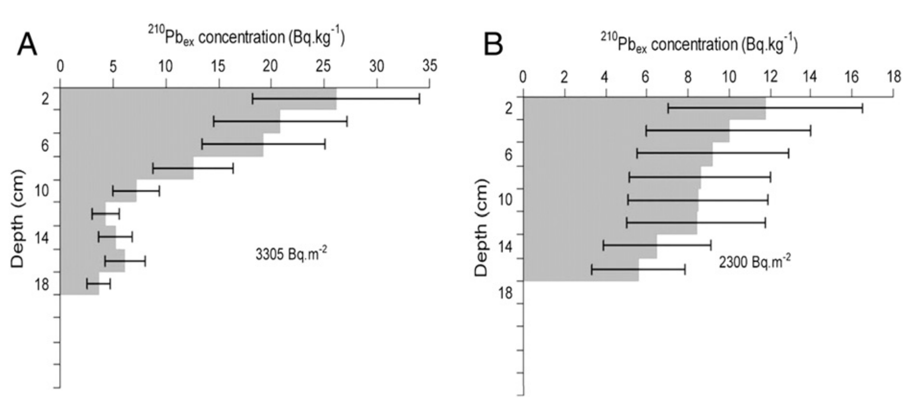

Fig. 3. Examples of $^{210}Pb_{ex}$ depth distribution in agricultural soil profiles in Morocco (A: Undisturbed soil; B: Cultivated eroded soil). Adapted from Benmansour et al. (2011).

or low ^{210}Pb input) coupled with a high spatial variability and a high measurement error may inhibit the use of this FRN (see Mabit et al., 2009, 2010).

The combined use of FRNs offers considerable potential to investigate the erosional history of a study site and the impact of land use and climate change, by providing information for different time windows (see Mabit et al., 2008; Dercon et al., 2012; Guzmán et al., 2013). In addition, use of two or more different and essentially independent radionuclides can increase the confidence in the results obtained. In most cases it is possible to measure the different FRNs at the same time using the same samples. In this context $^{210}Pb_{ex}$ is often used in combination with ^{137}Cs to provide information on soil redistribution over the past 100 years. If soil redistribution rates remain essentially constant over this period, the spatial variation of $^{210}Pb_{ex}$ inventories across a study site should theoretically be similar to that associated with ^{137}Cs inventories. Table 1 provides examples of where $^{210}Pb_{ex}$ has been used in combination with ^{137}Cs and includes studies where the results obtained by the radionuclides can be compared with independent evidence of soil and sediment redistribution rates established by more conventional monitoring techniques. In most cases the different radionuclides provide similar results, but contrasts can reflect land use change over the period of interest. Equally the results provided by the FRNs compare well with those provided by other direct and indirect monitoring techniques. Fig. 4 emphasises the general consistency between the results provided by $^{210}Pb_{ex}$ and ^{137}Cs for the soil erosion studies included in Table 1, by plotting the relationship between the results derived from two alternative approaches. This demonstrates a near 1 to 1 and highly significant linear relationship (i.e. $y = 1.07x - 0.642$; $r^2 = 0.93$).

In addition to the use of $^{210}Pb_{ex}$ for documenting soil redistribution rates, as discussed above, the radionuclide has also been successfully employed in a wide range of sediment source fingerprinting studies (e.g. He and Owens, 1995; Walling and Woodward, 1995; Owens and Walling, 2002; Froehlich, 2004). Such studies aim to establish the relative contribution of a range of potential sediment sources to the suspended sediment load of a river (see Walling, 2005, 2006). In this context, the $^{210}Pb_{ex}$ activity of a sample of the fine sediment transported by a river or stream can provide a sensitive fingerprint of the source of that sediment. Because the occurrence of $^{210}Pb_{ex}$ is primarily restricted to the surface horizons of the soil, by virtue of its fallout origin, the radionuclide affords an effective means of discriminating between sediment mobilised from surface sources and that originating from subsurface sources such as river banks and gullies. In addition, the mixing of the radionuclide into the plough layer by cultivation means that sediment mobilised from the surface of uncultivated land is likely to have a higher $^{210}Pb_{ex}$ activity than that mobilised from the surface of cultivated land (Walling and Woodward, 1992; Wallbrink and Murray, 1993). The contrast between the temporal behaviour of ^{210}Pb and ^{137}Cs, with the former being essentially continuous and the latter restricted to the period of bomb testing, means that the behaviour of the two radionuclides in surface materials could differ. For example, erosion of the surface of an uncultivated soil could remove all the ^{137}Cs originally contained in the surface material, but the $^{210}Pb_{ex}$ activity will be replenished by ongoing fallout. In most sediment source fingerprinting studies, there is a need to discriminate several potential sources and therefore $^{210}Pb_{ex}$ is commonly used in conjunction with other FRNs and/or other sediment properties (e.g. geochemical, mineral magnetic, bulk and isotopic carbon and nitrogen, and compound specific stable isotopes) as part of a composite fingerprint (e.g. He and Owens, 1995; Walling and Woodward, 1995; Wallbrink and Murray, 1996; Walling et al., 1999; Huisman and Karthikeyan, 2012; Owens et al., 2012; Wilson et al., 2012; Hancock and Revill, 2013; Olley et al., 2013).

In another application, several workers (e.g. Matisoff et al., 2005; Evrard et al., 2010) have proposed that the $^{7}Be/^{210}Pb_{ex}$ ratio in the fine sediment transported by river can provide evidence of its 'age' or , alternatively, the relative contribution of 'new' and 'old' sediment to the river's suspended sediment load. In this context 'age' refers to the time the sediment has remained in the river channel since being eroded and transported from the surrounding catchment. 'New' and 'old' similarly refer to sediment newly delivered to the channel which will be ^{7}Be enriched and ^{7}Be deficient sediment resuspended from the channel bed or banks. It is argued that since both ^{7}Be and $^{210}Pb_{ex}$ have a similar source as contemporary atmospheric fallout, fallout will be characterized by a distinctive $^{7}Be/^{210}Pb_{ex}$ ratio and freshly labelled sediment will evidence a similar ratio. Because ^{7}Be has a short half-life, the $^{7}Be/^{210}Pb_{ex}$ ratio associated with freshly mobilised suspended sediment will progressively decline as it remains longer in the system and the 'age' of the sediment can be estimated from the magnitude of the ratio. Equally, if freshly mobilised or 'new' suspended sediment is assumed to have a ratio close to that associated with fallout, and 'dead' sediment is assumed to have ratio approaching zero, the proportion of 'new' sediment will be directly related to the value of the ratio. However, while attractive in its apparent simplicity, Walling (2013) has shown that approach involves a number of assumptions that compromise such interpretation of the $^{7}Be/^{210}Pb_{ex}$ ratio.

While, there has been much recent interest in the use of Pb as a tracer of soil and sediment movement on hillslopes and through river basins, several studies have shown that, under certain circumstances, the $^{210}Pb_{ex}$ signal may be altered during its transport through the landscape and therefore be non-conservative. For example, studies of wildfire-affected landscapes (e.g. Blake et al., 2009; Wilkinson et al., 2009; Owens et al., 2012; Perreault et al., 2013) have shown that $^{210}Pb_{ex}$ can be concentrated in surface soils due to the combustion of organic matter at the soil surface (e.g. leaf litter) by the fire and this may influence its ability to trace soil and sediment redistribution in such landscapes (Smith et al., 2013). While this concentration effect has also been noted for other FRNs, like ^{137}Cs, the stronger association of $^{210}Pb_{ex}$ with organic matter means that it may be more pronounced than other FRNs. The well-recognised affinity of $^{210}Pb_{ex}$ to SOM has been exploited by several innovative studies aimed at improving current understanding of carbon fluxes and dynamics in the soil profile associated with erosion processes and with the impact of land use changes (e.g. Yoo et al., 2011; Özden et al., 2013; Teramage et al., 2013). Similarly, Braakhekke et al. (2011, 2012) have recently initiated studies using $^{210}Pb_{ex}$ as a tracer of SOM transport in mineral soil profiles in The Netherlands and in Central Germany.

We believe that greater emphasis on developing a better understanding of spatio-temporal patterns of $^{210}Pb_{ex}$ is essential to upscale its use to the watershed and river basin scales using adapted sampling strategies, GIS and geostatistical tools. Furthermore, investigating the relationship between this FRN and soil quality indicators could provide additional information regarding soil fertility degradation processes.

6. Assessing sedimentation rates and dating sediment in freshwater lakes and reservoirs

True varves, which occur rarely in most sedimentary sequences, with the exception of lakes in glacierized basins (Ojala et al., 2012), and other non-radiometric dating methods (e.g. fossil markers, pollen, pollution markers) can provide important stratigraphic time-markers. However, radiometric dating is the only technique of general applicability that is able to provide an absolute age determination for lake sediments (Caroll and Lerche, 2003).

The capacity of $^{210}Pb_{ex}$ determinations to provide information on the chronology of a sediment deposit and thereby to estimate the sedimentation rate is founded on its source and relatively long half-life. In general terms, the rate of decline of $^{210}Pb_{ex}$ activity with depth in a sediment core provides the basis for establishing an age–depth relationship and for estimating sediment accumulation rates (SAR). If SAR are high, the $^{210}Pb_{ex}$ activity will decline slowly with depth. On the other hand, if accumulation rates are low, the $^{210}Pb_{ex}$ activity will reduce rapidly with depth.

Table 1
Comparison of soil redistribution magnitudes provided by FRN (i.e. ^{137}Cs and $^{210}Pb_{ex}$) and conventional direct/indirect measurements.

Reference(s)	Location	Climate	Type of site	Objective(s)	Mean reference inventory (Bq m^{2})		Net soil redistribution		
					^{137}Cs	^{210}Pb	^{137}Cs	^{210}Pb	Conventional methods
Benmansour et al. (2013)	Marchouch (6°42′W, 33° 47′N), 68 km south east from Rabat, Morocco	Mediterranean semiarid climate	Experimental agricultural field	Estimate soil redistribution rates and the assessment of the effectiveness of soil conservation techniques	Undisturbed site (n = 12): 1445 Bq m^{-2}	Undisturbed site (n = 12): 3305 Bq m^{-2}	−14.3 t ha^{-1} yr^{-1} (MBM2) and −15.5 t ha^{-1} yr^{-1} (MBM3)	−12.1 t ha^{-1} yr^{-1} (MBM)	−12.1 t ha^{-1} yr^{-1} (RUSLE2; period: 10 years)
Belyaev et al. (2004)	Tver region, the Osuga river basin, northwest of the Russian Plain	Continental, precipitation of 600 mm	Agricultural field	Evaluate rates of soil and sediment redistribution during a period of intensive agriculture	4013 Bq m^{-2} (n = 5)	8392 Bq m^{-2} (n = 3)	−24.2 t ha^{-1} yr^{-1}	−10 t ha^{-1} yr^{-1}	−12 t ha^{-1} yr^{-1} USLE (period of 48 years)
Blake et al. (2009)	Blue Gum Creek catchment, Nattai National Park, New South Wales, Australia	Humid temperate climate	Wildfire affected eucalypt forest	Deriving sediment budget for wildfire-affected forested area	Burnt plateau area (n = 20): 493 ± 34 Bq m^{-2}	Burnt plateau area (n = 20): 1923 ± 171 Bq m^{-2}	−0.19 ± 0.06 t ha^{-1} yr^{-1}	−0.75 ± 0.13 t ha^{-1} yr^{-1}	–
O'Farrell et al. (2007)	Haypress basin, Tennessee Valley, in the Marin Headlands of northern California, USA	Mediterranean, with annual precipitation of 760 mm	Undisturbed soils covered with coastal grasses; poison oak and coyote bush on the hollows	Estimates of the erosion rates and processes	Flat crest of the catchment (n = 2): 1102 and 1057 Bq m^{-2}	Flat crest of the catchment (n = 2): 1508 and 1904 Bq m^{-2}	−2.0 ± 0.6 t ha^{-1} yr^{-1} (DMM)	−2.7 ± 0.7 t ha^{-1} yr^{-1} (DMM)	−3.2 ± 0.9 t ha^{-1} yr^{-1} (pound sediment volume)
Fukuyama et al. (2008)	Near the town of Taiki, in Mie prefecture, central Japan (136°25′E, 34°21′N)	Humid subtropical climate	Japanese cypress plantations	Confirming the dominant erosion processes	Undisturbed, ridge top of the study catchment (n = 1, using scraper plate): 1710 Bq m^{-2} and (n = 45, using a steel core sampler): 2948 Bq m^{-2}	Undisturbed, ridge top of the study catchment (n = 1, using scraper plate): 4354 Bq m^{-2} and (n = 45, using a steel core sampler): 8453 Bq m^{-2}	−2.1 t ha^{-1} yr^{-1} (DMM)	−3.3 t ha^{-1} yr^{-1} (DMM)	–
Gaspar et al. (2013)	Estaña lake catchment, central part of the Pre-Pyrenees, NE Spain	Continental Mediterranean	Different land use (natural forest, abandoned fields and cultivated fields)	Test the applicability of the combined FRN techniques in soil erosion studies for different soil types, land use and slope gradients	Level undisturbed sites with a mature and natural vegetation cover (n = 9): 1570 ± 80 Bq m^{-2}	Level undisturbed sites with a mature and natural vegetation cover (n = 3): 1943 ± 78 Bq m^{-2}	Mean erosion: −14.5 ± 9.3 t ha^{-1} yr^{-1} mean deposition: 5.9 ± 7.4 t ha^{-1} yr^{-1}	Mean erosion: −9.6 ± 24.6 t ha^{-1} yr^{-1} mean deposition: 22.2 ± 39.0 t ha^{-1} yr^{-1}	–
Kato et al. (2010)	Kherlen-bayan Ulaan (KBU) and in Baganuur (BGN) within the Kherlen River Basin, eastern Mongolia	Cold, semi-arid climate	Steppe grassland with tall-grass prairie	Soil erosion estimates and applicability of the $^{210}Pb_{ex}$ technique for semi-arid grass hillslopes	Permanent grassland: 1889 Bq m^{-2} (KBU) and flat terrace: 1696 Bq m^{-2} (BGN)	Permanent grassland: 6310 Bq m^{-2} (KBU) and flat terrace: 6427 Bq m^{-2} (BGN)	KBU catchment: −1.44 t ha^{-1} yr^{-1} (DMM) BGN catchment: −0.49 t ha^{-1} yr^{-1} (DMM)	KBU catchment: −2.29 t ha^{-1} yr^{-1} (DMM) BGN catchment: −1.14 t ha^{-1} yr^{-1} (DMM)	–
Porto et al. (2006)	Calabria, southern Italy,	Mediterranean climate	Three small forest/rangeland catchments	Explore the use of fallout ^{210}Pb to estimate rates of water-induced soil erosion on uncultivated land	Close to a permanent grassland with minimal slope: 2492 Bq m^{-2}, a small clearing between the trees: 2430 Bq m^{-2}, close to an undisturbed rangeland with some scattered oaks: 2445 Bq m^{-2}	An area of undisturbed rangeland with minimal slope within catchment W1, scrapper plate (n = 1) 5266 Bq m^{-2}, soil cores (n = 6): 5440 Bq m^{-2}	−8.6 t ha^{-1} yr^{-1} for W1 −14 t ha^{-1} yr^{-1} for W2 −11.3 t ha^{-1} yr^{-1} for W3	−6.2 t ha^{-1} yr^{-1} for W1 −12.1 t ha^{-1} yr^{-1} for W2 −11.1 t ha^{-1} yr^{-1} for W3	−12.4 t ha^{-1} yr^{-1} for W1 −19.2 t ha^{-1} yr^{-1} for W2 −7.80 t ha^{-1} yr^{-1} for W3

Porto and Walling (2012)	40 km north of Reggio Calabria in southern Italy	Mediterranean climate	Plots with different cropping and tillage systems	Validation of the FRN-based techniques for soil erosion studies	Cultivated field very close to the study site ($n = 8 + 1$): 5321 ± 104 Bq m^{-2}	Cultivated field very close to the study site ($n = 8 + 1$): 7145 ± 227 Bq m^{-2}	-9.5 to -12.0 t ha^{-1} yr^{-1} (MBM with tillage)	-6.5 to -18.2 t ha^{-1} yr^{-1} (MBM with tillage)	-9.4 to -38.5 t ha^{-1} yr^{-1} (5 runoff plots with different management systems; period: 4 years)
Porto et al. (2013)	Bonis catchment (1.39 km^2), mountain area of Sila Greca, Calabria, southern Italy	Dry period in summer and rainfall almost uniformly distributed in the rest of the year	Forest stands (93%) and no tree cover (6%)	Establishing sediment budget	Small clearing between the trees and 2 undisturbed rangelands with some scattered pines (composite slices of 24 cores): 3112 Bq m^{-2}	Small clearing between the trees and 2 undisturbed rangelands with some scattered pines (composite slices of 24 cores): 10,900 Bq m^{-2}	Slopes: -3.69 t ha^{-1} yr^{-1} (DMM) catchment divides: -1.99 t ha^{-1} yr^{-1} (DMM) valley bottoms: 3.58 t ha^{-1} yr^{-1} (DMM)	Slopes: -4.05 t ha^{-1} yr^{-1} (DMM) catchment divides: -4.83 t ha^{-1} yr^{-1} (DMM) valley bottoms: 1.27 t ha^{-1} yr^{-1} (DMM)	Sediment output from the study catchment: 0.25 t ha^{-1} yr^{-1} (period: 5 years)
Porto et al. (2014)	Small (0.86 ha) cultivated catchment in Sicily, Italy	Mediterranean climate	Cultivation since the early 1950s mainly with wheat	Assess the relative contribution of interrill/rill and gully erosion to soil loss	Permanent pasture with minimal slope ($n = 8$): 432 Bq m^{-2}	Permanent pasture with minimal slope ($n = 8$): 2800 Bq m^{-2}	-38.8 t ha^{-1} yr^{-1}	-34.2 t ha^{-1} yr^{-1}	–
Wakiyama et al. (2010)	Tsuzura River watershed, a midstream tributary of the Shimanto River basin, in southern Japan	Temperate monsoon humid	Japanese cypress plantation, natural broadleaf forest stands, and Japanese cedar plantation	Applicability of the ^{210}Pb$_{ex}$ technique	Downstream site of the flat ridge tops ($n = 2$): 3067 Bq m^{-2}	Downstream site of the flat ridge tops ($n = 2$): 19703 Bq m^{-2}	Japanese cypress stand (bare): -1.61 t ha^{-1} yr^{-1} (DMM) Japanese cypress stand (fern): -1.13 t ha^{-1} yr^{-1} (DMM) Japanese cedar stand (bare): -0.58 t ha^{-1} yr^{-1} (DMM) Broadleaf forest stand: -0.83 t ha^{-1} yr^{-1} (DMM)	Japanese cypress stand (bare): -1.24 t ha^{-1} yr^{-1} (DMM) Japanese cypress stand (fern): -1.14 t ha^{-1} yr^{-1} (DMM) Japanese cedar stand (bare): 0.65 t ha^{-1} yr^{-1} (DMM) Broadleaf forest stand: 0.98 t ha^{-1} yr^{-1} (DMM)	Japanese cypress: -9.35 t ha^{-1} yr^{-1} (period: 2 years) Japanese cedar: -0.97 t ha^{-1} yr^{-1} (period: 2 years) broadleaf forest: -6.99 (1 plot) and -2.70 (10 runoff plots) (period: 2 years)
Walling et al. (1995)	Butsford Barton near Colebrooke, Devon, UK	Temperate maritime	Small cultivated field	Comparing the estimates provided by ^{137}Cs and unsupported ^{210}Pb$_{ex}$ measurements	Neighbouring areas of undisturbed permanent grassland	Neighbouring areas of undisturbed permanent grassland	Net erosion rate for the field: -6.5 t ha^{-1} yr^{-1}	Net erosion rate for the field: -9 t ha^{-1} yr^{-1}	–
Walling et al. (2003a) Collins et al. (2001)	Upper Kaleya River basin (63 km^2), southern Zambia (16°11′S, 28°02′E)	distinct wet (November–March) and dry (April–October) seasons	Cultivated and grazing land within areas of communal and commercial farming	Applicability of the ^{210}Pb$_{ex}$ technique	Located within the study catchment and characterized by minimal slope ($n = 36$): 202.0 ± 22.8 Bq m^{-2}	Undisturbed location ($n = 12$): 2670 ± 402 Bq m^{-2}	Commercial cultivation: 4.3 t ha^{-1} yr^{-1} (DMM) communal cultivation: 2.5 t ha^{-1} yr^{-1} (DMM) bush grazing: 2.9 t ha^{-1} yr^{-1} (DMM)	Commercial cultivation: 4.5 t ha^{-1} yr^{-1} (DMM) communal cultivation: 1.2 t ha^{-1} yr^{-1} (DMM) bush grazing: 3.7 t ha^{-1} yr^{-1} (DMM)	–
Yang et al. (2011)	Mojiagou Basin, on the outskirts of Changchun, Jilin Province, Northeast China	Temperate sub-humid continental monsoon	Cultivated field (corn), ploughed along the contours	Validation of the FRN-based techniques for soil erosion studies	Sites with gentle slopes and wild grass ($n = 5$): 2918 ± 420 Bq m^{-2}	Sites with gentle slopes and wild grass ($n = 5$): 8954 ± 360 Bq m^{-2}	-4.5 to -37.6 t ha^{-1} yr^{-1} (MBM)	-3.4 to -38.1 t ha^{-1} yr^{-1} (MBM)	–
Zheng et al. (2007)	Shangqiao catchment, Neijiang, Sichuan Province, China	Subtropical climate	Cultivated field manually tilled from the bottom to the top of the slope	Soil erosion estimates associated with manual tillage	Grassland and flat cultivated land, and undisturbed area near a pine forest ($n = 6 + 1$): 2065 Bq m^{-2}	Grassland and flat cultivated land, and undisturbed area near a pine forest ($n = 6 + 1$): 18,902 Bq m^{-2}	-0.20 to -0.68 cm yr^{-1} (MBM) (mean annual rate of net soil loss 31.9 t ha^{-1} yr^{-1})	-0.19 to -0.78 cm yr^{-1} (MBM) (mean annual rate of net soil loss 30.1 t ha^{-1} yr^{-1})	-69.3 t ha^{-1} yr^{-1} (runoff plots; long-term monitoring data)

NB: MBM = Mass Balance Model; MBM2 = Mass Balance Model 2; MBM3 = Mass Balance Model 3; DMM = Diffusion and Migration Model; PDM = Profile Distribution Model.

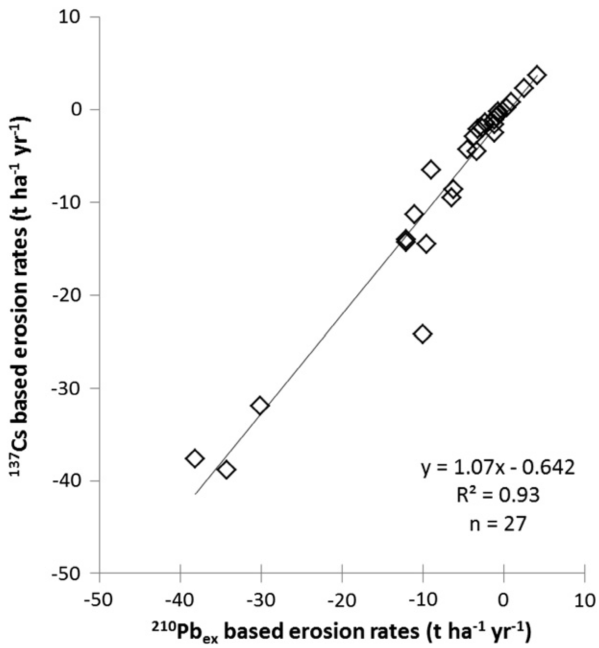

Fig. 4. Comparison of soil redistribution rates obtained with the ^{137}Cs and the ^{210}Pb$_{ex}$ methods. NB: Based on the data set presented in Table 1, each point representing the ^{137}Cs and ^{210}Pb$_{ex}$ pair information originating from the same study site.

This application of ^{210}Pb$_{ex}$ was first proposed by Goldberg (1963) to date glacier ice, and was subsequently applied to lacustrine (Krishnaswamy et al., 1971) and marine sediments (Koide et al., 1972). The development of the approach proceeded rapidly with other applications in various environments. In the meantime, the dating models used to interpret the ^{210}Pb$_{ex}$ depth distribution in sediment cores increased in variety and complexity (Robbins and Edgington, 1975; Appleby and Oldfield, 1978; Christensen, 1982; Abril et al., 1992; Caroll and Lerche, 2003). Currently, an advanced search in Scirus (http://www.scirus.com) with the keywords "sediment" and "^{210}Pb" produces more than 21,000 results which include 13,500 scientific articles. For an extensive review of such dating applications the reader is referred to Sánchez-Cabeza and Ruíz-Fernández (2012). It should be recognised that after half a century of investigations, the ^{210}Pb$_{ex}$ method has facilitated an improved understanding of a wide range of natural processes in recent sediments.

The most commonly used models for converting ^{210}Pb$_{ex}$ data from a sediment profile into a sediment chronology are: the Constant Flux and Constant Sedimentation Rate (CF–CSR) model (see Krishnaswamy et al., 1971; Appleby and Oldfield, 1978), the Constant Initial Concentration (CIC) model (see Pennington et al., 1976) and the Constant Rate of Supply (CRS) model (see Appleby and Oldfield, 1983). Descriptions of the models and the associated equations have been reported in the above references and by Appleby (2001).

The CRS model assumes that the flux of unsupported ^{210}Pb onto the sediment–water interface (SWI) is constant over time. The model works with the partial inventories below a given sediment horizon at depth z (obtained by numerical integration from the determined ^{210}Pb$_{ex}$ concentrations, the bulk density and the thickness of each sediment slice), and, by comparing them with the overall core inventory is able to generate a chronology – the ages at the limits of each sediment slice – and provide values of SAR (see Table 2). The latter can be linked to the major environmental impacts encountered by the investigated sedimentary system. Therefore, accurate estimation of the total inventory is of critical importance for the application of the CRS model (Appleby, 2001; MacKenzie et al., 2011). Sometimes the core is shorter than the equilibrium depth (the depth at which the total ^{210}Pb is in equilibrium with the supporting ^{226}Ra), or analytical uncertainties are too large. The total inventory can be estimated in these cases by using reference accumulation rates (when the older sections of the core follow an exponential decrease with mass depth) or reference dates (by using an independently dated reference level). Details on these methods can be found in Appleby (2001).

Appleby et al. (1979) used three laminated sediment cores from lakes in east Finland to provide an empirical validation of the CRS model in scenarios with varying SAR. Since then, the CRS model has become the most widely applied dating approach. It has been validated by several independent methods, although, some empirical evidence of its limitations has been provided by varved sediments (Wan et al., 1987; Reinikainen et al., 1997; Lamoureux, 1998).

The CIC model assumes that the ^{210}Pb$_{ex}$ flux co-varies with SAR and therefore that the initial concentration at the SWI remains constant over

Table 2
Summary of the main ^{210}Pb-based models for sediment radiometric dating.

Model	Assumptions[a]/notes	Analytical solutions	Some references
CRS: Constant Rate of Supply	[1], [2]/it can be formulated in terms of actual depths	$\Sigma(m) = \Sigma_0 \exp(-\lambda t)$	Appleby and Oldfield (1978) and Appleby (2001)
CIC: Constant Initial Concentration	[1]; $\phi(t)/w(t) = Cte$	$A(m) = A_0 \exp(-\lambda t)$	Robbins and Edgington (1975) and Robbins (1978)
CF–CSR: Constant flux and Constant Sedimentation Rate	[1], [2], [3]	$A(m) = A_0 \exp(-\lambda m/w)$; $t = m/w$	Krishnaswamy et al. (1971) and Smith and Walton (1980)
SIT: Sediment Isotope Tomography	[1]/it can be formulated in terms of actual depths	$A(m) = A_o^* \exp[(-B^* m + \sum_{n=1}^{N} a_n^* \sin\left(\frac{n\pi m}{m_{\max}}\right) + \sum_{n=1}^{N} b_n^* \left(1 - \cos\left(\frac{n\pi m}{m_{\max}}\right)\right)]$	Caroll and Lerche (2003)
NID–CSR: Non-Ideal-Deposition, Constant Sedimentation Rate	[1], [2], [3], fractioning of fluxes, depth distribution	$A(m) = C_1 e^{-\lambda \frac{m}{w}} + C_2 e^{-\alpha m}; C_2 = \frac{-\alpha g \phi}{\alpha w - \lambda}; C_1 = \frac{(1-g)\phi}{w} - C_2$	Abril and Gharbi (2012)
CMZ-CS: Complete Mixing Zone with constant SAR	[2], [3], $k_m = \infty, m \geq m_a$; $k_m = 0, m < m_a$	$A(m) = C = \frac{\phi}{w + \lambda m_a}, m \geq m_a$; $A(m) = C \exp[-\lambda(m - m_a)/w], m < m_a$	Robbins and Edgington (1975)
IMZ: Incomplete Mixing Zone	[2], [3]	A linear combination of solutions for CF-CS and CMZ-CS with coefficients g and $(1-g)$, being $g \in [0, 1]$	Abril et al. (1992)
CF-CS-Constant Diffusion	[2], [3], $k_m(m) = Cte$	$A(m) = \frac{\phi}{w - k_m \beta} \exp(\beta m); \beta = \frac{w - \sqrt{w^2 + 4\lambda k_m}}{2k_m}$	Robbins (1978) and Laissaoui et al. (2008)
CF-CS-depth dependent diffusion and/or translocational mixing	[2], [3], $k_m(m) = f(m)$; may include local sources and sinks	General numerical solution	Smith et al. (1986), Abril (2003a,b, 2004), Abril and Gharbi (2012) and Robbins (1986)

Inventories below the horizon at mass depth m: $\Sigma(m) = \int_m^\infty A(m')dm'$; $\Sigma_0 = \Sigma(m = 0)$.

[a] [1] Non post-depositional redistribution; [2] constant ^{210}Pb$_{ex}$ fluxes at the SWI; [3] constant SAR $A(m)$, ^{210}Pb$_{ex}$ activity concentration in solids at mass depth m.

time. This model permits estimation of the age of each sediment slice and its corresponding SAR history (see Table 2). The CIC model has proved its value in aquatic environments in which the $^{210}Pb_{ex}$ inputs are dominated by fluxes of sediment carrying $^{210}Pb_{ex}$ mobilised from the surrounding catchment, as can be the case with urbanized reservoirs (McCall et al., 1984). It is important to note that when $^{210}Pb_{ex}$ fluxes and SAR are both constant, the CRS and CIC models converge to the CF–CSR model (Table 2) and the $^{210}Pb_{ex}$ concentration follows an exponential decrease with mass depth. A good example is provided by Lima et al. (2005) in laminated sediments from the Pettaquamscutt River basin, in southern Rhode Island (USA).

As noted by Noller (2000) and Pennington et al. (1976), there are various major assumptions associated with using the $^{210}Pb_{ex}$ approach for sediment dating: (i) $^{210}Pb_{ex}$ is quickly removed from the atmosphere and freshwater and is sequestered in soils and sediments; (ii) $^{210}Pb_{ex}$ is immobile once deposited; (iii) $^{210}Pb_{ex}$ does not migrate down the sediment column; (iv) supported ^{210}Pb is in secular equilibrium with ^{226}Ra; and (v) ^{226}Ra remains constant with depth. As indicated below, there are other implicit assumptions in most of the models, but, first, a critical review of these five key assumptions is presented.

Assumption (v) allows for the direct estimation of the supported ^{210}Pb fraction from the asymptotic constant value (at depth) observed in the depth profile of total ^{210}Pb activity concentration. However, in many cases ^{226}Ra concentrations are not uniform with depth. Recent advances in gamma spectrometry allow the simultaneous measurement of ^{210}Pb and ^{226}Ra, making this assumption irrelevant. Assumption (iv) is met in most cases, if the sediment is underwater, although it has to be revisited in cores where sharp gradients of ^{226}Ra concentrations are found. Only a small fraction of the ^{222}Rn generated by the decay of ^{226}Ra reaches the pore space where it can diffuse throughout, resulting in steady-state profiles of supported ^{210}Pb slightly different from those for ^{226}Ra (Appleby, 2001). Assumption (iii) is necessary for CRS and CIC models, but is not valid in some sedimentary systems. Alternate models are needed in cases where major bioturbation processes produce complete mixing in the uppermost sediment layers (Robbins and Edgington, 1975). Examples of incomplete mixing (Abril et al., 1992), uniform diffusion (Robbins, 1978; Laissaoui et al., 2008) and translocational mixing – e.g. through a conveyor belt effect due to worm activity (Robbins, 1986; Smith et al., 1986) – have also been described (see Table 2 for model equations). In fact, this is the main restriction of the method, since any dataset with a $^{210}Pb_{ex}$ profile can be simultaneously explained by different models. Even in the case of a well-defined exponential decrease in the $^{210}Pb_{ex}$ activity profile, the profile can be explained by a CFCS model, a model with uniform diffusion, or by a model with a constant increase of SAR over time (i.e. acceleration). One of the most exciting scientific debates related to the radiometric dating of recent sediments concerned questions about sediment mixing and the acceleration of sedimentation rates (see Abril, 2003a, 2004). Thus, the flattening of the $^{210}Pb_{ex}$ profile observed in the uppermost sediment layers of several core profiles can be explained by both complete mixing (or partial mixing) or SAR acceleration. This aspect has been well-known since the beginning of radiometric dating by $^{210}Pb_{ex}$ and has generated much controversial discussion. Nevertheless, this apparent difficulty can be solved when the $^{210}Pb_{ex}$ dating method is complemented by other dating methods as discussed further below.

Assumption (i) is not entirely relevant since ^{210}Pb-based radiometric dating models operate with fluxes at the SWI instead of atmospheric deposition. Pre-depositional processes operating on atmospheric fluxes can be recognised by comparing the latter with those determined at the SWI using the ^{210}Pb-based dating method, and further insight can be obtained by studying artificial fallout radionuclides (Abril and García-León, 1994; Robbins et al., 2000; Trabelsi et al., 2012). Most of the ^{210}Pb-based radiometric dating models assume constant fluxes at the SWI (or fluxes co-varying with SAR, as in the CIC model). Nevertheless, the Sediment Isotope Tomography (SIT) model (Caroll and Lerche, 2003), through the application of Fourier analysis and under the assumption of non-post-depositional redistribution, can run with non-correlated and time-dependent fluxes and SAR. Due to the infinite number of numerical solutions, the SIT model has to be validated by several independent time-marks, more or less regularly distributed within the core.

It should be pointed out that the different radiometric dating models reflect a set of assumptions about the expected functioning of the sedimentary system (e.g. fluxes of $^{210}Pb_{ex}$, SAR and diffusion) which permit particular solutions to be found from the general and unique physical problem of mass conservation for solids and for the particle-associated tracers, in sediments that undergo accretion and compaction (Abril, 2003b).

Due to natural early compaction in sediments and core shortening during sampling operation and storage (Glew et al., 2001), actual depths (z, measured below the SWI) and sedimentation velocities, r (with physical dimensions $L\,T^{-1}$) are not meaningful magnitudes. Instead, the mass depth variable, m (the amount of dry matter per unit surface lying above any given sediment horizon at depth z) and the SAR (with physical dimensions of $M\,L^{-2}\,T^{-1}$) can be used. Thus, if

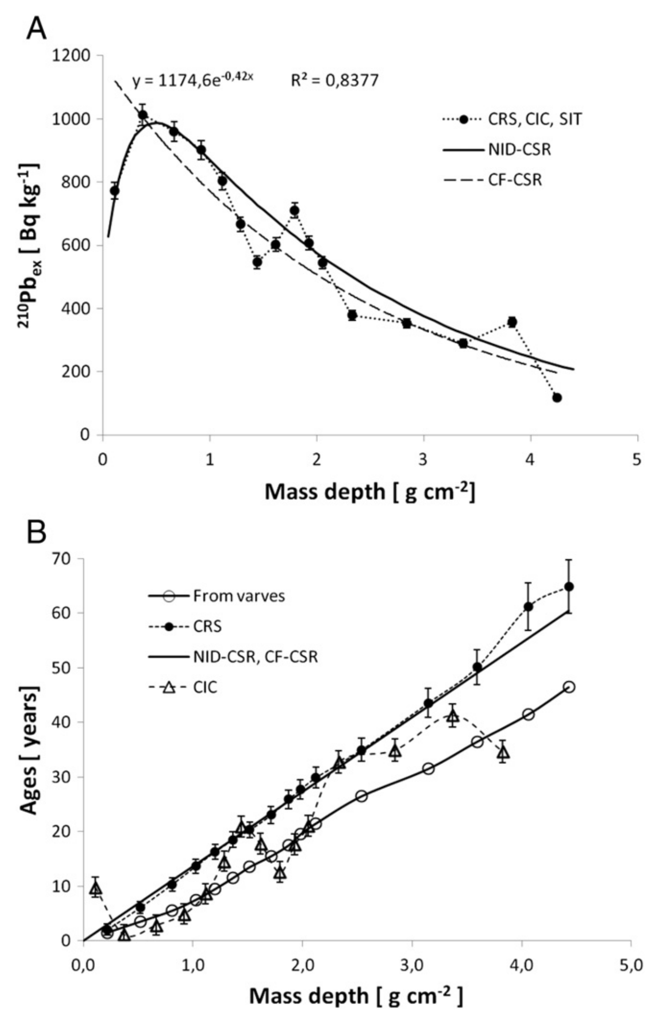

Fig. 5. Example of application of different ^{210}Pb dating models to a sediment core. (A) Measured (points with error-bars) $^{210}Pb_{ex}$ versus mass depth profile for a sediment core sampled in 1971 in Santa Barbara Basin (from Koide et al., 1973). The exponential fit corresponds to the CF–CSR model while the continuous line is the best fit obtained with the NID–CSR model (parameter values: with $\phi = 925 \pm 9$ Bq m^{-2} yr^{-1}, $w = 0.073 \pm 0.001$ g cm^{-2} yr^{-1}, $g = 0.61 \pm 0.04$ and $\alpha = 4.2 \pm 0.5$ g^{-1} cm^{2}). Models CRS and CIC are "mapping" models, contained as particular cases of the SIT model. (B) Chronologies produced with the previous models, compared against the one from varve counting (from Koide et al., 1973). Error bars correspond to 1σ propagated uncertainties.

compaction was neglected, it is possible to find analytical solutions for steady-state profiles in terms of actual depths under a wide variety of assumptions and models (see Robbins, 1978). If bulk densities are assumed to be steady-state (which is possible even under variable SAR when the conductivity of solids co-varies with the rates), the same set of analytical solutions can be reencountered, but in terms of *m*, and then, accounting for compaction (Abril, 2011). Under unsteady compaction, the governing equation for radioactive tracers should be coupled with the equation for mass conservation of solids given by the theory of early compaction (Abril, 2003b, 2011).

The diffusion term accounts for the random motion of individual particles, with a null net mass flow. When required, the mathematical description can be improved through a two-phase model (solids and pore-water) assuming local equilibrium for the solid-to-liquid partitioning of the tracer (Robbins and Jasinski, 1995). Nevertheless all these assumptions can be inappropriate to account for fast initial redistribution processes of radioactive fluxes within the pore space, particularly in sediments with high porosities. Thus, all models assume ideal deposition at the SWI, with new radioactive input being deposited above the previously existing material. However in sediments with high porosities, this assumption appears quite unrealistic as part of the incoming flux could penetrate rapidly through the connected pore spaces, which can be modelled as depth-distributed sources (see Abril and Gharbi, 2012). Such processes can explain subsurface $^{210}Pb_{ex}$ maxima observed in sediment cores, as well as a penetration of ^{137}Cs to greater depth than expected from sedimentation rates and diffusion. A similar situation was reported for an investigated floodplain, where fluxes were distributed almost exponentially with depth in the surface sediment, which could have been buried by further accretion during individual flood events (He, 1993).

A complete and rigorous mathematical treatment of different models and the analytical and numerical methods involved in their respective solutions is beyond the scope of the present review. A summary of the most common ^{210}Pb-based radiometric dating models is presented in Table 2, and, to provide an objective comparison of their advantages and limitations, a laminated sediment core sampled in 1971 in the Santa Barbara Basin, California, USA (Koide et al., 1973) was included to test the different model-chronologies against the one derived from varve counting (see Fig. 5).

The supported fraction was estimated to be 3.5 ± 0.5 dpm g^{-1} (Koide et al., 1972). Bulk densities were estimated from the reported water and organic matter contents, assuming typical densities for inorganic solids (2.5 g cm^{-3}), organic matter (1.1 g cm^{-3}) and seawater (1.03 g cm^{-3}). Discussion will be limited to those models which exclude post-depositional redistribution. In Fig. 5a, the CF–CSR model corresponds to the exponential fit (a single-mean-value of SAR can be obtained from the scaling factor in this exponential, as presented in Table 2), while the Non-Ideal-Deposition, Constant Sedimentation Rate (NID–CSR) model is the best fit to a model including a fraction of fluxes undergoing non-ideal deposition (as an exponential depth distributed source; see figure caption for model parameters). These are two examples of "curve fitting models". In the same panel, the CRS and CIC models exactly fit to the measured profile since they interpret any deviation from the ideal exponential decay as a change in SAR (CRS model) or in the flux (CIC model), in such a way that they can produce as many values of SAR/flux as data points contained in the profile. These are examples of "mapping models". The SIT model assumes that both, SAR and fluxes can vary independently with time, and it contains, as particular cases, the CRS and CIC models, and it can operate through an extensive multi-parametric fit.

Fig. 5b shows the chronologies (given as ages before the date of sampling) obtained from the previous models, along with that derived from varve counting. To apply the CRS model, it is necessary to know the total inventory, but this core was not long enough (e.g. $^{210}Pb_{ex}$ in Fig. 5a does not disappear within the core mass depth). In these cases, some corrections are needed, and the one used here involves estimating the missing part of the inventory from an extrapolation of the exponential decay pattern observed in Fig. 5a (the reference accumulation rate method). The age of each sediment slice is then computed using the analytical solution given in Table 2. CF–CSR and NID–CSR models produced virtually identical results for the mean SAR value. The corresponding plot of the ages versus mass depth is a straight line in quite good agreement with the CRS chronology. Finally, to apply the CIC model it is necessary to have a reliable estimate of the initial concentration, a difficult task in this particular profile which shows a sub-surface maxima. Without any additional constraint, this value is an ad hoc hypothesis. For our test, a value of 1050 ± 50 Bq kg^{-1} has been adopted. The corresponding chronology is then derived using the analytical solution presented in Table 2. It is worthy of note that CIC ages show fluctuations that seem to violate the stratigraphic principle, but this can be overcome by adopting a polynomical smoothing of the data. Without any independent time mark, the SIT model cannot constrain the variability in SAR and fluxes, in such a way that there exists a virtually unlimited number of possible solutions, the CIC and CRS chronologies being two particular cases.

Although error propagation and the χ^2 tests have not been discussed in detail, from this exercise it is clear that neither the high quality of the fits (as in the cases of CF–CSR and NID–CSR models) nor the consistency with other models (e.g. CRS and the two curve-fitting models) can ensure the accuracy of the derived chronologies (in this example they largely deviate from the varve chronology). Therefore, as suggested by Smith (2001), the authors of this review highly recommend confirming the reliability of model-derived ^{210}Pb dates using an independent dating technique. This is often accomplished by comparing ^{210}Pb model dates against ages determined by artificial fallout radionuclides such as ^{137}Cs (e.g. Pennington et al., 1973; Walling et al., 2002), Pu isotopes ($^{239+240}Pu$; $^{238}Pu/^{239+240}Pu$) (e.g. Wan et al., 1987; Hancock et al., 2011), ^{241}Am (e.g. Appleby et al., 1991) and ^{14}C (e.g. Massa et al., 2012).

Some attempts have been made to extend and adapt this methodology to study sedimentary processes in reservoirs (McCall et al., 1984) and depositional zones of riverine systems (Chakrapani and Subramanian, 1993; Jweda and Baskaran, 2011; Aalto and Nittrouer, 2012). Sedimentation in these aquatic environments may be highly irregular, with some episodic extreme events, and in riverine systems it may coexist with some erosion events. Thus, the usual assumptions involved in ^{210}Pb dating models are rarely met in these scenarios. Particularly, a few decades after a dam construction, neither $^{210}Pb_{ex}$ inventories in sediments nor their $^{210}Pb_{ex}$ activity versus depth profiles are in steady-state. In many cases, severe erosion leads to a fast colmation of reservoirs, with deposits up to tens of metres. Despite this intrinsic complication, some new strategies have been proposed to overcome such limitations. Thus, measurement of the thickness of sediment over the original basin floor (McCall et al., 1984) or the inter-comparison of bathymetric surveys at different dates can serve to provide chronological references and a gross-estimate of SAR in reservoirs. In riverine–estuarine systems, multi-isotopic (e.g. ^{210}Pb, ^{226}Ra, ^{137}Cs, ^{7}Be) analysis, multi-core intercomparison, multi-model testing, granulometric speciation, and normalization of $^{210}Pb_{ex}$ concentrations to some stable elements (e.g. Al), among other strategies, have been successfully applied (Álvarez-Iglesias et al., 2007; Jweda and Baskaran, 2011; Aalto and Nittrouer, 2012).

7. Assessing sedimentation rates on floodplains and wetlands

The $^{210}Pb_{ex}$ approach has been adapted to permit the investigation of other sedimentary environments, such as river floodplains, wetlands and saltmarshes. In particular, it has been used to provide a chronology of sedimentary deposits and enable sedimentation rates to be determined (e.g. Du and Walling, 2012). This has enabled researchers to investigate changes in sedimentation rates and link these to changes within the contributing catchment (such as land use) and changes in climate (e.g. Craft and Casey, 2000; Walling et al., 2003b; Aalto and Nittrouer, 2012), as well as investigating changes in sediment-associated contaminant fluxes in river basins (Owens and Walling, 2003; Wang et al., 2004). While the overall concept of using $^{210}Pb_{ex}$ to date floodplain and wetland sediments is similar to that described above for lacustrine environments, the processes responsible for incorporating $^{210}Pb_{ex}$ into the deposited sediment are likely to be more complex. In some cases, wetlands do not receive external inputs of sediment from outside the wetland; they are a closed system. In these situations, sediment accretion is mainly associated with the build-up of autochthonous organic material and $^{210}Pb_{ex}$ is derived primarily from atmospheric fallout. Thus, dating follows the approaches described above (Section 6). In the case, of wetlands receiving both autochthonous and allochthonous sediment (i.e. an open system) and in the case of accreting floodplains, $^{210}Pb_{ex}$ is derived from both direct fallout of ^{210}Pb to the sediment surface and that associated with the deposition of sediment mobilised from the upstream catchment during storm events (He and Walling, 1996a). In this situation, it is not appropriate to assume a constant rate of supply or a constant initial concentration. A number of additional factors therefore need to be taken into consideration. Whereas the annual input of direct atmospheric fallout ^{210}Pb to a lake is commonly assumed to be contained within the sediment deposited during that year, in the case of floodplain and wetland sediments, it will be distributed approximately exponentially with depth in the surface horizon of the existing floodplain or wetland sediment, which may itself be buried by further accretion during individual flood events (He and Walling, 1996a). In this case, the average sedimentation rate R (g cm^{-2} yr^{-1}) over the past ca. 100 years can be estimated by distinguishing the input of ^{210}Pb associated with atmospheric fallout and that associated with deposited sediment. Thus, in the case of floodplain environments, He and Walling (1996a) developed the CICCS (constant initial concentration and constant sedimentation rate) model and R can be determined using the following equation:

$$R = \lambda_{Pb} \frac{A_{inv} - A_{inv,At}}{C_t} \quad (2)$$

where:

λ_{Pb} decay constant of ^{210}Pb;

A_{inv} $^{210}Pb_{ex}$ inventory (mBq cm^{-2}) measured for a specific point on a floodplain;

$A_{inv,\,At}$ local $^{210}Pb_{ex}$ inventory (mBq cm^{-2}), measured by analysing soil samples collected from undisturbed land close to the floodplain above the inundation level;

C_t Initial $^{210}Pb_{ex}$ concentration (mBq g^{-1}) in catchment-derived sediment.

This approach has the advantage that only a single assessment of the total $^{210}Pb_{ex}$ inventory of a floodplain core, rather than information on the depth distribution of $^{210}Pb_{ex}$ activity is required (He and Walling, 1996a); although it is still possible to use the $^{210}Pb_{ex}$ depth profile to determine sediment chronology (e.g. Owens et al., 1999; Walling et al., 2003b; Du and Walling, 2012). In addition, since only the total inventory of the sediment core is measured, the potential effects of biological activity in causing mixing within the sediment column and thus perturbing the $^{210}Pb_{ex}$ depth distribution can be essentially ignored. The CICCS model does require the determination of $^{210}Pb_{ex}$ concentration associated with the catchment-derived deposited sediment (i.e. C_t). This can be obtained from measurements of contemporary sediment (i.e. suspended sediment or overbank deposits collected from scrapes or floodplain deposition mats) with allowance made for particle size effects. One of the main benefits of the CICCS model is that it enables many sediment cores to be analysed, and therefore enable spatial patterns of overbank sedimentation to be determined (e.g. He and Walling, 1996a).

Recently, in northern Bolivia the application of a revised geochronological methodology using a new conceptual model of ^{210}Pb input during floods (the constant initial reach clay activity, unknown sedimentation (CIRCAUS) model) has been used to investigate decadal and annual recurrence frequency of floodplain sedimentation (see Aalto and Nittrouer, 2012), and shows great potential.

In estuarine and coastal wetland systems, additional complications can be envisaged in determining accretion rates linked to (i) sediment delivery dynamics and (ii) saline pore waters. Delivery of reworked material during tidal cycles and fluctuating inputs of catchment material in response to landscape disturbance can lead to complex accretion dynamics that do not conform to standard models (e.g. Leorri et al., 2013). Furthermore, mobility of ^{137}Cs due to saline pore waters (Foster et al., 2006) can make corroboration of ^{210}Pb dates difficult in the absence of other independent stratigraphic markers.

8. Conclusion

This review summarizes and critically examines the assumptions related to the use of $^{210}Pb_{ex}$ as a tracer of soil and sediment redistribution and as a geochronometer. As highlighted, the analysis of this isotope should be undertaken with care to ensure reliable determination of $^{210}Pb_{ex}$ activity concentrations in soil and/or sediment. Accurate measurement of this isotope – including quality control and verification of analytical methodologies – still remains a challenge for the scientific community.

Although considerable progress has been made in using $^{210}Pb_{ex}$ as a tracer in terrestrial and aquatic environments under a range of different climatic conditions, it is clear that there is still a need to pursue the validation of the results obtained with $^{210}Pb_{ex}$ and to promote the approach further through combining radiotracing techniques and traditional measurement methods. Some of the assumptions related to the Pb interactions between soils/sediment and plants should undoubtedly be investigated further under controlled and/or natural conditions to clearly quantify transfer factors and adsorption/absorption of this isotope by plants in various environment. Development and improvement of the $^{210}Pb_{ex}$ technique will depend on the ability of the scientific community to address some of the research challenges outlined in this review.

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References

Aalto, R., Nittrouer, C.A., 2012. ^{210}Pb geochronology of flood events in large tropical river systems. Phil. Trans. R. Soc. A 370 (1966), 2040–2074.
Abril, J.M., 2003a. Difficulties in interpreting fast mixing in the radiometric dating of sediments using ^{210}Pb and ^{137}Cs. J. Paleolimnol. 30, 407–414.
Abril, J.M., 2003b. A new theoretical treatment of compaction and the advective–diffusive processes in sediments. A reviewed basis for radiometric dating models. J. Paleolimnol. 30, 363–370.
Abril, J.M., 2004. Constraints on the use of ^{137}Cs as a time-marker to support CRS and SIT chronologies. Environ. Pollut. 129, 31–37.
Abril, J.M., 2011. Could bulk density profiles provide information on recent sedimentation rates? J. Paleolimnol. 46, 173–186.
Abril, J.M., García-León, M., 1994. The integrated atmospheric flux effect in a radiogeochronological model. J. Environ. Radioact. 24, 65–79.
Abril, J.M., Gharbi, F., 2012. Radiometric dating of recent sediments: beyond the boundary conditions. J. Paleolimnol. 48 (2), 449–460.
Abril, J.M., García-Leon, M., García-Tenorio, R., Sánchez, C.I., El-Daoushy, F., 1992. Dating of marine sediments by an incomplete mixing model. J. Environ. Radioact. 15, 135–151.
Adriano, D.C., 1986. Trace Elements in the Terrestrial Environment. Springer-Verlag, New York.
Ahmed, A.A., Mohamed, A., Ali, A.E., Barakat, A., Abd, E.L., Hady, M., El-Hussein, A., 2004. Seasonal variations of aerosol residence time in the lower atmospheric boundary layer. J. Environ. Radioact. 77, 275–283.

Albrecht, A., 1999. Radiocesium and ^{210}Pb in sediments, soils and surface waters of a high alpine catchment: a mass balance approach relevant to radionuclide migration and storage. Aquat. Sci. 61, 1–22.

Alloway, B.J., 1995. Heavy Metals in Soils, 2nd ed. Chapman and Hall, London (286 pp.).

Al-Masri, M.S., Al-Akel, B., Nashawani, A., Amin, Y., Khalifa, K.H., Al-Ain, F., 2008. Transfer of ^{40}K, ^{238}U, ^{210}Pb, and ^{210}Po from soil to plant in various locations in south of Syria. J. Environ. Radioact. 99, 322–331.

Álvarez-Iglesias, P., Quintana, B., Rubio, B., Pérez-Arlucea, M., 2007. Sedimentation rates and trace metal input history in intertidal sediments from San Simón Bay (Ría de Vigo, NW Spain) derived from ^{210}Pb and ^{137}Cs chronology. J. Environ. Radioact. 98, 229–250.

Anagu, I., Ingwersen, J., Utermann, J., Streck, T., 2009. Estimation of heavy metal sorption in German soils using artificial neural networks. Geoderma 152, 104–112.

Appel, C., Ma, L., 2002. Concentration, pH, and surface charge effects on cadmium and lead sorption in three tropical soils. J. Environ. Qual. 31, 581–589.

Appleby, P.G., 2001. Chronostratigraphic techniques in recent sediments. In: Last, W.L., Smol, J.P. (Eds.), Tracking Environmental Change Using Lake Sediments, Basin Analysis, Coring, and Chronological Techniques, Development in Paleoenvironmental Research. Kluwer Academic Publishers, Dordrecht, p. 576.

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

Appleby, P.G., Oldfield, F., 1978. The calculation of lead-210 dates assuming a constant rate supply of unsupported ^{210}Pb to the sediment. Catena 5, 1–8.

Appleby, P.G., Oldfield, F., 1983. The assessment of ^{210}Pb from sites with varying sedimentation accumulations rates. Hydrobiology 103, 29–35.

Appleby, P.G., Oldfield, F., 1992. Application of lead-210 to sedimentation studies. In: Ivanovich, M., Harman, R.S. (Eds.), Uranium-Series Disequilibrium: Application to Earth, Marine and Environment Sciences. Clarendon Press, Oxford, pp. 731–738.

Appleby, P.G., Oldfield, F., Thompson, R., Huttunen, P., 1979. ^{210}Pb dating of annually laminated lake sediments from Finland. Nature 280, 53–55.

Appleby, P.G., Richardson, N., Nolan, P.J., 1991. ^{241}Am dating of lake sediments. Hydrobiologia 214, 35–42.

Baskaran, M., 1995. A search of seasonal variability on the depositional fluxes of ^{7}Be and ^{210}Pb. J. Geophys. Res. 100 (D2), 2833–2840.

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

Baskaran, M., 2011b. Dating of biogenic and inorganic carbonates using ^{210}Pb–^{226}Ra disequilibrium method: a review. In: Baskaran, M. (Ed.), Handbook of Environmental Isotope Geochemistry. Advances in Isotope Geochemisty, vol. 2. Springer, pp. 789–809 (Chapter 37).

Baskaran, M., Coleman, C.H., Santschi, P.H., 1993. Atmospheric depositional fluxes of ^{7}Be and ^{210}Pb at Galveston and College Station, Texas. J. Geophys. Res. 98 (D11), 20555–20571.

Beks, J.P., Eisma, D., van der Plicht, J., 1998. A record of atmospheric ^{210}Pb deposition in The Netherlands. Sci. Total Environ. 222, 35–44.

Belyaev, V.R., Markelov, M.V., Golosov, V.N., Belyaev, Y.R., Tishkina, E.V., 2004. Applying various methods for assessing soil and sediment redistribution within an intensively cultivated dry valley subcatchment. In: Golosov, V.N., Belyaev, V.R., Walling, D.E. (Eds.), Sediment Transfer Through the Fluvial System. IAHS Publication, 288, pp. 93–103.

Benmansour, M., Laissaoui, A., Benbrahim, S., Ibn Majah, M., Chafik, A., Povinec, P., 2006. Distribution of anthropogenic radionuclides in Moroccan coastal waters and sediments. Radioactivity in the Environment — Book Series, 8, pp. 145–150.

Benmansour, M., Nouira, A., Benkdad, A., Ibn Majah, M., Bouksirat, H., El Oumri, M., Mossadek, R., Duchemin, M., 2011. Estimates of long and short term soil erosion rates on farmland in semi-arid West Morocco using caesium-137, excess lead-210 and beryllium-7 measurements. Impact of Soil Conservation Measures on Erosion Control and Soil Quality. IAEA-TECDOC-1665. International Atomic Energy Agency Publication, Vienna, Austria, pp. 159–174 (http://www-pub.iaea.org/books/IAEABooks/8612/Impact-of-Soil-Conservation-Measures-on-Erosion-Control-and-Soil-Quality).

Benmansour, M., Mabit, L., Nouira, A., Moussadek, R., Bouksirate, H., Duchemin, M., Benkdad, A., 2013. Assessment of soil erosion and deposition rates in a Moroccan agricultural field using fallout ^{137}Cs and $^{210}Pb_{ex}$. J. Environ. Radioact. 115, 97–106.

Benninger, L.K., Krishnaswami, S., 1981. Sedimentary processes in the inner New York Bight: evidence from excess ^{210}Pb and $^{239+240}$Pu. Earth Planet. Sci. Lett. 53, 158–174.

Bird, G., 2011. Provenancing anthropogenic Pb within the fluvial environment: developments and challenges in the use of Pb isotopes. Environ. Int. 37, 802–819.

Blake, W.H., Wallbrink, P.J., Wilkinson, S.N., Humphreys, G.S., Doerr, S.H., Shakesby, R.A., Tomkins, K.M., 2009. Deriving hillslope sediment budgets in wildfire-affected forests using fallout radionuclide tracers. Geomorphology 104, 105–116.

Boshkova, T., Minev, L., 2001. Correction for self-attenuation in gamma-ray spectrometry of bulk samples. Appl. Radiat. Isot. 54 (5), 777–783.

Braakhekke, M.C., Wutzler, T., Reichstein, M., Kattge, J., Beer, C., Schrumpf, M., Schöning, I., Hoosbeek, M.R., Kruijt, B., Kabat, P., 2011. Modeling the vertical soil organic matter profile using $^{210}Pb_{ex}$ measurements and Bayesian inversion. Biogeosci. Discuss. 8, 7257–7312.

Braakhekke, M.C., Wutzler, T., Beer, C., Kattge, J., Schrumpf, M., Schöning, I., Hoosbeek, M. R., Kruijt, B., Kabat, P., Reichstein, M., 2012. Modeling the vertical soil organic matter profile using Bayesian parameter estimation. Biogeosci. Discuss. 9, 11239–11292.

Bradl, H.B., 2004. Adsorption of heavy metal ions on soils and soils constituents. J. Colloid Interface Sci. 277, 1–18.

Bunzl, K., Trautmannsheimer, M., 1999. Transfer of ^{238}U, ^{226}Ra and ^{210}Pb from slag contaminated soils to vegetables under field conditions. Sci. Total Environ. 231, 91–99.

Burton, W.M., Stewart, N.G., 1960. Use of long-lived natural radioactivity as an atmospheric tracer. Nature 186, 584–589.

Caillet, S., Arpagaus, P., Monna, F., Dominik, J., 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 (2), 241–256.

Caroll, J., Lerche, I., 2003. Sedimentary Processes: Quantification Using Radionuclides. Elsevier, Oxford.

Central Radioanalytical Laboratory of Food and Feed Safety Directorate, 2006. Central Radioanalytical Laboratory of Food Feed Safety Directorate — Hungarian Agricultural Authority. Radioanalytical Monitoring Network of Ministry of Agriculture and Rural Development. Annual Report 2006.

Chakrapani, G.J., Subramanian, V., 1993. Rates of erosion and sedimentation in the Mahanadi river basin, India. J. Hydrol. 149 (1–4), 39–48.

Cheng, H., Hu, Y., 2010. Lead (Pb) isotopic fingerprinting and its applications in lead pollution studies in China: a review. Environ. Pollut. 158 (5), 1134–1146.

Christensen, E.R., 1982. A model for radionuclides in sediments influenced by mixing and compaction. J. Geophys. Res. Oceans 87, 566–572.

Collins, A.L., Walling, D.E., Sichingabula, H.M., Leeks, G.J.L., 2001. Using ^{137}Cs measurements to quantify soil erosion and redistribution rates for areas under different land use in the Upper Kaleya River basin, southern Zambia. Geoderma 104, 229–323.

Craft, C.B., Casey, W.P., 2000. Sediment and nutrient accumulation in floodplain and depressional freshwater wetlands of Georgia, USA. Wetlands 20, 323–332.

Cutshall, N.H., Larsen, I.L., Olsen, C.R., 1983. Direct analysis of ^{210}Pb in sediment samples: self-absorption corrections. Nucl. Instrum. Methods Phys. Res. 206 (1–2), 309–312.

Davies, B.E., 1995. Lead, In: Alloway, B.J. (Ed.), Heavy Metals in Soils, 2nd ed., pp. 206–223 (Chapter 9).

Degryse, F., Smolders, E., Parker, D.R., 2009. Partitioning of metals (Cd, Co, Cu, Ni, Pb, Zn) in soils: concepts, methodologies, prediction and applications — a review. Eur. J. Soil Sci. 60, 590–612.

DeMaster, D.J., McKee, B.A., Nittrouer, C.A., Brewster, D.C., Biscaye, P.E., 1985. Rates of sediment reworking at the Hebble site based on measurements of Th-234, Cs-137 and Pb-210. Mar. Geol. 66, 133–148.

Dercon, G., Mabit, L., Hancock, G., Nguyen, M.L., Dornhofer, P., Bacchi, O.O.S., Benmansour, M., Bernard, C., Froehlich, W., Golosov, V.N., Haciyakupoglu, S., Hai, P.S., Klik, A., Li, Y., Lobb, D., Onda, Y., Popa, N., Rafiq, M., Ritchie, J.C., Schuller, P., Shakhashiro, A., Wallbrink, P., Walling, D.E., Zapata, F., Zhang, X., 2012. Fallout radionuclide-based techniques for assessing the impact of soil conservation measures on erosion control and soil quality: an overview of the main lessons learnt under an FAO/IAEA Coordinated Research Project. J. Environ. Radioact. 107, 78–85.

Desideri, D., Meli, M.A., Roselli, C., Feduzi, L., 2011. Alpha and gamma spectrometry for determination of natural and artificial radionuclides in tea, herbal tea and camomile marketed in Italy. Microchem. J. 98, 170–175.

Dörr, H., 1995. Application of ^{210}Pb in soils. Paleolimnology 13, 157–168.

Du, P., Walling, D.E., 2012. Using ^{210}Pb measurements to estimate sedimentation rates on river floodplains. J. Environ. Radioact. 103, 59–75.

Du, J.Z., Zhang, J., Baskaran, M., 2011. Application of short-lived radionuclides (^{7}Be, ^{210}Pb, ^{210}Po, ^{137}Cs and ^{234}Th) to trace the sources, transport pathways and deposition of particles/sediments in rivers, estuaries and coasts. In: Baskaran, M. (Ed.), Handbook of Environmental Isotope Geochemistry. Advances in Isotope Geochemisty, vol. 1. Springer, pp. 305–329 (Chapter 16).

Ebaid, Y.Y., Khater, A.E.M., 2006. Determination of ^{210}Pb in environmental samples. J. Radioanal. Nucl. Chem. 270 (3), 609–619.

Evans, L.J., 1989. Chemistry of metal retention by soils. Environ. Sci. Technol. 23, 1046–1059.

Evrard, O., Némery, J., Gratiot, N., Duvert, C., Ayrault, S., Lefèvre, I., Poulenard, J., Prat, C., Bonté, P., Esteves, M., 2010. Sediment dynamics during the rainy season in tropical highland catchments of central Mexico using fallout radionuclides. Geomorphology 124 (1–2), 42–54.

Ferraz, M.C.M.A., Lourenco, J.C.N., 2000. The influence of organic matter content of contaminated soils on the leaching rate of heavy metals. Environ. Prog. 19 (1), 53–58.

Foster, I.D.L., Mighall, T.M., Proffitt, H., Walling, D.E., Owens, P.N., 2006. Post-depositional ^{137}Cs mobility in the sediments of three shallow coastal lagoons, SW England. J. Paleolimnol. 35, 881–895.

Froehlich, W., 2004. Soil erosion, suspended sediment sources and deposition in the Maw-Ki-Syiem drainage basin, Cherrapunji, northwestern India. In: Golosov, V., Belyaev, V., Walling, D.E. (Eds.), Sediment Transfer through the Fluvial System. IAHS Publication, 288, pp. 138–146.

Fukuyama, T., Onda, Y., Takenaka, C., Walling, D.E., 2008. Investigating erosion rates within a Japanese cypress plantation using ^{137}Cs and $^{210}Pb_{ex}$ measurements. J. Geophys. Res. Earth Surf. 113. http://dx.doi.org/10.1029/2006JF000657.

Gaspar, L., Navas, A., Walling, D.E., Machín, J., Gómez Arozamena, J., 2013. Using ^{137}Cs and $^{210}Pb_{ex}$ to assess soil redistribution on slopes at different temporal scales. Catena 102, 46–54.

Glew, J.R., Smol, J.P., Last, W.M., 2001. Sediment core collection and extrusion. In: Last, W. L., Smol, J.P. (Eds.), Tracking Environmental Change Using Lake Sediments. Basin Analysis, Coring, and Chronological Techniques. Developments in Paleoenvironmental Research. Kluwer Academic Publishers, Dordrecht, pp. 171–203.

Goldberg, E.D., 1963. Geochronology with ^{210}Pb. In: IAEA (Ed.), Symposium on Radioactive Dating. IAEA, Vienna, pp. 121–131.

Golosov, V.N., Belyaev, V.R., Markelov, M.V., Ivanova, N.N., Kuznetsova, Y.U.S., 2011. Application of the radionuclide technique and other methods for assessing the effectiveness of soil conservation measures at the Novosil study site, Orel region, Central Russia. Impact of Soil Conservation Measures on Erosion Control and Soil Quality. IAEA-TECDOC-1665. International Atomic Energy Agency Publication, Vienna, Austria, pp. 131–157 (http://www-pub.iaea.org/books/IAEABooks/8612/Impact-of-Soil-Conservation-Measures-on-Erosion-Control-and-Soil-Quality).

Graham, I.J., Ditchburn, R.G., Barry, B.J., 2004. ^{210}Pb–^{137}Cs dating of glacial lake sediments. N.Z. Sci. Rev. 61 (2), 45–47.

Gustafsson, J.P., Tiberg, C., Edkymish, A., Kleja, D.B., 2011. Modelling lead (II) sorption to ferrihydrite and soil organic matter. Environ. Chem. 8 (5), 485–492.

Guzmán, G., Quinton, J.N., Nearing, M.A., Mabit, L., Gómez, J.A., 2013. Sediment tracers in water erosion studies: current approaches and challenges. J. Soils Sediments 13, 816–833.

Hancock, G.J., Revill, A.T., 2013. Erosion source discrimination in a rural Australian catchment using compound-specific isotope analysis (CSIA). Hydrol. Process. 27 (6), 923–932.

Hancock, G.J., Leslie, C., Everett, S.E., Tims, S.G., Brunskill, G.J., Haese, R., 2011. Plutonium as a chronomarker in Australian and New Zealand sediments: a comparison with ^{137}Cs. J. Environ. Radioact. 102 (10), 919–929.

He, Q., 1993. Interpretation of Fallout Radionuclide Profiles in Sediments From Lake and Floodplain Environment(Ph.D. Thesis) University of Exeter, UK.

He, Q., Owens, P.N., 1995. Determination of suspended sediment provenance using caesium-137, unsupported lead-210 and radium-226: a numerical mixing model approach. In: Foster, I.D.L., Gurnell, A.M., Webb, B.W. (Eds.), Sediment and Water Quality in River Catchments. Wiley, Chichester, pp. 207–227.

He, Q., Walling, D.E., 1996a. Use of fallout ^{210}Pb to investigate longer-term rates and patterns of overbank sediment deposition on the floodplains of lowland rivers. Earth Surf. Process. Landf. 21, 141–154.

He, Q., Walling, D.E., 1996b. Interpreting particle size effects in the adsorption of ^{137}Cs and unsupported ^{210}Pb by mineral soils and sediments. J. Environ. Radioact. 30, 117–137.

He, Q., Walling, D.E., 1997. The distribution of Fallout ^{137}Cs and ^{210}Pb in undisturbed and cultivated soils. Appl. Radiat. Isot. 48 (5), 677–690.

Hou, X., Roos, P., 2008. Critical comparison of radiometric and mass spectrometric methods for the determination of radionuclides in environmental, biological and nuclear waste samples. Anal. Chim. Acta. 608 (2), 105–139.

Huisman, N.L.H., Karthikeyan, K.G., 2012. Using radiometric tools to track sediment and phosphorus movement in an agricultural watershed. J. Hydrol. 450–451, 219–229.

Hult, M., 2007. Low-level gamma spectrometry using Ge-detectors. Metrologie S87–S94.

IAEA, 2009. Quantification of radionuclide transfer in terrestrial and freshwater environments for radiological assessments. IAEA-TECDOC 1616. International Atomic Energy Agency Publication, Vienna, Austria (616 pp. http://www-pub.iaea.org/books/IAEABooks/8103/Quantification-of-Radionuclide-Transfer-in-terrestrial-and-Freshwater-Environments-for-Radiological-Assessments).

IAEA, 2011. Analytical methodology for the determination of radium isotopes in environmental samples. Analytical Quality in Nuclear Applications Series, 19. International Atomic Energy Agency Publication, Vienna, Austria (59 pp. http://www-pub.iaea.org/MTCD/Publications/PDF/IAEA-AQ-19_web.pdf).

Imboden, D.M., Stiller, M., 1982. The influence of radon diffusion on the ^{210}Pb distribution in sediments. J. Geophys. Res. Oceans 87 (C1), 557–565.

Jia, G., Jia, J., 2012. Determination of radium isotopes in environmental samples by gamma spectrometry, liquid scintillation counting and alpha spectrometry: a review of analytical methodology. J. Environ. Radioact. 106, 98–119.

Jia, G., Torri, G., 2007. Determination of ^{210}Pb and ^{210}Po in soil or rock samples containing refractory matrices. Appl. Radiat. Isot. 65 (1), 1–8.

Jodlowski, P., 2006. Self-absorption correction in gamma-ray spectrometry of environmental samples — an overview of methods and correction values obtained for the selected geometries. Nukleonika 51, S21–S25.

Jodlowski, P., 2007. Correction factors to account for minor sample height variations in gamma-ray spectrometry. Nucl. Instrum. Methods Phys. Res. A 580, 238–241.

Jodlowski, P., Wachniew, P., Dinh, C.N., 2014. Monte Carlo validation of the self-attenuation correction determination with the Cutshall transmission method in ^{210}Pb measurements by gamma-spectrometry. Appl. Radiat. Isot. http://dx.doi.org/10.1016/j.apradiso.2013.11.062.

Jweda, J., Baskaran, M., 2011. Interconnected riverine lacustrine systems as sedimentary repositories: a case study in southeast Michigan using excess ^{210}Pb- and ^{137}Cs-based sediment accumulation and mixing models. J. Great Lakes Res. 37, 432–446.

Kabata Pendias, A., Pendia, H., 1992. Trace Elements in Soils and Plants, 2nd ed. CRC Press, Boca Raton, Fla. USA.

Kaste, J.M., Baskaran, M., 2011. Meteoric ^{7}Be and ^{10}Be as process tracers in the environment. In: Baskaran, M. (Ed.), Handbook of Environmental Isotope Geochemistry. Advances in Isotope Geochemistry, vol. 1. Springer, pp. 61–86 (Chapter 5).

Kato, H., Onda, Y., Tanaka, Y., 2010. Using ^{137}Cs and ^{210}Pb$_{ex}$ measurements to estimate soil redistribution rates on semi-arid grassland in Mongolia. Geomorphology 114, 508–519.

Khater, A.E.M., Ebaid, Y.Y., 2008. A simplified gamma-ray self-attenuation correction in bulk samples. Appl. Radiat. Isot. 66 (3), 407–413.

Klaminder, J., Yoo, K., 2008. Contaminants as tracers for studying dynamics of soil formation: mining an ocean of opportunities. In: Sparks, D.L. (Ed.), Advances in Agronomy, 100, pp. 15–57.

Klaminder, J., Bindler, R., Emteryd, O., Renberg, I., 2005. Uptakes and recycling of lead by boreal forest plants: quantitative estimates from a site in northern Sweden. Geochim. Cosmochim. Acta 69 (10), 2485–2496.

Klaminder, J., Renberg, I., Bindler, R., Appleby, P., Emteryd, O., Grip, H., 2006. Estimating the mean residence time of lead in the organic horizon of boreal forest soils using 210-lead, stable lead and a soil chronosequence. Biogeochemistry 78, 31–49.

Koide, M., Soutar, A., Goldberg, E.D., 1972. Marine Geochronology with ^{210}Pb. Earth Planet. Sci. Lett. 14, 442–446.

Koide, M., Bruland, K.W., Goldberg, E.D., 1973. ^{228}Th/^{232}Th and ^{210}Pb geochronologies in marine and lake sediments. Geochim. Cosmochim. Acta 37, 1171–1187.

Koiter, A.J., Owens, P.N., Petticrew, E.L., Lobb, D.A., 2013. The behavioural characteristics of sediment properties and their implications for sediment fingerprinting as an approach for identifying sediment sources in river basins. Earth Sci. Rev. 125, 24–42.

Komarek, M., Ettler, V., Chrastny, V., Mihaljevic, M., 2008. Lead isotopes in environmental sciences: a review. Environ. Int. 34 (4), 562–577.

Korte, N.E., Skopp, J., Fuller, W.H., Niebla, E.E., Alesii, B.A., 1976. Trace element movement in soils: influence of soil physical and chemical properties. Soil Sci. 122 (6), 350–359.

Kozak, K., Mietelski, J.W., Jasinska, M., Gaca, P., 2001. Decreasing of the natural background counting — passive and active method. Nukleonika 46 (4), 165–169.

Krishnaswamy, S.D., Lal, D., Martin, J.M., Meybeck, M., 1971. Geochronology of lake sediments. Earth Planet. Sci. Lett. 11, 407–414.

Laboratoire National Henri Becquerel, 2012. Decay Data Evaluation ProjectRecommended Data http://www.nucleide.org/DDEP_WG/DDEPdata.htm.

Laissaoui, A., Benmansour, M., Ziad, N., Majah, Ibn, Abril, J.M., Mulsow, S., 2008. Anthropogenic radionuclides in the water column and a sediment core from the Alboran Sea: application to radiometric dating and reconstruction of historical water column radionuclide concentrations. J. Paleolimnol. 40 (3), 823–833.

Lamoureux, S.F., 1998. Distinguishing Between the Geomorphic and Hydro-Meteorological Controls Recorded in Clastic Varved Sediments(Ph. D. Thesis) Department of Earth and Atmospheric Sciences. University of Alberta, Canada.

Leorri, E., Cearreta, A., García-Artola, A., Irabien, M.J., Blake, W.H., 2013. Relative sea-level rise in the Basque coast (N Spain): different environmental consequences on the coastal area. Ocean Coast. Manag. 77, 3–13.

Li, J., Li, Y., Wang, Y., Wu, J., 2010. Applicability study of using in-situ gamma-ray spectrometry technique for ^{137}Cs and ^{210}Pb$_{ex}$ inventories measurement in grassland environments. Appl. Radiat. Isot. 68 (6), 1143–1149.

Lima, A.L., Hubeny, J.B., Reddy, Ch.M., King, J.W., Hughen, K.A., Eglinton, T.I., 2005. High-resolution historical records from Pettaquamscutt River basin sediments: 1. ^{210}Pb and varve chronologies validate record of ^{137}Cs released by the Chernobyl accident. Geochim. Cosmochim. Acta 69, 1803–1812.

Mabit, L., Benmansour, M., Walling, D.E., 2008. Comparative advantages and limitations of the fallout radionuclides ^{137}Cs, ^{210}Pb$_{ex}$ and ^{7}Be for assessing soil erosion and sedimentation. J. Environ. Radioact. 99 (12), 1799–1807.

Mabit, L., Klik, A., Benmansour, M., Toloza, A., Geisler, A., Gerstmann, U.C., 2009. Assessment of erosion and deposition rates within an Austrian agricultural watershed by combining ^{137}Cs, ^{210}Pb$_{ex}$ and conventional measurements. Geoderma 150 (3–4), 231–239.

Mabit, L., Klik, A., Toloza, A., 2010. Radioisotopic measurements (^{137}Cs and ^{210}Pb) to assess erosion and sedimentation processes: case study in Austria. In: Zdruli, P., Pagliai, M., Kapur, S., Faz Cano, F. (Eds.), Land Degradation and Desertification: Assessment, Mitigation and Remediation. Springer, pp. 401–412 (Chapter 31).

Mabit, L., Meusburger, K., Fulajtar, E., Alewell, C., 2013. The usefulness of ^{137}Cs as a tracer for soil erosion assessment: a critical reply to Parsons and Foster (2011). Earth Sci. Rev. 127, 300–307.

MacKenzie, A.B., Hardie, S.M.L., Farmer, J.G., Eades, L.J., Pulford, I.D., 2011. Analytical and sampling constrains in ^{210}Pb dating. Sci. Total Environ. 409, 1298–1304.

Massa, C., Bichet, V., Gauthier, E., Perren, B.B., Mathieu, O., Petit, C., Monna, F., Giraudeau, J., Losno, R., Richard, H., 2012. A 2500 year record of natural and anthropogenic soil erosion in South Greenland. Quat. Sci. Rev. 32, 119–130.

Matisoff, G., Whiting, P.J., 2011. Measuring soil erosion rates using natural (^{7}Be, ^{210}Pb) and anthropogenic (^{137}Cs, ^{239}Pu, ^{240}Pu). In: Baskaran, M. (Ed.), Handbook of Environmental Isotope Geochemistry. Advances in Isotope Geochemisty, vol. 1. Springer, pp. 487–519 (Chapter 25).

Matisoff, G., Wilson, C.G., Whiting, P.J., 2005. The ^{7}Be/^{210}Pb$_{XS}$ ratio as an indicator of suspended sediment age or fraction new sediment in suspension. Earth Surf. Process. Landf. 30 (9), 1191–1201.

McCall, P.L., Robbins, J.A., Matisoff, G., 1984. ^{137}Cs and ^{210}Pb transport and geochronologies in urbanized reservoirs with rapidly increasing sedimentation rates. Chem. Geol. 44, 33–65.

Navas, A., Valero-Garcés, B., Gaspar, L., 2011. Radionuclides and stable elements in the sediments of Yesa Reservoir, Central Spanish Pyrenees. J. Soils Sediments 11, 1082–1098.

Noller, J.S., 2000. Lead-210 geochronology. Quaternary Geochronology: Methods and Applications. American Geophysical Union, Washington, pp. 115–120.

Nozaki, Y., 1977. The study of lead-210 in the marine environment. J. Oceanogr. Soc. Jpn. 33 (6), 342–349.

O'Farrell, C.R., Heimsath, A.M., Kaste, J.M., 2007. Quantifying hillslope erosion rates and processes for a coastal California landscape over varying timescales. Earth Surf. Process. Landf. 32, 544–560.

Ojala, A.E.K., Francus, P., Zolitschka, B., Besonen, M., Lamoureux, S.F., 2012. Characteristics of sedimentary varve chronologies — a review. Quat. Sci. Rev. 43, 45–60.

Oldfield, F., Appleby, P.G., 1984. The role of ^{210}Pb dating in sediment based erosion studies. Drainage Basin Erosion and Sedimentation — A Conference on Erosion, Transportation and Sedimentation in Australian Drainage Basins. Newcastle University, NSW, pp. 175–182.

Olley, J., Burton, J., Smolder, K., Pantus, F., Pietsch, T., 2013. The application of fallout radionuclides to determine the dominant erosion process in water supply catchments of subtropical South-east Queensland, Australia. Hydrol. Process. 27 (6), 885–895.

Owens, P.N., Walling, D.E., 2002. Changes in sediment sources and floodplain deposition rates in the catchment of the River Tweed, Scotland, over the last 100 years: the impact of climate and land use change. Earth Surf. Process. Landf. 27, 403–423.

Owens, P.N., Walling, D.E., 2003. Temporal changes in the heavy metal and phosphorus content of suspended sediment transported by Yorkshire rivers, UK over the last 100 years, as recorded by floodplain deposits. Hydrobiologia 494, 185–191.

Owens, P.N., Walling, D.E., Leeks, G.J.L., 1999. Use of floodplain sediment cores to investigate recent historical changes in overbank sedimentation rates and sediment sources in the catchment of the River Ouse, Yorkshire, UK. Catena 36, 21–47.

Owens, P.N., Blake, W.H., Giles, T.R., Williams, N.D., 2012. Determining the effects of wildfire on sediment sources using ^{137}Cs and unsupported ^{210}Pb: the role of natural landscape disturbances and driving forces. J. Soils Sediments 12, 982–994.

Özden, B., Uğur, A., Esetlili, T., Esetlili, B.C., Kurucu, Y., 2013. Assessment of the effects of physical–chemical parameters on ^{210}Po and ^{210}Pb concentrations in cultivated and uncultivated soil from different areas. Geoderma 192, 7–11.

Parsons, A.J., Foster, I.D.L., 2011. What can we learn about soil erosion from the use of ^{137}Cs? Earth Sci. Rev. 108 (1–2), 101–113.

Pennington, W., Cambray, R.S., Fisher, E.M., 1973. Observations on lake sediments using ^{137}Cs fallout as a tracer. Nature 242, 324–326.

Pennington, W., Tutin, T.C., Cambray, R.S., Eakins, J.D., Harkness, D.D., 1976. Radionuclide dating of recent sediments of Bleham Tarn. Freshw. Biol. 6, 317–331.

Perreault, L.M., Yager, E.M., Aalto, R., 2013. Application of $^{210}Pb_{ex}$ inventories to measure net hillslope erosion at burned sites. Earth Surf. Process. Landf. 38, 133–145.

Persson, B.R.R., Holm, E., 2011. Polonium-210 and lead-210 in the terrestrial environment: a historical review. J. Environ. Radioact. 102, 420–429.

Porto, P., Walling, D.E., 2012. Validating the use of ^{137}Cs and $^{210}Pb_{ex}$ measurements to estimate rates of soil loss from cultivated land in southern Italy. J. Environ. Radioact. 106, 47–57.

Porto, P., Walling, D.E., Callegari, G., Catona, F., 2006. Using fallout lead-210 measurements to estimate soil erosion in three small catchments in Southern Italy. Water Air Soil Pollut. Focus 6, 657–667.

Porto, P., Walling, D.E., Callegari, G., Capra, A., 2009. Using caesium-137 and unsupported lead-210 measurements to explore the relationship between sediment mobilisation, sediment delivery and sediment yield for a Calabrian catchment. Mar. Freshw. Res. 60, 680–689.

Porto, P., Walling, D.E., Callegari, G., 2013. Using ^{137}Cs and $^{210}Pb_{ex}$ measurements to investigate the sediment budget of a small forested catchment in southern Italy. Hydrol. Process. 27 (6), 795–806.

Porto, P., Walling, D.E., Capra, A., 2014. Using ^{137}Cs and $^{210}Pb_{ex}$ measurements and conventional surveys to investigate the relative contributions of interrill/rill and gully erosion to soil loss from a small cultivated catchment in Sicily. Soil Tillage Res. 135, 18–27.

Povinec, P.P., 2008. Low-level gamma-ray spectrometry for environmental samples. J. Radioanal. Nucl. Chem. 276 (3), 771–777.

Preiss, N., Genthon, C., 1997. Use of a new database of lead-210 for global aerosol model validation. J. Geophys. Res. 102, 25347–25357.

Preiss, N., Meliérè, M., Pourchet, M., 1996. A compilation of data on lead-210 concentration in surface air and fluxes at the air–surface and water–sediment interfaces. J. Geophys. Res. 101, 28847–28862.

Quindos, L.S., Sainz, C., Fuente, I., Nicolas, J., Quindos, L., Arteche, J., 2006. Correction by self-attenuation in gamma-ray spectrometry for environmental samples. J. Radioanal. Nucl. Chem. 270 (2), 339–343.

Ravichandran, M., Baskaran, M., Santschi, P.H., Bianchi, T.S., 1995. Geochronology of sediments in the Sabine–Neches estuary, Texas, U.S.A. Chem. Geol. 125, 291–306.

Reinikainen, P., Meriläinen, J.J., Virtanen, A., Veijola, H., Äystö, J., 1997. Accuracy of ^{210}Pb dating in two annually laminated lake sediments with high Cs background. Appl. Radiat. Isot. 48, 1009–1019.

Renberg, I., Brännvall, M.L., Bindler, R., Emteryd, O., 2002. Stable lead isotopes and lake sediments a useful combination for the study of atmospheric lead pollution history. Sci. Total Environ. 292, 45–54.

Robbins, J.A., 1978. Geochemical and geophysical applications of radioactive lead isotopes. In: Nriagu, J.O. (Ed.), Biochemistry of Lead. Elsevier, Amsterdam, pp. 285–393.

Robbins, J.A., 1986. A model for particle-selective transport of tracers in sediments with conveyer belt deposit feeders. J. Geophys. Res. Oceans 91, 8542–8558.

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

Robbins, J.A., Jasinski, A.W., 1995. Chernobyl fallout radionuclides in Lake Sniardwy, Poland. J. Environ. Radioact. 26, 157–184.

Robbins, J.A., Mudroch, A., Oliver, B.G., 1990. Transport and storage of ^{137}Cs and ^{210}Pb in sediments of Lake St. Clair. Can. J. Fish. Aquat. Sci. 47, 572–587.

Robbins, J.A., Holmes, C., Halley, R., Bothner, M., Shinn, E., Graney, J., Keeler, G., tenBrink, M., Orlandini, K.A., Rudnick, D., 2000. Time-averaged fluxes of lead and fallout radionuclides to sediments in Florida Bay. J. Geophys. Res. 105 (C12), 28805–28821.

Robu, E., Giovani, C., 2009. Gamma-ray self-attenuation corrections in environmental samples. Rom. Rep. Phys. 61 (2), 295–300.

Saïdou, Bochud, F., Laedermann, J.P., Buchillier, T., Moise, K.N., Froidevaux, P., 2007. Calibration of an HPGe detector and self-attenuation correction for ^{210}Pb: verification by alpha spectrometry of ^{210}Po in environmental samples. Nucl. Instrum. Methods Phys. Res. Sect. A 578 (3), 515–522.

Saïdou, Bochud, F., Laedermann, J.P., Kwato Njock, M.G., Froidevaux, P., 2008. A comparison of alpha and gamma spectrometry for environmental natural radioactivity surveys. Appl. Radiat. Isot. 66, 215–222.

San Miguel, E.G., Perez-Moreno, J.P., Bolivar, J.P., Garcia-Tenorio, R., Martin, J.E., 2002. ^{210}Pb determination by gamma spectrometry in voluminal samples (cylindrical geometry). Nucl. Instrum. Methods Phys. Res. Sect. A 493 (1–2), 111–120.

Sánchez-Cabeza, J.A., Ruíz-Fernández, A.C., 2012. ^{210}Pb sediment radiochronology: an integrated formulation and classification of dating models. Geochim. Cosmochim. Acta 82, 183–200.

Santos, I.R., Burnett, W.C., Godoy, J.M., 2008. Radionuclides as tracers of coastal processes in Brazil: review, synthesis, and perspectives. Braz. J. Oceanogr. 56 (2), 115–131.

Selim, H.M. (Ed.), 2011. Dynamics and Bioavailability of Heavy Metals in the Rootzone. Taylor & Francis Group, CRC Press, USA (313 pp.).

Selim, H.M. (Ed.), 2012. Competitive Sorption and Transport of Heavy Metals in Soils and Geological Media. CRC Press, USA (426 pp.).

Shakhashiro, A., Mabit, L., 2009. Results of an IAEA inter-comparison exercise to assess ^{137}Cs and total ^{210}Pb analytical performance in soil. Appl. Radiat. Isot. 67 (1), 139–146.

Simms, A.D., Woodroffe, C., Jones, B.G., Heijnis, H., Mann, R.A., Harrison, J., 2008. Use of ^{210}Pb and ^{137}Cs to simultaneously constrain ages and sources of post-dam sediments in the Cordeaux reservoir, Sydney, Australia. J. Environ. Radioact. 99 (7), 1111–1120.

Smith, J.N., 2001. Why should we believe ^{210}Pb sediment geochronologies? J. Environ. Radioact. 55, 121–123.

Smith, J.N., Walton, A., 1980. Sediment accumulation rates and geochronologies measured in the Saguenay Fjord using the Pb-210 dating method. Geochim. Cosmochim. Acta 44 (2), 225–240.

Smith, J.N., Boudreau, B.P., Noshkin, V., 1986. Plutonium and ^{210}Pb distributions in northeast Atlantic sediments: subsurface anomalies caused by non-local mixing. Earth Planet. Sci. Lett. 81 (1), 15–28.

Smith, H.G., Blake, W.H., Owens, P.N., 2013. Discriminating fine sediment sources and the application of sediment tracers in burned catchments: a review. Hydrol. Process. 27 (6), 943–958.

Stiller, M., Imboden, D.M., 1986. ^{210}Pb in Lake Kinnerett waters and sediments: residence times and fluxes. In: Sly, P.G. (Ed.), Sediment and Water Interactions. Springer, pp. 501–511 (Chapter 44).

Strawn, D.G., Sparks, D.L., 1999. The use of XAFS to distinguish between inner-and outer-sphere lead adsorption complexes on montmorillonite. J. Colloid Interface Sci. 216 (2), 257–269.

Strawn, D.G., Sparks, D.L., 2000. Effect of soil organic matter on the kinetics and mechanism of Pb(II) sorption and desorption in soil. Soil Sci. Soc. Am. J. 64, 144–156.

Tamponnet, C., 2009. Behaviour of Lead-210 in continental environment: comparison with stables isotopes of lead. Radioprotection 44 (5), 285–289.

Tangahu, B.V., Rozaimah Sheikh Abdullah, S., Basri, H., Idris, M., Anuar, N., Mukhlisin, M., 2011. A Review on heavy metals (As, Pb, and Hg) uptake by plants through phytoremediation. Int. J. Chem. Eng. http://dx.doi.org/10.1155/2011/939161 (31 pp.).

Taylor, A., Blake, W.H., Smith, H.G., Mabit, L., Keith-Roach, M.J., 2013. Assumptions and challenges in the use of fallout beryllium-7 as a soil and sediment tracer in river basins. Earth Sci. Rev. 126, 85–95.

Teramage, M.T., Onda, Y., Kato, H., Wakiyama, Y., Mizugaki, S., Hiramatsu, S., 2013. The relationship of soil organic carbon to $^{210}Pb_{ex}$ and ^{137}Cs during surface soil erosion in a hillslope forested environment. Geoderma 192, 59–67.

Trabelsi, Y., Gharbi, F., El Ghali, A., Oueslati, M., Samaali, M., Abdelli, W., Ben Tekaya, M., Benmansour, M., Mabit, L., Ben M'Barek, N., Baccouche, S., Reguigui, N., Abril, J.M., 2012. Recent sedimentation rates in Garaet El Ichkeul Lake, NW Tunisia, as affected by the construction of dams and a regulatory sluice. J. Soils Sediments 12 (5), 784–796.

Trivedi, P., Dyer, J.A., Sparks, D.L., 2003. Lead sorption onto ferrihydrite. Part 1. A macroscopic and spectroscopic assessment. Environ. Sci. Technol. 37, 908–914.

Turekian, K.Y., Nozaki, Y., Benninger, L.K., 1977. Geochemistry of atmospheric radon and radon products. Annu. Rev. Earth Planet. Sci. 5, 227–255.

Van Hoof, P.L., Andren, A.W., 1989. Partitioning and transport of ^{210}Pb in Lake Michigan. J. Great Lakes Res. 15 (3), 498–509.

Vesterbacka, P., Ikäheimonen, T.K., 2005. Optimization of ^{210}Pb determination via spontaneous deposition of ^{210}Po on a silver disk. Anal. Chim. Acta. 545 (2), 252–261.

Villa, M., Hurtado, S., Manjón, G., García-Tenorio, R., 2007. Calibration and measurements of ^{210}Pb using two independent techniques. Radiat. Meas. 42 (9), 1552–1560.

Wakiyama, Y., Onda, Y., Mizugaki, S., Asai, H., Hiramatsu, S., 2010. Soil erosion rates on forested mountain hillslopes estimated using ^{137}Cs and $^{210}Pb_{ex}$. Geoderma 159, 39–52.

Wallbrink, P.J., Murray, A.S., 1993. Use of fallout radionuclides as indicators of erosion processes. Hydrol. Process. 7, 297–304.

Wallbrink, P.J., Murray, A.S., 1996. Determining soil loss using the inventory ratio of excess lead-210 to cesium-137. Soil Sci. Soc. Am. J. 60 (4), 1201–1208.

Wallbrink, P.J., Olley, J.M., Hancock, G., 2002a. Estimating residence times for fine sediment in river channels using fallout ^{210}Pb. IAHS Publ. 276, 425–433.

Wallbrink, P.J., Walling, D.E., He, Q., 2002b. Radionuclide measurements using HpGe gamma spectrometry. In: Zapata, F. (Ed.), Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides. Kluwer, Dordrecht, pp. 67–96.

Walling, D.E., 2005. Tracing suspended sediment sources in catchments and river systems. Sci. Total Environ. 344, 159–184.

Walling, D.E., 2006. Tracing versus monitoring: new challenges and opportunities in erosion and sediment delivery research. In: Owens, P.N., Collins, A.J. (Eds.), Soil Erosion and Sediment Redistribution in River Catchments. CABI, Wallingford, pp. 13–27.

Walling, D.E., 2012. Fallout radionuclides in the study of erosion and sedimentation. In: Meyers, R.A. (Ed.), Encyclopedia of Sustainability Science and Technology. Springer.

Walling, D.E., 2013. Beryllium-7: the Cinderella of fallout radionuclide sediment tracers? Hydrol. Process. 27, 830–844.

Walling, D.E., He, Q., 1994. Rates of overbank sedimentation on the flood plains of several British rivers during the past 100 years. IAHS Publ. 224, 203–210.

Walling, D.E., He, Q., 1999a. Changing rates of overbank sedimentation on the floodplains of British rivers over the past 100 years. In: Brown, A.G., Quine, T.A. (Eds.), Fluvial Processes and Environmental Change. Wiley, Chichester, pp. 207–222.

Walling, D.E., He, Q., 1999b. Using fallout lead-210 measurements to estimate soil erosion on cultivated land. Soil Sci. Soc. Am. J. 63, 1404–1412.

Walling, D.E., He, Q., 1999c. Improved models for estimating soil erosion rates from ^{137}Cs measurements. J. Environ. Qual. 28, 61–622.

Walling, D.E., Quine, T.A., 1991. Use of ^{137}Cs measurements to investigate soil erosion on arable fields in the UK: potential applications and limitations. J. Soil Sci. 42, 147–165.

Walling, D.E., Woodward, J.C., 1992. Use of radiometric fingerprints to derive information on suspended sediment sources. Erosion and Sediment Transport Monitoring Programmes in River Basins (Proceedings of the Oslo Symposium, August 1992). IAHS Publication, 210, pp. 153–164.

Walling, D.E., Woodward, J.C., 1995. Tracing sources of suspended sediment in river basins: a case study of the River Culm, Devon, UK. Mar. Freshw. Res. 46, 327–336.

Walling, D.E., He, Q., Quine, T., 1995. Use of caesium-137 and lead-210 as tracers in soil erosion investigation. IAHS Publ. 229, 163–172.
Walling, D.E., Owens, P.N., Leeks, G.J.L., 1999. Fingerprinting suspended sediment sources in the catchment of the River Ouse, Yorkshire, UK. Hydrol. Process. 13, 955–975.
Walling, D.E., He, Q., Appleby, P.G., 2002. Conversion models. In: Zapata, F. (Ed.), Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides. Kluwer, Dordrecht, pp. 111–162.
Walling, D.E., Collins, A.L., Sichingabula, H.M., 2003a. Using unsupported lead-210 measurements to investigate soil erosion and sediment delivery in a small Zambian catchment. Geomorphology 52, 193–213.
Walling, D.E., Owens, P.N., Foster, I.D.L., Lees, J.A., 2003b. Changes in the fine sediment dynamics of the Ouse and Tweed basins in the UK over the last 100–150 years. Hydrol. Process. 17, 3245–3269.
Walling, D.E., Zhang, Y., He, Q., 2011. Models for deriving estimates of erosion and deposition rates from fallout radionuclide (caesium-137, excess lead-210, and beryllium-7) measurements and the development of user-friendly software for model implementation. Impact of Soil Conservation Measures on Erosion Control and Soil Quality. IAEA-TECDOC-1665. International Atomic Energy Agency Publication, pp. 11–33 (http://www-pub.iaea.org/books/IAEABooks/8612/Impact-of-Soil-Conservation-Measures-on-Erosion-Control-and-Soil-Quality).
Wan, G.J., Santschi, P.H., Sturm, M., Farrenkothen, K., Lueck, A., Werth, E., Schuler, C., 1987. Natural (^{210}Pb, ^{7}Be) and fallout (^{137}Cs, $^{239+240}$Pu, ^{90}Sr) radionuclides as geochemical tracers of sedimentation in Greifensee, Switzerland. Chem. Geol. 63, 181–196.
Wang, G.P., Liu, J.S., Tang, J., 2004. Historical variation of heavy metals with respect to different chemical forms in recent sediments from Xianghai Wetlands, Northeast China. Wetlands 24, 608–619.
Welp, G., 1999. Inhibitory effects of the total and water-soluble concentrations of nine different metals on the dehydrogenase activity of a loess soil. Biol. Fertil. Soils 30 (1–2), 132–139.
Welp, G., Brümmer, G.W., 1999. Adsorption and solubility of ten metals in soil samples of different composition. J. Plant Nutr. Soil Sci. 162 (2), 155–161.
Wilkinson, S.N., Wallbrink, P.J., Hancock, G.J., Blake, W.H., Shakesby, R.A., Doerr, S.H., 2009. Fallout radionuclide tracers identify a switch in sediment sources and transport-limited sediment yield following wildfire in eucalypt forest. Geomorphology 110, 140–151.
Wilson, C.G., Papanicolaou, A.N.T., Denn, K.D., 2012. Partitioning fine sediment loads in a headwater system with intensive agriculture. J. Soils Sediments 12 (6), 966–981.
Winkler, R., Rosner, G., 2000. Seasonal and long-term variation of ^{210}Pb concentration in air, atmospheric deposition rate and total deposition velocity in south Germany. Sci. Total Environ. 263, 57–68.
Wise, S.M., 1980. Caesium-137 and lead-210: a review of techniques and some applications in geomorphology. In: Cullingford, R.A., Davidson, D.A., Lewin, J. (Eds.), Timescales in Geomorphology. Wiley, New York, pp. 109–127.
Yang, Y.H., Yan, B.X., Zhu, H., 2011. Estimating soil erosion in Northeast China using ^{137}Cs and ^{210}Pb$_{ex}$. Pedosphere 21, 706–711.
Yoo, K., Ji, J.L., Aufdenkampe, A., Klaminder, J., 2011. Rates of soil mixing and associated 25 carbon fluxes in a forest versus tilled agricultural field: implications for modeling the soil carbon cycle. J. Geophys. Res. 116, G01014. http://dx.doi.org/10.1029/2010JG001304.
Zaborska, A., Caroll, J., Papucci, C., Pempkowiak, J., 2007. Intercomparison of alpha and gamma spectrometry techniques used in ^{210}Pb geochronology. J. Environ. Radioact. 93 (1), 38–50.
Zapata, F. (Ed.), 2002. Handbook for the Assessment of Soil Erosion and Sedimentation Using Environment Radionuclides. Kluwer Academic Publishers, The Netherlands (219 pp.).
Zhang, X., Qi, Y., Walling, D.E., He, X., Wen, A., Fu, J., 2006. A preliminary assessment of the potential for using ^{210}Pb$_{ex}$ measurement to estimate soil redistribution rates on cultivated slopes in the Sichuan Hilly Basin of China. Catena 68, 1–9.
Zhang, W., Ungar, K., Chen, J., St-Amant, N., Tracy, B.L., 2009. An accurate method for the determination of ^{226}Ra activity concentrations in soil. J. Radioanal. Nucl. Chem. 280 (3), 561–567.
Zheng, J.J., He, X.B., Walling, D.E., Zhang, X.B., Flanagan, D., Qi, Y.Q., 2007. Assessing soil erosion rates on manually-tilled hillslopes in the Sichuan Hilly Basin using ^{137}Cs and ^{210}Pb$_{ex}$ measurements. Pedosphere 17, 273–283.
Zupanc, V., Mabit, L., 2010. Nuclear techniques support to assess erosion and sedimentation processes: preliminary results of the use of ^{137}Cs as soil tracer in Slovenia. Dela 33, 21–36.