University "Tuscia" of Viterbo, Italy
Department of Innovation in Biological Systems, Food and Forestry (DIBAF)

Affiliation: University "Mediterranea" of Reggio Calabria, Italy
Department of Agriculture

PhD Course: Sciences, Technology and Biotechnology for Sustainability
Curriculum: Forest Ecology and Environmental Technologies

Cycle XXIX

Predicting erosion rates in three small Calabrian catchments using Caesium-137 (^{137}Cs) measurements. Theoretical approach and field measurements based on the re-sampling technique

SSD: AGR/08

PhD Thesis of
Vanessa Cogliandro

Tutor
Prof. Paolo Porto

PhD Course Coordinator
Prof. Mauro Moresi

College year 2016/2017

INDEX

Short Abstract

Extended abstract

I. Introduction

II. State-of-the-art

III. Materials and Methods

IV. Results and Discussion

V. Conclusion and Future Prospectives

References

List of papers

Short Abstract

The soil is a natural precious resource to defend because soil erosion represents an important threat to the long-term sustainability of agriculture and forestry in many areas of the world. During the last decades, a number of methods have been employed to predict soil erosion rates in different countries of the world, including southern Italy. One of the techniques that proved to be very successful in this field of research is based on the use of the radionuclide ^{137}Cs. The ^{137}Cs measurements are able to document rates of soil erosion for the past ca. 60 years since the commencement of the nuclear fallout in 1954.

The work undertaken in this thesis aims at monitoring and predicting erosion rates in three small Calabrian catchments using an improved theoretical approach, proposed to exploit the potential of the classic ^{137}Cs technique.

These catchments are equipped since 1978 for measuring rainfall, runoff and sediment yield. A number of field campaigns, undertaken during the period 1998-2001, provided the basis for a first application of the novel technique based on ^{137}Cs measurements.

The first application of this technique allowed a comparison between the predicted values of erosion rates provided by the radionuclide measurements (1954-2001) and the long-term values of measured soil erosion representative of the first monitored period (from 1978 to 2001). The results of these investigations are published elsewhere in the literature.

The activity carried out for this PhD Project and described in this dissertation is based on a second set of field campaigns, undertaken during the period from 2013 to 2015 within the same catchments. This strategy, better known as Re-sampling approach, consists in repeating the same sampling campaigns in order to recollect the soil samples, from the same sampling points, after a number of years. The new measurements of ^{137}Cs, looking back at an extended time window (1954-2015) allowed a comparison of soil erosion rates for different periods.

The main results obtained during this PhD Project demonstrated that two of the three catchments showed a significant increase in net soil loss in recent years and this was consistent with the increase shown by the annual sediment yields from the two catchments. The third catchment provided no clear evidence of a change in net soil loss and this was again consistent with the records of annual sediment yield. The overall results suggest that the recent period represents a period of changing sediment response within the study catchments. This suggestion is consistent with independent evidence provided by the available records of sediment yield from the catchments and the trend of increasing annual erosivity demonstrated by the records from two rainfall measuring stations in the vicinity of the study catchment.

The results from the study confirmed the viability of using the Re-sampling approach for ^{137}Cs measurements within areas with uncultivated soils, in order to document changing rates of soil redistribution in response to Global Change and more particularly changes in land use and precipitation erosivity.

Keywords: ^{137}Cs; re-sampling approach; soil loss; sediment yield; uncultivated soils; environmental change.

Extended Abstract

The research PhD project deals with the use of a novel technique aimed at predicting erosion rates in three small catchments (W1, W2 and W3) located in Calabria, Southern Italy. An improved theoretical method, consisting of a re-sampling approach, is proposed to exploit the potential of the classic ^{137}Cs technique. The re-sampling technique was applied to investigate erosion rates during the period 1999-2015. Comparison of the inventory documented by the initial sampling campaign (1998-2001) for each sampling with the second campaigns of sampling (2013-2015) at the same points provided the basis for estimating the mean annual soil redistribution rate for that point over the period between the two sampling campaigns (1999-2015).

In order to fulfill the proposed goals, the activity was carried out in several phases that required:

a) a review of the literature about the existing works concerning the re-sampling technique related to the use of ^{137}Cs for predicting soil erosion rates in uncultivated soils - this review includes also the use of the laboratory equipment (gamma detectors) useful to measure the radionuclide activity in soil samples;
b) the field work within the experimental catchments that included the ordinary sediment sampling to monitor the erosion processes at event scale on the three catchments (W1, W2, and W3) and two field campaigns within the catchments W1 and W3 devoted to collection of soil samples from selected points - to complete the scenario of the application of the radionuclides technique in different physical habitats, further research at University of Maryland Baltimore County was required;
c) the laboratory analyses for measuring the ^{137}Cs content in the soil samples collected within the experimental sites;
d) the processing of the new datasets obtained during the field campaigns to calculate the erosion rates at event, annual, and mean annual scale - to cover some of the gaps existing in the local database of soil erosion measurements available at the experimental sites, the application of a rainfall-runoff model was necessary at the Warsaw University of Life Sciences; therefore, it was possible to refine the procedures of reading and interpretation of rainfall and hydrometric data;
e) the comparison of the results with existing database in the area to set up a first experimental protocol for the application of this methodology;
f) a final calculation in order to estimate the mean annual soil redistribution rate for the three catchments in the recent time window between the two field campaigns.

Three experimental catchments (W1, W2 and W3) located near Crotone (39°09'10''N, 17°08'10''E) in the ephemeral headwaters of the larger Crepacuore basin were used for this purpose. These catchments, in view of their large existing databases in terms of rainfall, runoff and sediment yield measurements, represent a unique example of experimental site to test this new technique. They are equipped since 1978 for measuring rainfall, runoff and sediment yield. Each catchment has been instrumented by the National Research Council of Italy (CNR) "Soil Conservation Project" to monitor the effects of afforestation on their hydrological response and sediment yield. Runoff from the catchments is measured using H-flume structures equipped with mechanical stage recorders. The sediment load passing the gauging structure is measured using a Coshocton wheel sampler installed below the H-flume. This sampler collects an aliquot of ca. 1/200th of the flow that is diverted to a tank. After each storm event, representative samples are collected from the tank to determine the mean sediment concentration in the tank and the sediment yield is calculated as the product of this concentration and the total runoff volume for the event measured by the H-flume. The annual sediment yield is in turn calculated as the sum of the sediment loads for all storm events occurring during the year.

These three catchments (W1, W2 and W3) have never been cultivated and they originally supported a rangeland vegetation cover. W1 catchment still supports a rangeland vegetation cover dominated by *Lygeum spartum* and *Atriplex halimus*. In 1968, W2 and W3 were planted with *Eucalyptus occidentalis* trees. These trees were cut in 1978 and 1990 in catchment W2 and in 1986, 2006 and 2016 in catchment W3 with the tree cover being subsequently restored by natural re-growth. Within catchment W2, the tree cover is not uniform, and about 20% of its area, primarily along its northern margin, is characterized by discontinuous trees and a sparse grass cover. Elsewhere, the tree cover is relatively uniform. In catchment W3 the forest cover is currently almost continuous and only about 2-3% of the area is characterized by a grass cover.

The fieldwork related to this PhD programme consists of two types of activity: the ordinary sediment sampling campaigns and the soil sampling campaigns.

The ordinary sediment sampling campaigns was carried out within the experimental sites (W1, W2 and W3) and consisted of several field activities, most of them devoted to the measurements of rainfall, runoff and sediment yield after each natural storm event. These measurements concern collection of sediment samples, after each storm event, from the tank located at the catchments outlet. These measurements allowed to establish the mean sediment concentration in the tank at the event scale. The sediment concentrations associated with these samples are determined by oven drying at 105°C. The event sediment yield from the catchments is then calculated as the product of the mean sediment concentration and the total runoff volume for the event measured by the H-flume. These measurements, together with the existing dataset available for the previous monitoring period (1978-2015), served for validating the re-sampling technique proposed in this project.

The soil sampling campaigns consist of field activities directed to collect soil samples within the catchments in order to apply the re-sampling technique. A first application of the re-sampling technique was carried out in catchment W2 in 2013. The activity undertaken during this PhD programme related the catchments W1 and W3. In this respect, a first field campaign was undertaken in September 2014 and the second one in July 2015. These field activities concerned the collection of soil samples in 68 sites within the catchment W1 and in 83 sites within the W3. Soil sampling was carried out using a steel core tube (0.1 m diameter) driven into the ground by a motorized percussion corer to a depth of 30 cm. These samples were compared with those collected during a field campaign undertaken in 2001 for the catchment W1 and in 1998 for the W3, based on the same sampling locations.

An adjacent area of undisturbed rangeland with minimal slope and similar altitude was selected as the reference site. In this case, one set of samples was collected using a scraper plate, in order to provide samples from a larger and therefore more representative area.

All soil samples were initially oven dried at 105°C for 48 hours, disaggregated and passed through a 2 mm sieve. A representative sample of this fraction of each sample was then placed in a 0.5 kg Marinelli beaker or a plastic pot (in the case of smaller samplers) in order to measure its ^{137}Cs activity by gamma spectrometry. This set of samples was analyzed using 2 high resolution HPGe detectors in the Laboratory of the Department of Agriculture at the University "Mediterranea" of Reggio Calabria, Italy. Count times were ca. 40,000 s providing a precision of ca. ±10% at the 95% level of confidence. The efficiency of the detection system was calibrated using standard samples prepared by adding known amounts of radionuclide standards to representative soil samples.

The analysis consisted of the complete reading of the spectrum, identifying peaks (identified by the detector through the gamma rays emission produced by the caesium inside the sample) related to 660 keV, and in the calculation of these areas underneath. The information provided by the above analysis was used to calculate the ^{137}Cs content of the sample. This was expressed in two ways:

a) *^{137}Cs activity*, the concentration of ^{137}Cs per unit mass (expressed as Bq kg^{-1});
b) *^{137}Cs inventory*, the total amount of ^{137}Cs present in the entire soil profile per unit surface area (usually expressed as Bq m^{-2}).

For the catchment W1, the calculation of the mean annual sediment yield from the study catchments for the two periods for which sediment yield data are available gives 11.6 t ha^{-1} yr^{-1} for the period 1978-1994 and 35.9 t ha^{-1} yr^{-1} for the period 2009-2014. For the catchment W2, the mean annual sediment yield for the period 1978-1994 is estimated to be 17.5 t ha^{-1} yr^{-1} and for the period 2006-2012 is 31.02 t ha^{-1} yr^{-1}. For catchment W3, the corresponding values are 7.4 t ha^{-1} yr^{-1} for the periods 1978-1994 and 37.5 t ha^{-1} yr^{-1} for the period 2006-2015.

In this study, a diffusion and migration model has been used for the purpose of estimating the soil redistribution rates indicated by the ^{137}Cs inventories measured at the sampling sites. For the first sampling campaigns (1998-2001), the estimates of soil redistribution rate relate to periods from 1954 to 1998-2001 - where 1954 is assumed to represent the onset of bomb fallout. For the second sampling campaigns, these estimates relate to periods from 1954 to 2013-2015. By subtracting the total soil loss for the period 1954 to 2013-2015 from the total soil loss for the period 1954 to 1998-2001, it was possible to estimate the total soil loss, and thus the mean annual soil loss, for the period 1998-2001 to 2013-2015.

For the first sampling, the estimated mean annual soil redistribution rates for the catchment W1 was of -8.6 t ha^{-1} yr^{-1}, for the catchment W2 it was of -14.0 t ha^{-1} yr^{-1} and for the catchment W3 it was of -11.3 t ha^{-1} yr^{-1} (negative values indicate erosion). For the second sampling the estimates of mean annual soil redistribution rates for the catchment W1 is of -9.4 t ha^{-1} yr^{-1}, for the catchment W2 it is -13.6 t ha^{-1} yr^{-1} and for the catchment W3 it is -12.4 t ha^{-1} yr^{-1}. By calculating the total soil loss for the period 1954-2001 as the product of the soil redistribution rate and the period involved in years, and subtracting this from the total soil loss for the period 1954-2015, it is possible to estimate the total soil loss and thus the mean annual soil loss for the period 2001-2015. This latter value was estimated to be -12.3 t ha^{-1} yr^{-1} for the catchment W1, -12.6 t ha^{-1} yr^{-1} for the catchment W2 and -16.4 t ha^{-1} yr^{-1} for the catchment W3.

The overall results suggest that the recent period represents a period of changing sediment response within the study catchments W1 and W3. For the W2 catchment, the estimates provided by the ^{137}Cs measurements do not show significant difference between the two periods. The higher magnitude of the values of sediment yield could reflect an additional contribution to the sediment output from the study catchment from channel erosion, which is not included in the estimates of mean annual soil loss from the catchment slopes provided by the ^{137}Cs measurements. This apparent contrast is more likely to reflect the uncertainties associated with the two different approaches to characterizing sediment output from the study catchment, which have been captured by the uncertainty attributed to the estimates.

The results from the study demonstrate the viability of using a re-sampling approach for ^{137}Cs measurements within areas with uncultivated soils, in order to provide information on changing rates of soil redistribution in response to Global Change and more particularly changes in land use and precipitation erosivity.

INTRODUCTION

1.1 Overview

Soil erosion and associated sedimentation are natural landscape forming processes. However, they can be accelerated by human activities (deforestation, overgrazing, urban sprawl, change in land use and non-sustainable farming practices) that can have negative impacts on agricultural production as well as watershed management and conservation of natural resources (Lal, 2000; Walling, 2001).

Erosion can be caused by geological factors and by exogenous processes, like water, wind and ice. In particular, the water erosion is the process of disintegration of the soil surface that occurs as a result of rainfall and runoff.

About the erosion phenomenology, these processes consist in the detachment of soil particles, their entrainment, the transport and the deposition of these particles termed sediments. The erosion process begins when the tangential forces are exerted by the impact of raindrops on the soil surface and the current generated by water runoff. This phenomenon depends on the amount of these efforts, the resistance to separation of the particles from the soil mass, the transportability of the sediment and the transport capacity of the agent erosive (Bagarello and Ferro, 2006).

In order to understand perfectly the dynamics of this phenomenon, it is important to know the water erosion factors: the climate, the soil, the morphology, the covering and the land use. These main factors influencing soil erosion can be grouped into three categories:

a) *Energy factors* - These are related to the capacity of several agents such as rainfall, runoff, wind, ice, gravity and also mechanical tools (machinery) to generate erosion. In this regard, erosivity is the potential aggressivity of the causal agent. Landscape morphological features (e.g. slope gradient, aspect, form and length) are intrinsically linked to these energy factors (Mabit *et al*., 2014a). Water erosion has been identified as one of the major causes of soil degradation in both developed and developing countries (FAO, 2005; Mabit *et al*., 2000, 2002). On any given slope, surface runoff characteristics, and hence water erosivity, are closely related to rainfall event characteristics (e.g. rainfall intensity, event duration, raindrop size), antecedent conditions and the hydrological properties of the soil surface and shallow subsurface (Morgan, 2005). Water erosion occurs as laminar or sheet erosion, rill or interrill erosion and gully erosion. Changes in vegetation and soil hydrological conditions can also produce superficial mass movements on sloping lands and lands with low soil stability (Torri, 2004). Wind velocity and evaporation are the main factors controlling spatial patterns and rates of wind erosion. Wind selectively mobilizes and transports soil particles to nearby areas via processes of rolling and saltation as well as to remote locations as dry deposition/dust carried in suspension (Goosens, 2002).

b) *Resistance factors* - These are related to the soil erodibility or the susceptibility of the soil to be eroded. Soil erodibility is directly dependent on the physico-mechanical properties of the soil (e.g. degree of aggregate stability) and indirectly dependent on other properties (linked to chemical, physico-chemical, and biological factors) that affect the former (Mabit *et al.*, 2014a).

c) *Protection factors* - In practice, several types of erosion can occur in a given location according to the local land use and management practices (Torri, 2004) but there are also a range of factors that can reduce erosivity. Interception of rainfall is a major factor in this regard and good cover of plant biomass (vegetation or crop residues) will protect the soil from water and wind and the erosion losses will accordingly be reduced. In contrast, removal of vegetation by deforestation and overgrazing will result in enhanced erosion and deposition off-site. The loss/removal of scarce natural vegetation in arid zones leaves the soil prone to wind and water erosion and promotes desertification. In many parts of the developing world, resource-poor farmers who practice subsistence agriculture on communal lands do not have sufficient economic and technical resources to permit adequate soil management and conservation, and they cannot make investments to improve soil quality. It is therefore necessary to consider the importance of socio-economic and political factors that are drivers of the degradation of soil resources by (a) (b) erosion. The availability of resources and the economic incentives provided by relevant bodies and associated agricultural policies also exert a very important influence on land use and its intensification. These factors can have a great influence on soil erosion (Stocking, 2003; Boardman *et al.*, 2003).

Soil erosion results not only in a loss of surface soil and thus a reduction in soil depth and water holding capacity, but it can also cause changes in soil properties, due to selective mobilisation and redistribution of soil constituents as well as loss of topsoil. Changes in soil properties can result in reduced crop productivity and a deterioration in soil quality, which may impact on ecosystem functioning and the longer term sustainability of the soil resource (Karlen et al., 1997). Accelerated erosion decreases soil productivity (on-site impacts), increases downstream sedimentation, and is related to environmental pollution problems (off-farm impacts) in agroecosystems (Pimentel *et al.*, 1994; Schmitter *et al.*, 2010). On-site and off-site impacts of erosion can be summarised as follows:

a) *On-site effects* - Soil erosion affects farmers particularly through soil (including colloidal particles), nutrient and organic matter losses with consequent decline in soil fertility/productivity and crop yield reduction. These problems may occur as a consequence of two major processes. First, continual removal of surface soil by erosion leads to soil degradation reflected in the reduction of soil fertility and breakdown of soil structure (Elwell, 1984; Mabit & Bernard, 1998). Compaction and poor internal drainage can act to exacerbate this problem by reducing the infiltration capacity of the soil and enhancing surface runoff. Secondly, extensive rills and gullying may lead to a reduction of the cultivable areas. The impact of this process has been widely recognised and various estimates of its severity have been established (Whitlow, 1986; Valentini *et al.*, 2005). In the long-term, soil redistribution leads to spatial variability in soil quality, i.e. depletion or removal of soil particles and constituents, such as nutrients and organic matter, in eroding areas and enrichment in depositional areas (Mabit & Bernard, 1998; Mabit *et al.*, 2008; Pansak *et al.*, 2008).

b) *Off-site effects* - Sediment and associated potential pollutants (mainly nutrients, pesticides, and pesticide metabolites) can reach watercourses and contribute to the eutrophication and contamination of water resources affecting aquatic ecosystems and wetlands. They can also increase or create siltation problems and other major off-site impacts such as muddy floods and mudflows which can damage human infrastructure and habitation. In both developed and developing countries, the problem of sedimentation in water conveyance systems and reservoirs increases costs of water treatment and management of dams. Land degradation by soil erosion and deposition also has wider negative ecological and socioeconomic impacts. These are directly related to food insecurity and increased malnutrition leading often to higher levels of poverty, rural migration, social unrest and overall poor economic development (Pimentel, 2006; UNEP, 2007).

Therefore, soil erosion and sedimentation cause not only on-site degradation of a non-renewable natural resource, but also off-site problems such as downstream sediment deposition in fields, floodplains and water stream (Zapata *et al.*, 2003). These problems and concern over the degradation of the landscape by erosion, and their impact on soil fertility and crop productivity in agriculture land, water pollution, and sedimentation in lakes, reservoirs, and floodplains are well documented (Brown and Wolf, 1984; UNEP, 1992; Lal, 1994; Walling, 1989, 2000).

In summary, soil erosion is one of the most serious environmental problems in the world today because it seriously threatens agriculture and the natural environment. Adequate food supplies depend on productive land. Soil erosion throughout the world is proceeding largely uncontrolled, with minimal conservation. This is in part because the amount of soil that erodes with each rain or windstorm is almost imperceptible. For instance, 15 tonnes of soil lost from a hectare of land during a single storm removes only about 1 mm of soil from the surface. Such soil losses are measurable by a number of standard methods, but quantitative monitoring is almost a non-existent feature of development programs (Pimental *et al.*, 1993).

1.2 Introduction

Recent concern for problems of accelerated soil erosion and associated land degradation in many areas of the world (Pimentel, 1993) and recognition of the many adverse off-site impacts of soil erosion (Clark *et al.,* 1985) have emphasized the need for improved information on rates of soil loss and thus for reliable means of assessing soil erosion rates across a range of environments (Porto *et al*., 2001). In the case of erosion, attention focused largely on soil erosion on agricultural land and emphasized the assessment of rates of soil loss and their implications for crop productivity and the sustainability of land use practices, as well as the design of soil conservation measures. In general, the degree of attention given to the study of erosion and sediment transport was broadly proportional to the magnitude of erosion rates and sediment loads. (Walling *et al.*, 2011). Current concerns about both on-site and off-site adverse effects associated with accelerated soil loss, generate an urgent need for obtaining reliable quantitative data on the extent and actual rates of soil erosion worldwide. Such data are required for a more comprehensive assessment of the magnitude of erosion effects, to obtain a better understanding of the processes and the main controlling factors, validate new soil erosion/sedimentation prediction models, and provide a basis for developing scientifically sound land use policies and selecting effective soil conservation measures and land management strategies, including assessment of their economic and environmental impacts (Toy *et al.*, 2002; Walling, 2001).

For Southern Italy, Sorriso-Valvo *et al.* (1995) suggested that soil erosion rates as high as 10 mm year^{-1} (ca. 100-150 t ha^{-1} year^{-1}) can occur in the mountainous Calabrian region. In such contexts, there has been a drive to reduce soil erosion by implementing improved management practices and erosion control measures. In some agricultural areas, where soil erosion is particularly severe, land use change and afforestation have been seen as the most appropriate means of reducing erosion risk. Some uncertainty does, however, exist regarding the effectiveness of afforestation programmes in reducing soil loss and catchment sediment yields, including the most appropriate forest species, particularly when the climate and soil conditions mean that the resulting forest cover may be sparse and not well-developed. Eucalyptus trees have, for example, been widely planted, but the canopy cover provided by such trees is generally less dense than that associated with other tree species. Furthermore, the need to obtain some economic return from the afforested land means that forest harvesting, frequently involving clearcutting, is an essential feature of the changed land management (Porto *et al.*, 2009). To examine and thus validate the effectiveness of afforestation in reducing soil erosion and catchment sediment yields, a number of catchment experiments were initiated in southern Italy, near Crotone in Calabria, from 1978 within the framework of the National Research Council of Italy (CNR) 'Soil Conservation Project'.

Traditional soil erosion monitoring is dealt with using three different ways: direct erosion measurements (erosion plots, experimental catchments), erosion modelling and prediction methods (using mathematical and empirical models) and indirect measurements (using conventional markers as tracers to estimate soil translocation). The selection of the method to be used basically depends on the objectives of the study and the availability of resources (Elliot *et al.*, 1991).

The quest for alternative techniques of soil erosion assessment to complement existing methods and to meet new requirements has directed attention to the use of radionuclides, as tracers for documenting rates and spatial patterns of soil redistribution within the landscape (Ritchie & McHenry, 1990; Walling & Quine, 1995). The use of fallout radionuclides to obtain information on soil and sediment redistribution rates within a catchment is founded on the existence of a number of natural and manmade radionuclides that reach the land surface as fallout, primarily as wet fallout in association with rainfall, and are rapidly and strongly fixed by the surface soil or sediment. The subsequent redistribution of these radionuclides within a catchment or river system is a direct reflection of the movement of the soil or sediment particles to which the radionuclides are attached. By studying the post-fallout redistribution and fate of the selected fallout radionuclide, it is possible to obtain information on soil and sediment redistribution and, therefore, on erosion and deposition rates. The fallout radionuclide most widely used for this purpose is cesium-137 (^{137}Cs) (Ritchie JC and Ritchie CA, 2008).

The ^{137}Cs technique has been used worldwide in soil erosion studies and is considered one of the best technique available to assess medium-long term average soil erosion rates for disturbed points in a field (Loughran, 1989; Quine, 1999; Walling *et al.*, 2003; IAEA, 2006). This technique only provides the estimates of total soil erosion. In order to better control soil erosion, it is also important to assess the relative contributions of each erosion process toward the total soil erosion and the variation of their contributions across the landscape. To do so, the ^{137}Cs technique has to be used in conjunction with erosion models (e.g., Van Oost, *et al.*, 2003; Li *et al.*, 2008; Tiessen *et al.*, 2009). Traditionally, the use of the ^{137}Cs technique involves a comparison of ^{137}Cs inventory measured at the study site to that measured at a reference site, ideally an adjacent non-eroded site vegetated with perennial grass or low herb (Pennock and Appleby, 2002).

Further recent application of fallout radionuclide measurements in investigations of soil redistribution rates directed attention to permit documenting of changing erosion rates. In this PhD dissertation, a *Re-sampling* approach was also used. Using this novel approach, it is possible to compare the estimates of soil redistribution rate derived for the locations sampled in a first sampling period with a new recent time window. The period between the two sampling campaigns needs to be sufficiently long to make sure that there is a change in inventory (due to the soil redistribution and the radioactive decay) and to exceeds the uncertainty that can be ascribed to the precision of the laboratory measurements and sampling variability. One first activity in using the re-sampling approach employed by Porto *et al.* (2014) in this study area, located near Crotone, but others many Authors employed this novel technique worldwide (see §2.7).

State-of-the-art

2.1 Introduction to radionuclides

In the investigations of soil erosion and sedimentation, work has focused on the use of a particular group of environmental radionuclides, namely fallout radionuclides (FRNs), which include artificial radionuclides such as ^{137}Cs originated from thermonuclear weapon tests in the 1950s-1960s, and geogenic radioisotopes such as ^{210}Pb and also more recently cosmogenic radioisotopes such as ^{7}Be. They have been used worldwide to obtain rates and patterns of soil erosion and deposition at several temporal and spatial scales (Mabit *et al.*, 2008; Zapata *et al.*, 2003; Zapata & Nguyen, 2009). The need for improved management of erosion- and sediment related problems has necessarily directed attention to the development of an alternative technique for assessing erosion rates and documenting soil and sediment redistribution in the landscape. The use of fallout radionuclides, including caesium-137 (^{137}Cs), excess lead-210 (^{210}Pb$_{ex}$), and beryllium-7 (^{7}Be), has attracted much interest. It provides an essentially unique means of assembling retrospective, spatially distributed information on rates of soil loss and soil and sediment redistribution within the landscape over different timescales, without the need for expensive long-term monitoring (Zapata *et al.*, 2003; Walling, 2006, 2012; Mabit *et al.*, 2008; Zapata & Nguyen, 2010; Matisoff & Whiting, 2011). Most studies employing fallout radionuclides in soil erosion and sediment redistribution investigations have been based on the use of ^{137}Cs (half-life 30.17 yr) and ^{137}Cs measurements have now been successfully used in soil erosion and sediment budget investigation worldwide (Ritchie and Ritchie, 2005; Mabit *et al.*, 2013).

The International Atomic Energy Agency (IAEA) has played a key role in these methodological developments and applications. The Soil and Water Management and Crop Nutrition (SWMCN) Sub-programme of the Joint FAO/IAEA Division of Nuclear Techniques in Food and Agriculture has strengthened national capacities for using these nuclear-based techniques and has disseminated them through international co-operation in research, training and other outreach activities in FAO and IAEA Member States. In this way, both UN organizations meet their mandates and contribute to their commitment towards achieving the UN Millennium Development Goals of Extreme Poverty and Hunger Eradication and Environmental Sustainability (United Nations, 2009).

Early applications of the ^{137}Cs technique in soil erosion research were made by several groups in Australia (Loughran *et al.*, 1988; Campbell *et al.*, 1982; Elliot *et al.*, 1990; Wallbrink & Murray, 1993), Canada (De Jong *et al.*, 1983; Kachanoski, 1993; Pennock *et al.*, 1995), England

(Walling & Quine, 1991; Walling & Bradley, 1988; Walling & He 1999a and 1999b), and the USA (Ritchie & McHenry, 1973, 1975 and 1990). These pioneering groups provided a solid foundation for the application of the ^{137}Cs technique to measure soil redistribution in agricultural and natural landscapes. One important lesson learned from these early studies was that there is a need for harmonized protocols for a worldwide and consistent application of this technique.

The development, refinement and calibration of the ^{137}Cs technique have provided a universal tool to quantify soil redistribution rates in a range of natural and agro-ecosystems, and have paved the way for a wider application of the technique, particularly in assessing the effectiveness of soil conservation technologies in controlling or mitigating soil erosion and associated degradation processes (Mabit *et al.*, 2014a).

2.2 General information and basis of the nuclear technique

An atom consists of a central nucleus (proton and neutron), which is relatively heavy and carries a positive electrical charge (proton), surrounded by a cloud of low mass, negatively-charged electrons. A complete atom is electrically neutral, which means that the number of electrons or their negative charge are equal to the amount of (opposite) positive charge on a proton in the nucleus, and there is no overall charge on the neutral atom.

The nucleus of an atom is made of particles collectively named nucleons. There are two types of nucleons: protons and neutrons. They have practically equal masses but protons carry a positive electrical charge and neutrons possess no charge. The number of proton plus 11 neutrons is referred to as the mass number (A). The number of protons (Z) determines the charge of the nucleus and is referred to as the atomic number. The neutron number (N) is defined as A minus Z. Isotopes of a particular element are defined as containing the same number of protons but may have different numbers of neutrons. Thus, isotopes with different numbers of neutrons in their nuclei but the same number of protons have similar chemical properties.

Most of the approximately one thousand known isotopes are stable and do not disintegrate with time. The presence of neutrons keeps the proton together and so stabilizes the nucleus. However, when the ratio of neutrons to protons is outside a particular value, which varies with each isotope, the nucleus becomes unstable and spontaneously emits particles and/or electromagnetic radiation. This phenomenon, which characterizes radionuclides, is called radioactive decay and is a statistical process in which the decay rate is proportional to the number of radioactive nuclei of a particular type present at any time (t) and is usually accompanied by the emission of charged particles and/or gamma rays.

Three main types of emitted radiation exist: alpha, beta and gamma. Since most commonly used radionuclides in soil erosion studies are gamma-emitters, we describe in this study only gamma radiation (γ). Alpha or beta decay processes may leave the product nucleus either in its ground state or more frequently in an excited state. A nucleus in an excited state may give up its excitation energy and return to ground state in a variety of ways. The most commonly used radionuclides in soil erosion studies are gamma-emitters. These gamma radiations are photons or quanta packages of energy transmitted in the form of wave motion with energies typically from several keV to several MeV. They are distinguished from only by the fact that they come from the nucleus. Most gamma rays are somewhat higher in energy than x-rays and are, therefore, very penetrating. Gamma radiation can travel very large distances and lose its energy by interacting with atomic electrons. Depending on their energies, they can be stopped by a thin piece of aluminium foil, or can penetrate through several centimeters of lead.

The SI unit of activity is the Becquerel (Bq), which is defined as one nuclear disintegration per second. Before the introduction of SI units, the most commonly used radioactivity unit was the Curie (Ci) corresponding to 3.7 x 1010 nuclear disintegrations per second (Mabit *et al.*, 2014a).

2.2.1 ^{137}Cs origin and characteristics

Caesium (Cs) is the fifty-fifth atomic element of Mendeleev's periodic table. It is the most electropositive and the most alkaline element among alkali metals. Its atomic mass is variable from 125 to 145. Only the stable isotope of caesium, ^{133}Cs, is natural. The most well-known radioactive isotopes of caesium are ^{134}Cs ($t_{1/2} = 2.06$ years) and ^{137}Cs ($t_{1/2} = 30.17$ years).

There are no natural sources of ^{137}Cs. Indeed, it is an artificial or 'man-made' radionuclide generated as a product of nuclear fission. ^{137}Cs is a component of the ^{137}I decay chain as outlined in Equation (2.1). It emits a strong gamma-ray (662.66 KeV) making its measurement in environmental samples using gamma detector facilities relatively easy without the need for special chemical preparation or separation (Ritchie & McHenry, 1990).

$$^{137}\mathrm{I} \rightarrow {}^{137}\mathrm{Xe} \rightarrow {}^{137}\mathrm{Cs} \rightarrow {}^{137\,m}\mathrm{Ba} \rightarrow {}^{137}\mathrm{Ba} \tag{2.1}$$

The main sources of ^{137}Cs present in the environment are (i) atmospheric nuclear weapons tests carried during the period from the late 1950s to the 1970s, (ii) releases from the Chernobyl nuclear power plant accident in 1986 and (iii) local releases from nuclear reactor and waste reprocessing plants (Mabit *et al.*, 2014b).

Bomb-derived ^{137}Cs was injected into the stratosphere where it circulated globally before reaching the soil surface as fallout (Carter & Moghissi 1977). The fallout was associated primarily with precipitation, but locally, around the nuclear test sites, dry fallout was also significant (Ritchie & McHenry, 1990). The global deposition of ^{137}Cs fallout extended from the mid-1950s until the 1970s, with maximum fallout occurring in 1963 in the northern hemisphere and slightly later in the southern hemisphere. After moratoriums on testing and the Test Ban Treaty signed in 1963, global radioactive fallout rates decreased steadily, except for minor periods in 1971 and 1974 caused by nuclear testing by non-treaty countries.

2.3 Fallout radionuclides in soils

Fallout radionuclides have been widely used as environmental tracers in studying sediment and associated erosion, transport and deposition processes in drainage basins. More particularly they have been used for evaluating soil erosion rates by estimating the residence times of radionuclides in top soils (e.g. de Jong *et al*, 1986; Loughran *et al.*, 1987; Walling & Quine, 1990,1991,1992; Ritche & McHenry, 1991).

This approach is able to overcome several of the limitations associated with more traditional methods. Of particular importance is its potential to provide retrospective information on medium-term average rates of soil redistribution on the basis of a single site visit and representative distributed data for fields and larger areas, without the need to disturb the system by installing measuring equipment. For some applications, particularly those requiring spatially distributed information on soil redistribution rates, fallout radionuclides arguably provide an essentially unique means of assembling data that cannot be obtained using alternative approaches (Porto & Walling, 2012). ^{137}Cs measurements are primarily used to generate information on mean annual erosion rates over the past ca. 50 years. They have been use data range of scales, extending from studies of individual fields (Garcia-Oliva *et al*., 1995; Porto & Walling, 2012), through studies of small and intermediate-sized catchments (Golosov *et al.,* 2008; Mabit *et al*., 2007; Navas *et al*., 2005; Porto *et al*., 2001, 2003) to national scale soil erosion surveys (Walling & Zhang, 2010).

The application of the ^{137}Cs technique is based on several assumptions:

1. ^{137}Cs was deposited as fallout in association with precipitation, during and shortly after the period of thermonuclear weapons testing (i.e. mid-1950s to mid-1970s);
2. Fallout ^{137}Cs reaching the ground surface was rapidly and strongly adsorbed by soil and sediment particles with the result that subsequent redistribution of ^{137}Cs reflects the movement of these particles;
3. By assuming that the initial spatial distribution of ^{137}Cs fallout inputs was uniform, any deviations in the measured ^{137}Cs content of soil and sediment samples from the local

fallout inventory represent the net impact of soil redistribution during the period since fallout occurred;

4. Estimates of soil erosion and sedimentation can be derived from establishing relationships between ^{137}Cs loss or gain and soil erosion or deposition.

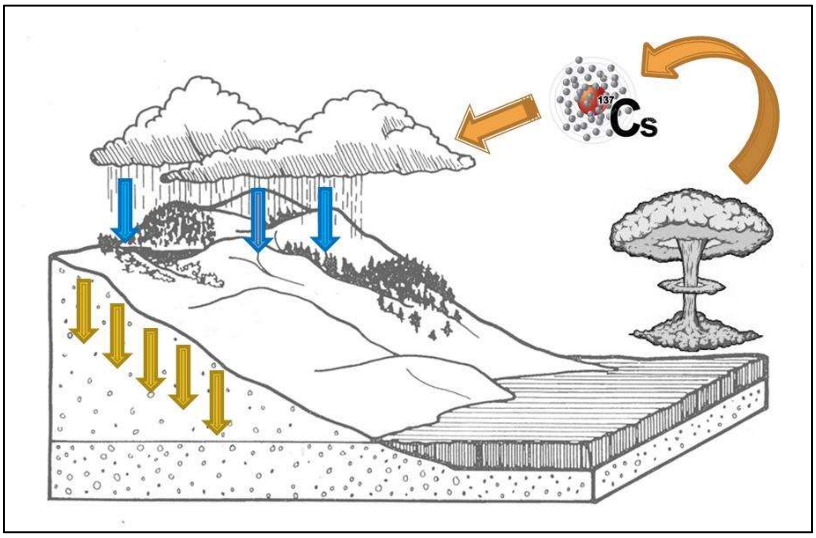


Figure 2.1. Example of the basis of the ^{137}Cs approach

The artificial radionuclide ^{137}Cs (half-life = 30.2 year) was introduced into the environment by the atmospheric testing of thermonuclear weapons during a period extending from the mid-1950s to the mid-1970s. After release into the stratosphere, ^{137}Cs was distributed globally and deposited as fallout, usually in association with precipitation. The temporal variation in fallout closely resembled the temporal variation of ^{137}Cs concentrations in the atmosphere, which in turn reflected the frequency of the bomb tests. Because most of the weapons tests were located in the USA and the former USSR, the fallout of ^{137}Cs was higher in the northern than the southern hemisphere. At the regional scale, fallout data indicate that there is a close relationship between ^{137}Cs concentrations and total annual precipitation (Cawse & Horrill, 1986). Although little empirical data exists concerning potential local variations in the ^{137}Cs fallout from weapons testing, a central assumption of the approach is that there is uniform total deposition at the local scale. This assumption is based upon the principle that although precipitation varies locally during individual storms, the net effect of the superposition of this rainfall over several years is in an essentially uniform local pattern of total precipitation and therefore of total fallout. An additional input of ^{137}Cs was provided by the accident at the Chernobyl nuclear power plant on 26 April 1986. This released ^{137}Cs as a debris cloud at ground level, with the result that most of the ^{137}Cs deposition occurred as wet fallout in association with precipitation above this cloud. Most of this ^{137}Cs is found in the northern hemisphere, with only very small quantities occurring outside of Europe and the former USSR (Pourchet *et al.*, 1988). In contrast to the fallout resulting from weapons testing, the ^{137}Cs derived from Chernobyl exhibits marked local spatial variation, reflecting the fact that the Chernobyl plume did not reach the stratosphere and also that the subsequent fallout was short-lived and associated with a small number of rainfall events (Walling & Collins, 2000).

2.3.1 Behaviour of ^{137}Cs in soil

For the use of ^{137}Cs as a soil erosion tracer it is important that ^{137}Cs has a limited mobility in soil. After its deposition at the soil surface it is rapidly and strongly adsorbed on the cation exchange sites of fine soil particles (clay and organic particles) and can thus be considered as being essentially non-exchangeable (Ritchie & McHenry, 1990; Davis, 1963; Eyman & Kevern, 1975; Lomenick & Tamura 1965). The ^{137}Cs intercepted by the plant canopy can be transferred to the soil via wash-off and the biological uptake from soils by vegetation can be considered negligible (Dahlman et al., 1975). If adsorbed by the vegetation, the ^{137}Cs is released to soils when the vegetation dies and decays (Davis, 1963; Dahlman *et al.*, 1975; Rogowski & Tamura, 1970a and 1970b). The rapid and effective adsorption of ^{137}Cs by soil and sediment particles is supported by empirical evidence from numerous laboratory and field investigations (Livens and Baxter, 1988; Livens and Rimmer, 1988; He and Walling, 1996). Strong adsorption is often associated with clay particles (Livens and Loveland, 1988), but has also been observed for a range of different mineral soils (Livens and Baxter, 1988). The likelihood of strong adsorption is further supported by the low rates of vertical migration of ^{137}Cs observed for many soil types in both laboratory and field experiments (Squire and Middleton, 1966; Frissel and Pennders, 1983).

The vertical distribution or profile of ^{137}Cs in soils reflects a range of physical, physicochemical and biological processes operating in the soil system (He & Walling, 1997). Due to the operation of various physical, chemical and biological processes in the soil system, fallout radionuclides can be redistributed in soil profiles. The processes involved are as follows (He, 1993):

- *The physical processes*: In the soil solution, soil particles, and therefore the associated fallout radionuclides, can be redistributed by physical processes, such as the movement of soil particles by the flow of pore water or the diffusion of soil particles in the pore water. Both the external conditions, such as the rainfall intensity, and the soil characteristics, such as the soil physical properties can influence such processes;

- *The physic-chemical processes*: If ion exchange is responsible for the adsorption of fallout Cs-137 and Pb-210 by soil particles and the reaction is reversible, then both Cs-137 and Pb-210 can be replaced by other ions from soil particles and re-enter the soil solution. Fallout radionuclides in solution can move in the soil solution due to molecular diffusion. Radionuclides released from one site in the soil can be re-adsorbed at another site. Pore water can move fallout radionuclides in solution released from soil particles further down in the soil profile. These processes are affected by the soil chemical properties as well as physical properties.
- *Biological processes*: Bioturbation associated with vertical mixing by soil fauna can also redistribute fallout radionuclides by redistributing soil particles containing fallout radionuclides in the soil profiles. The intensity of these processes is controlled by the biological activity within the soil system.

The importance of physico-chemical processes is higher in some extreme environments, where the biological activity is low and climatic or geological factors play a key role. In most undisturbed soils, physico-chemical processes and bioperturbation are the major factors responsible for the redistribution of fallout ^{137}Cs in the soil profile, while in cultivated soils, the redistribution of ^{137}Cs is primarily the result of mechanical mixing associated with cultivation. Lateral redistribution of ^{137}Cs in soils by biological and chemical processes is insignificant in comparison with the movement of ^{137}Cs by physical processes i.e. erosion and transport by water and wind (Ritchie & McHenry, 1990). For these reasons, and since ^{137}Cs fallout was relatively uniformly distributed across the landscape and strongly adsorbed by soil particles, it can be used as a tracer for studying the physical processes of erosion and sedimentation. The spatial variability of ^{137}Cs inventories can provide quantitative information on rates and patterns of erosion and sedimentation.

A fundamental assumption of the ^{137}Cs approach is that after the initial adsorption of ^{137}Cs by the mineral fraction, all subsequent vertical and lateral redistribution occurs in association with the erosion, transport and deposition of soil and sediment particles. This assumption has been supported by numerous studies (Evans & Dekker, 1966; Rogowski & Tamura, 1970; Ritchie & McHenry, 1990; Walling & Quine, 1993).

2.4 The application of the ^{137}Cs method

The application of the ^{137}Cs technique for determining rates and patterns of soil loss is based on several assumptions. The key assumptions and requirements of the ^{137}Cs technique have been fully described in many publications (Ritchie & McHenry, 1990; Walling & Quine, 1991, 1993; De Jong *et al.*, 1983; Loughran *et al.,* 1988). Essentially, the use of ^{137}Cs measurements in the assessment of catchment erosion and sedimentation is based upon three main acceptances:

- the distribution of the total atmospheric fallout of ^{137}Cs is uniform;
- fallout ^{137}Cs is rapidly and strongly adsorbed by soil and sediment particles upon reaching the ground surface;
- subsequent redistribution of ^{137}Cs occurs in association with soil erosion and sedimentation.

The basic principle of the method is simple, but there are some methodological issues which require careful consideration and some pitfalls which can present serious obstacles for the successful use of this method, if the user is not aware of them. The representativeness and accuracy of the results obtained, depends heavily on the correct implementation of the ^{137}Cs method (Mabit *et al.*, 2014b).

The key feature of ^{137}Cs behavior in the soil, on which the method is founded, is the strong bonding of ^{137}Cs to soil particles and its chemical stability in soil environments. This makes it possible to make the fundamental assumption that ^{137}Cs can move only together with soil particles to which it is bound and that the major processes causing its redistribution in the landscape are key mechanical processes such as water, wind and tillage

The redistribution assessment is commonly based on the ^{137}Cs inventory (total radionuclide activity per unit area) measured at a given sampling site compared with a reference site (inventory representing the cumulative atmospheric fallout input at the site, taking due account of the different behaviour of cultivated and non-cultivated soils). Because direct long-term measurements of atmospheric fallout are rarely available, the cumulative input or reference inventory is usually established by sampling adjacent stable and nearly undisturbed sites, where neither significant erosion nor additional deposition have occurred (Fig. 2.2).

The determination of ^{137}Cs inventories requires soil cores to be taken according to a carefully planned field sampling design. Measurement of ^{137}Cs concentration vs. depth is required for cores representative of each landscape unit within the study site. It is essential to determine the inventory of ^{137}Cs at a reference site (i.e. a location undisturbed by erosion or sedimentation) near to the study area (Mabit *et al.*, 2008). Sampling sites can be characterized as erosional or depositional by comparing the ^{137}Cs inventory of the given site with that of the reference site. Inventories lower than the reference value represent soil losses by erosion and those in excess of the reference level indicate deposition/accumulation of soil (Walling *et al.*, 2002).

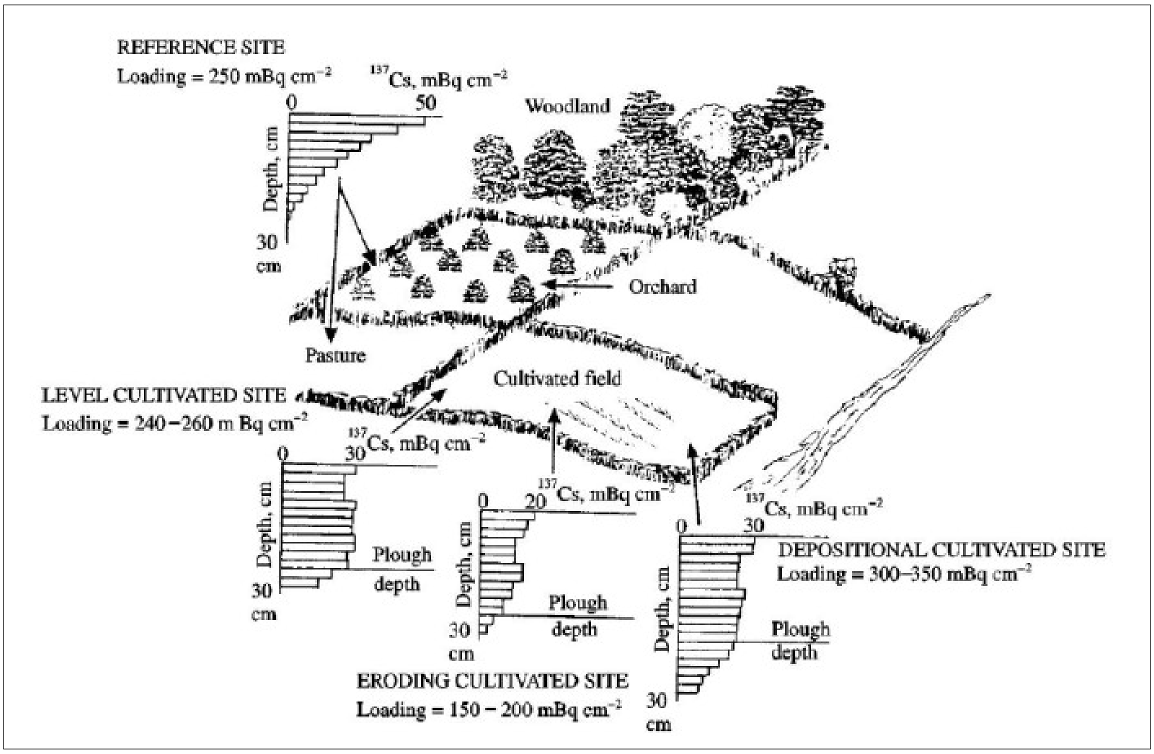


Fig 2.2 - Diagram illustrating the application of the ^{137}Cs method to study soil erosion and deposition within a landscape (Walling & Quine, 1990).

To derive quantitative estimates of soil erosion and deposition rates from ^{137}Cs measurements, it is necessary to use conversion models that are developed on the basis of physical processes influencing the relationship between magnitude of the reduction or increase in the ^{137}Cs inventory and the soil redistribution (Walling *et al.*, 2002; Walling & He, 1999a). The soil redistribution data obtained (soil erosion and sedimentation rates and patterns) represent an integrated measurement of all effects leading to soil redistribution occurring during the period extending from the main phase of atmospheric fallout input to the time of sampling. When using several FRNs, soil redistribution data over different time scales can be obtained using a single sampling campaign, thereby avoiding time-consuming, costly installations and procedures commonly required to monitor study sites over extended periods. The ^{137}Cs method is the most widely used and validated for medium-term (50 years) soil erosion assessment. The use of ^{210}Pb$_{ex}$ has been extended from the previous sediment dating application to provide a method for longer term (100 years) soil and sediment redistribution studies (Walling & He, 1999b). Recent developments of the ^{7}Be application permit short-term (days to months) soil erosion assessment in connection to individual rainfall events and seasonal changes in land use/management practices (Blake et al., 1999; Sepulveda *et al.*, 2008; Walling *et al.*, 2009).

2.5 Description of the conversion models

Use of environmental radionuclide measurements to estimate rates of erosion and deposition is founded on comparison of the inventories at individual sampling points with a reference inventory, representing the local fallout input and thus the inventory to be expected at a site experiencing neither erosion nor deposition. A measured inventory for an individual sampling point less than the reference value is indicative of erosion, whereas an inventory greater than the reference value is indicative of deposition. Although such comparisons of measured inventories with the local reference value provide useful qualitative information on the spatial distribution of erosion and deposition in the landscape and on the relative magnitude of the values involved, in most instances quantitative estimates of erosion and deposition rates are required. The derivation of quantitative estimates is heavily dependent upon the existence of a reliable means of converting the magnitude of the measured inventory at a specific sampling point, relative to the local reference inventory, to an estimate of the rate of erosion or deposition at that point.

Many different approaches have been used to convert ^{137}Cs measurements to quantitative estimates of erosion and deposition rates (Walling & He, 1999(b), 2000). These methods include both empirical relationships, and theoretical models and accounting procedures. The models varied in complexity from the simple proportional model to more complex mass balance models and models which attempt to describe the key processes controlling the distribution of ^{137}Cs in the soil profile. Models applicable to both cultivated and undisturbed (e.g. rangeland and permanent pasture) soils were included.

It is also possible to adapt some of conversion procedures and models used for ^{137}Cs to ^{210}Pb$_{ex}$ and ^{7}Be, provided key contrasts with ^{137}Cs are take into account. It has been shown that together these three radionuclides are able to provide information on soil redistribution over temporal scales ranging from a few days (^{7}Be), through decades (^{137}Cs) to around 100 years (^{210}Pb$_{ex}$). Furthermore, use of the individual radionuclides in combination offers potential to identify temporal trends in soil erosion and sedimentation rates and to elucidate the erosional history of a study site.

Each model has its specific set of parameters although some of these parameters are common between models. It is important to recognize that the individual models are different in their underlying assumptions, processes descriptions and representation of temporal variation. A sound understanding of the models and their parameters is an essential precursor to their applications. In order to avoid the possible misuse of the models, these issues will be addressed in the following sections.

A brief description of the theoretical basis of the models and some guidelines regarding the advantages and limitations of the individual models are presented. The first three models described below are for application to cultivated soils and the last two for use with uncultivated soils (rangelands or permanent pasture) (Walling *et al.*, 2006).

2.5.1 *The proportional model*

The proportional model is probably the simplest of the conversion models used for ^{137}Cs measurements (De Jong *et al.*, 1983; Fredericks & Perrens, 1988; Martz & de Jong, 1987; Walling & Quine, 1990; Di Stefano *et al.*, 1999). It is based on the premise that ^{137}Cs fallout inputs are completely mixed within the plough or cultivation layer and the simple assumption that the depth of soil removed by erosion is directly proportional to the fraction of the original ^{137}Cs inventory removed from the soil profile by erosion since the beginning of ^{137}Cs fallout or the onset of cultivation, whichever is later. Thus, if half of the ^{137}Cs input has been removed, the total soil loss over the period is assumed to be 50% of the plough depth. Division of this depth by the number of years elapsed since the beginning of significant ^{137}Cs fallout provides an estimate of the annual rate of surface lowering. Inclusion of a value for the bulk density of the soil makes it possible to calculate an erosion rate in units of mass per unit time (i.e. t ha^{-1} yr^{-1}). A particle size correction factor is often incorporated into the model to make allowance for the size selectivity of sediment mobilization and the preferential association of ^{137}Cs with the finer fractions of the soil. The model can be represented as follows:

$$Y = 10\,\frac{B\;d\;X}{100\;T\;P} \tag{2.2}$$

where:

Y is the mean annual soil loss (t ha^{-1} yr^{-1});

d is the depth of plough or cultivation layer (m);

B is the bulk density of soil (kg m^{-3});

X is the percentage reduction in total ^{137}Cs inventory (defined as $[(A_{ref}-A)/A_{ref}]\times 100$);

A_{ref} is the local ^{137}Cs reference inventory (Bq m^{-2});

A is the measured total ^{137}Cs inventory at the sampling point (Bq m^{-2});

T is the time elapsed since the initiation of ^{137}Cs accumulation or the commencement of cultivation whichever is later (yr);

P is the particle size correction factor for erosion (see below).

In cases where the ^{137}Cs inventory A for a sampling point is greater than the local reference inventory A_{ref}, deposition of sediment may be assumed and the annual deposition rate Y' (t ha^{-1} yr^{-1}) may be estimated using the following equation:

$$Y' = 10 \frac{B\ d\ X'}{100\ T\ P'} \tag{2.3}$$

where:

X' is the percentage increase in total ^{137}Cs inventory (defined as $[(A-A_{ref})/A_{ref}] \times 100$);

P' is the particle size correction factor for deposition.

The particle size correction factors P and P' are introduced to take account of the potential particle size selectivity of erosion and deposition processes. If erosion results in selective removal of fines, the eroded sediment is likely to be enriched in ^{137}Cs, since in most environments ^{137}Cs is preferentially associated with the fine fraction. Failure to take account of this in the conversion model will result in overestimation of erosion rates, because the reduction in the inventory will reflect the loss of a smaller mass of soil, by virtue of its increased ^{137}Cs content. Equally, deposition will frequently involve the deposition of coarser particles, which is likely to be characterized by a reduced ^{137}Cs content, when compared with the eroded soil. If this is not taken into account, deposition rates could be under estimated. P represents the ratio of the ^{137}Cs concentration in the sediment mobilized by erosion to that in the original soil and its value is generally 1.0 or greater. Similarly, P' represents the ratio of the ^{137}Cs concentration in deposited sediment to that of the mobilized sediment and its value is commonly less than 1.0.

Advantages and limitations: The proportional model requires very little information to provide an estimate of the erosion or deposition rate at the sampling point. In addition to the values of ^{137}Cs inventory for the sampling points and the local reference inventory, only an estimate of the plough depth and a value for the bulk density of the soil are required. It is therefore easy to apply. However, the assumptions of this model are a considerable oversimplification of reality in terms of the accumulation of ^{137}Cs in the soil. The accumulation of ^{137}Cs takes place over several years and some of the fallout input will remain at the soil surface prior to incorporation into the soil profile by cultivation. If some of the ^{137}Cs accumulated on the surface is removed by erosion prior to incorporation into the profile the estimates of soil loss provided by the model will overestimate actual rates of soil loss. Perhaps more importantly, however, this model does not take into account the fact that the depth of the plough layer is maintained by incorporating soil from below the original plough depth as the surface is lowered by erosion. This in turn causes a progressive dilution of ^{137}Cs concentrations in the soil within the plough layer. As a result, the model is likely to underestimate the actual rates of soil loss. Similarly, if the model is used to estimate deposition rates, the values obtained will underestimate the actual deposition rates, since no account is taken of the progressive reduction in the ^{137}Cs content of the deposited soil as erosion proceeds upslope. In view of these fundamental limitations, use of the proportional model is not generally recommended. It is included in the software package to permit comparison of the results obtained with those provided by other more reliable models (IAEA, 2011).

2.5.2 *A simplified mass balance model*

Mass balance models attempt to overcome some of the limitations of the simple proportional model by taking account of both inputs and losses of ^{137}Cs from the profile over the period since the onset of ^{137}Cs fallout and the progressive lowering of the base of the plough layer in response to removal of soil from the soil surface by erosion (Zhang *et al.*, 1990) proposed a simplified mass balance model, which assumes that the total ^{137}Cs fallout occurred in 1963, instead of over the longer period extending from the mid-1950s to the mid-1970s. In its original form this simplified mass balance model did not take account of particle size effects but a correction factor P. The erosion rate Y (t ha^{-1} yr^{-1}) can be calculated as:

$$Y = \frac{10\,d\,B}{P}\left[1-\left(1-\frac{X}{100}\right)^{\frac{1}{t-1963}}\right] \tag{2.4}$$

where:

d is the depth of the plough or cultivation layer (m);

B is the soil bulk density (kg m^{-3});

X is the percentage reduction in total ^{137}Cs inventory (defined as $[(A_{ref}-A)/A_{ref}]\times 100$);

P is the particle size correction factor.

For a depositional site ($A(t) > A_{ref}$), assuming a constant deposition rate R' (kg m^{-2} yr^{-1}) at the site, the sediment deposition rate can be estimated from the ^{137}Cs concentration of the deposited sediment $C_d(t')$ (Bq kg^{-1}) according to:

$$R' = \frac{A_{ex}(t)}{\int_{1963}^{t} C_d\,(t')\,e^{-\lambda(t-t')}dt'} \tag{2.5}$$

where:

$A_{ex}(t)$ is the excess ^{137}Cs inventory of the sampling point over the reference inventory at year t (the measured inventory less the local reference inventory) (Bq m^{-2});

$C_d(t')$ is the ^{137}Cs concentration of deposited sediment at year t' (Bq kg^{-1});

λ is the decay constant for ^{137}Cs (yr^{-1});

P' is the particle size correction factor.

Generally, the ^{137}Cs concentration $C_d(t')$ of deposited sediment can be assumed to be represented by the weighted mean of the ^{137}Cs concentration of sediment mobilized from the upslope contributing area. $C_d(t')$ and can therefore be calculated as:

$$C_d(t') = \frac{1}{\int_S R\,dS} \int_S P'\, C_e(t')\, R\, dS \tag{2.6}$$

where:

S is the upslope contributing area (m^2);

$C_e(t')$ is the ^{137}Cs concentration of sediment mobilized from an eroding point (Bq kg^{-1}), which can be calculated according to:

$$C_e(t') = \frac{P}{d} A_{ref}\, e^{\lambda(t-t')} \left(1 - P\frac{R}{d}\right)^{t'-1963} \tag{2.7}$$

where:

$A_{ref}(t)$ is A_{ref} in year t.

Advantages and limitations: The simplified mass balance model takes into account the progressive reduction in the ^{137}Cs concentration of the soil within the plough layer due to the incorporation of soil containing negligible ^{137}Cs from below the original plough depth and thus represents an improvement over the proportional model. This model is also easy to use and requires only information on plough depth. However, this model does not take into account the possible removal of freshly deposited ^{137}Cs fallout before its incorporation into the plough layer by cultivation, which may occur during rainfall events which produce surface runoff and therefore erosion. The assumption that the total ^{137}Cs fallout input occurs in 1963 is also an oversimplification (IAEA, 2011). Since the model requires information on the ^{137}Cs concentration of soil eroded from upslope, the sampling points used with the model must be arranged along a downslope transect. Normally, a uniform spacing of sampling points is assumed. The first (upper) point on the transect should represent an eroding point (Walling *et al*., 2014).

2.5.3 *Mass balance model*

A more comprehensive mass balance model requires consideration of the time-variant fallout ^{137}Cs input and the fate of the freshly deposited fallout before its incorporation into the plough layer by cultivation. For an eroding point ($A(t)<A_{ref}$), the change in the total ^{137}Cs inventory $A(t)$ with time can be represented as (Walling & He, 1997):

$$A(t) = A(t_0)\, e^{-\left(\frac{PR}{d}+\lambda\right)(t-t_0)} + \int_{t_0}^{t} \left[1 - P\gamma\left(1 - e^{-\frac{R}{H}}\right)\right] I(t')\, e^{-\left(\frac{PR}{d}+\lambda\right)(t-t')}\, dt' \tag{2.8}$$

where:

t_0 represents the year when cultivation started (yr);
γ is the proportion of the annual ^{137}Cs input susceptible to removal by erosion;
H is the relaxation mass depth of the initial distribution of fallout ^{137}Cs in the soil profile (kg m^{-2}).
$A(t_0)$ (Bq m^{-2}) is the ^{137}Cs inventory at t_0 (yr):

$$A(t_0) = \int_{1954}^{t_0} I(t')\, e^{-\lambda(t'-t_0)}\, dt' \tag{2.9}$$

where:

$I(t')$ is the annual ^{137}Cs deposition flux (Bq m^{-2} yr^{-1}).

The erosion rate R can be estimated by solving Equation 2.7 numerically, when the ^{137}Cs deposition flux and values of the relevant parameters are known. The ^{137}Cs concentration of mobilized sediment $C_e(t')$ can be expressed as:

$$C_e(t') = \frac{I(t')}{R} P\gamma \left(1 - e^{-R/H}\right) + P\frac{A(t')}{d} \tag{2.10}$$

For a depositional point ($A(t) > A_{ref}$), the mean soil deposition rate R' can be calculated from the following Equation:

$$R' = \frac{A - A_{ref}}{\int_{t_0}^{t} C_d\, (t')\, e^{-\lambda(t-t')} dt'} \tag{2.11}$$

where:

$C_d(t')$ (Bq kg^{-1}) essentially comprises two components, the first of which is associated with the removal of the freshly deposited ^{137}Cs, and the second is associated with erosion of the accumulated ^{137}Cs stored in the soil. Again, it can be estimated from the ^{137}Cs concentrations of the mobilized sediment from the upslope eroding area S; $C_d(t')$ can be expressed as:

$$C_d(t') = \frac{1}{\int_S R\, dS} \int_S P'\, C_e(t')\, R\, dS \tag{2.12}$$

Advantages and limitations: The mass balance model described here takes account of both the temporal variation of the ^{137}Cs fallout input and its initial distribution in the surface soil, prior to incorporation into the plough layer by tillage. Results obtained using this model are likely to be more realistic than those provided by the simplified mass balance model presented in the previous section. However, in order to use this model, information on the plough depth *h*, relaxation mass depth *H* and the parameter γ is required. Although the first is relatively easy to estimate from the ^{137}Cs depth distribution at a non-eroding point, the latter two are more difficult to specify. The relaxation mass depth *H* is best estimated experimentally for the local soil type and the parameter γ can be estimated from available long-term rainfall records and information on the timing of tillage operations at the study site. Since the model requires information on the ^{137}Cs concentration of soil eroded from upslope, the sampling points used with the model must be arranged along a downslope transect (IAEA, 2011). Normally, a uniform spacing of sampling points is assumed. The first (upper) point on a transect should represent an eroding point (Walling *et al.*, 2014).

2.5.4 *The Profile Distribution Model (for uncultivated soils)*

For uncultivated soils, the depth distribution of ^{137}Cs in the soil profile will be significantly different from that in cultivated soils, where the ^{137}Cs is mixed within the plough or cultivation layer. In many situations, the depth distribution of ^{137}Cs in an undisturbed stable soil will exhibit an exponential decline with depth that may be described by the following function (cf. Zhang *et al.*, 1990; Walling & Quine, 1990):

$$A'(x) = A_{ref}(1 - e^{-x/h_0}) \tag{2.13}$$

where:

$A'(x)$ is the amount of ^{137}Cs above the depth x (Bq m^{-2});

A_{ref} is the ^{137}Cs reference inventory (Bq m^{-2});

x is the depth from the soil surface (kg m^{-2});

h_0 is the coefficient describing profile shape (kg m^{-2}).

If it is assumed that the total ^{137}Cs fallout occurred in 1963 and that the depth distribution of the ^{137}Cs in the soil profile is independent of time, the *erosion rate Y* for an eroding point (with total ^{137}Cs inventory Au (Bq m^{-2}) less than the local reference inventory A_{ref} (Bq m^{-2})) can be estimated as:

$$Y = \frac{10}{(t-1963)P}\ln(1-\frac{X}{100})h_0 \tag{2.14}$$

where:

Y is the annual soil loss (t ha^{-1} yr^{-1});

t is the year of sample collection (yr);

X is the percentage ^{137}Cs loss in total inventory in respect to the local ^{137}Cs reference value (defined as $[(A_{ref}-A_u)/A_{ref}] \times 100$);

A_u is the measured total ^{137}Cs inventory at the sampling point (Bq m^{-2});

P is the particle size correction factor.

For a depositional location, a tentative estimate of the deposition rate R' can be estimated from the excess ^{137}Cs inventory $A_{ex}(t)$ (Bq m^{-2}) (defined as A_u-A_{ref}) and the ^{137}Cs concentration of deposited sediment C_d:

$$R' = \frac{A_{ex}}{\int_{t_0}^{t} C_d(t')e^{-\lambda(t-t')}dt'} = \frac{A_u - A_{ref}}{\frac{P'}{\int_S RdS}\int_S A_{ref}\left(1-e^{-R/h_0}\right)dS} \tag{2.15}$$

Advantages and limitations: The profile shape model is simple and easy to use. However, this model involves a number of simplifying assumptions. Most importantly, it does not take account of the time-dependent behaviour of the ^{137}Cs fallout input and the progressive development (downward movement) of the depth distribution of the ^{137}Cs within the soil profile after deposition from the atmosphere. As such, it is likely to overestimate rates of soil loss, since the amount of ^{137}Cs removed with a given depth of soil is likely to be greater than suggested by the present-day depth distribution of ^{137}Cs in the soil (IAEA, 2011).

2.5.5 *The diffusion and migration model (for uncultivated soils)*

Although the depth distribution model described above can be used to obtain approximate estimates of soil erosion or deposition rates for uncultivated soils, a more realistic approach needs to consider the time-dependent depth distribution behavior, which will reflect the time-dependent ^{137}Cs fallout input and the progressive redistribution of ^{137}Cs in the soil profile after deposition from the atmosphere (e.g. Pegoyev & Fridman, 1978; Walling & He, 1993; He & Walling, 1997). In many situations, the redistribution of ^{137}Cs in uncultivated soils can be described using a one-dimensional diffusion and migration model characterised by an effective diffusion coefficient and migration rate (cf. Pegoyev & Fridman, 1978; Reynolds *et al.*, 1982; He & Walling, 1997). For example, in some situations, the ^{137}Cs depth profile in uncultivated soils exhibits a broad concentration peak with the maximum concentration located below the soil surface. The variation of the ^{137}Cs concentration $C_u(t)$ (Bq kg^{-1}) in surface soil with time t (yr) may be approximated as:

$$C_u(t) \approx \frac{I(t)}{H} + \int_0^{t-1} \frac{I(t')e^{-R/H}}{\sqrt{D\pi(t-t')}} e^{-V^2(t-t')/(4D)-\lambda(t-t')} dt' \tag{2.16}$$

where:

D is the diffusion coefficient (kg^2 m^{-4} a^{-1});

V is the downward migration rate of ^{137}Cs in the soil profile (kg m^{-2} a^{-1}).

For an eroding point, if sheet erosion is assumed, then the erosion rate R may be estimated from the reduction in the ^{137}Cs inventory $A_{ls}(t)$ (Bq m^{-2}) (defined as the ^{137}Cs reference inventory A_{ref} less the measured total ^{137}Cs inventory A_u (Bq m^{-2})) and the ^{137}Cs concentration in the surface soil $Cu(t')$ given by Equation 2.16 according to:

$$\int_0^t PRC_u(t')e^{-\lambda(t-t')}dt' = A_{ls}(t) \tag{2.17}$$

For a depositional location, the deposition rate R' can be estimated from the ^{137}Cs concentration of deposited sediment $C_d(t')$ and the excess ^{137}Cs inventory $A_{ex}(t)$ (defined as the total measured ^{137}Cs inventory A_u less the local reference inventory A_{ref}) using the following relationship (Walling & He, 1999b):

$$R' = \frac{A_{ex}}{\int_{t_0}^{t} C_d(t')e^{-\lambda(t-t')}dt'} = \frac{A_u - A_{ref}}{\int_{t_0}^{t} C_d(t')e^{-\lambda(t-t')}dt'} \tag{2.18}$$

where $C_d(t')$ can be calculated from:

$$C_d(t') = \frac{1}{\int_S R dS} \int_S P' P C_u(t') R dS \tag{2.19}$$

Advantages and limitations: Since the diffusion and migration model described here takes into account the time-dependent behaviour of both the ^{137}Cs fallout input and its subsequent redistribution or downward movement in the soil profile, it therefore represents an improvement over the profile shape model presented in the previous section. However, to use this model, more information on the behaviour of ^{137}Cs in undisturbed soils is needed. More particularly, estimates of the diffusion coefficient D and the migration rate V are required. These can be derived from detailed measurements of the ^{137}Cs depth distribution (Walling *et al.*, 2014).

2.6 Particle size correction factors for eroding and deposition site

The particle size correction factors aim to take account of the grain size selectivity of erosion and sedimentation processes. For an eroding site, the correction factor is a function of the ratio of the ^{137}Cs concentration of mobilised sediment to that of the original soil (cf. He & Walling, 1996). The particle size correction factors *P* and *P'* provide a means of taking account of the particle size selectivity of erosion and deposition processes and thus the enrichment or depletion of eroded and deposited sediment in ^{137}Cs relative to the original soil. The correction factor *P*, which is applied when estimating erosion rates, is defined as the ratio of the ^{137}Cs concentration in mobilized sediment to that of the original soil, whereas *P'*, which is used when estimating deposition rates, is defined as the ratio of ^{137}Cs content of deposed sediment to that of the mobilized sediment. Both factors could be estimated directly by measuring the ^{137}Cs concentration of the relevant soil or sediment, but, since such data are unlikely to be available, they are more commonly estimated indirectly, by comparing the particle size distributions of the two components of the ratio. It is well known that particle size exerts a strong influence on the ^{137}Cs content. One useful approach to estimating *P* and *P'* from particle size data is that reported by He and Walling (1996), which makes use of measurements of the specific surface area of the soil or sediment involved. An estimate of the specific surface area of a sediment sample can be readily derived from its particle size distribution, by assuming spherical particles (Walling et al., 2002). In order to estimate values for the particle size correction factor for eroding sites (*P*) and depositional sites (*P'*), information on the grain size distribution of the soils and the mobilised sediment is needed. Values of *P* and *P'* can be estimated using the procedures described by He and Walling (1996). If the specific surface area of mobilised sediment is S_{ms} (m^2 g^{-1}), and that of the original soil is S_{sl} (m^2 g^{-1}), P can be calculated as:

$$P = \left(\frac{S_{ms}}{S_{sl}} \right)^{v} \tag{2.20}$$

where v is a constant with a value of ca. 0.65. If the specific surface area of deposited sediment is S_{ds} (m^2 g^{-1}), P' can be calculated as:

$$P^{'} = \left(\frac{S_{ds}}{S_{ms}} \right)^{v} \tag{2.21}$$

The value of v in Equation (2.21) is the same as that in Equation (2.20). The following relationship therefore exists:

$$PP^{'} = \left(\frac{S_{ms}}{S_{sl}} \right)^{v} * \left(\frac{S_{ds}}{S_{ms}} \right)^{v} = \left(\frac{S_{ds}}{S_{sl}} \right)^{v} \tag{2.22}$$

2.7 Sampling objectives

The design of effective and appropriate sampling strategies is an important and critical step for successfully applying ^{137}Cs or other FRNs in soil erosion and sedimentation studies. The strategy will depend on the study objectives. According to Pennock and Appleby (2002), investigations can be classified into three different categories: soil redistribution studies, floodplain/lake and reservoir sedimentation studies, and integrated catchment studies.

Further the selection of the study site and the background data collection are very important. The selection of an appropriate site is a key requirement in any study. It is recommended that several sites are visited to support selection of a site that is representative of the wider landscape or land use type under investigation. If the study is to be carried out in a country or region where the ^{137}Cs method has not previously been applied, or if the study is to be carried by a research team which does not have experience of the method, it is recommended that a small site is selected and that geographical conditions and the dynamics of soil redistribution are not too complex. Application of the ^{137}Cs method and interpretation of the results obtained is much easier for smaller sites with relatively simple geographical conditions and limited variability of soil redistribution processes (Mabit *et al.*, 2014b).

Before formulating the sampling strategy, a reconnaissance survey to include background data collection will be required. This requires two main actions:

1. Enquiries, collection and compilation of background environmental and socio-economic information like using existing references, official documents and governmental reports; the major datasets can be included as follows (see Anderson & Ingram, 1993): climatic data over several years (rainfall, temperature, etc.); geomorphology and pedology (topography, typical landforms of the region, soil type and texture); land use (type of land use since the 1950s, type and frequency of tillage, specific soil conservation practices if applied); messages or records (earlier erosion events, flooding or extreme weather events, conditions of their occurrence and their characteristics).
2. A reconnaissance survey of the selected study area, because the objective is to recognize the specific environmental conditions of the study area including the identification of potential suitable reference sites, the typical topography of the study area, soil types and land use. This is a key step that will allow refinement of the sampling strategy.

2.8 Advantages and limitations ^{137}Cs techniques for assessing erosion and sedimentation

The ^{137}Cs technique possesses a number of major advantages over traditional approaches to document erosion and deposition rates. They can be summarized as follows (Walling & Quine, 1995; Mabit *et al.*, 2008, Zupanc & Mabit, 2010):

a) The approach provides retrospective information. Past erosion rates can be estimated from samples collected at the present time;
b) The technique provides estimates of medium-term average (50 years) rates of soil redistribution and therefore integrate land use and climatic variability, including extreme events;
c) The technique provides information on both erosion and deposition and therefore permits quantification of net erosion and sediment export;
d) The assessment of soil redistribution provided by ^{137}Cs integrates all processes involving soil particle movements (water, wind and tillage erosion);
e) The technique permits the quantification of soil loss and deposition associated with sheet erosion, which is difficult to assess using other classical approaches at the field scale, due to the limited depths of soil removal and accumulation commonly involved;
f) Estimates are based on individual sampling points within the landscape, and spatially distributed information on rates and patterns of soil redistribution can therefore be generated;

g) The technique does not require costly and labour intensive long-term monitoring programs. The results can be obtained from a single visit to the site for soil sampling;

h) Although ^{137}Cs is commonly used retrospectively, by comparing the inventories measured at a study site with the local reference inventory, it can also be used for ongoing monitoring of soil redistribution by comparing the inventories measured at specific points within the study site between successive sampling campaigns separated by a period of several years e.g. (Lobb & Kachanoski, 1999; Loughran & Balog, 2006; Tiessen *et al.,* 2009);

i) Sampling is relatively simple and cost-effective and can be completed in a relatively short time, depending on the sampling density and the size of the area investigated;

j) The site disturbance during sampling is minimal and will not interfere with seeding and cultivation operations. Furthermore, there is no disturbance of natural runoff and erosion processes, such as might occur with the installation of bounded erosion plots. Whole fields and the complete slope profile can be sampled.

The main constraints or limitations are listed below (Mabit *et al.*, 2008):

a) In some areas, especially in the southern hemisphere, ^{137}Cs inventories are low and gamma analyses will require greater count times to obtain an acceptable precision for the measurements;

b) The Chernobyl accident provided additional fallout inputs in some areas. These inputs must be taken into account, when using ^{137}Cs measurements to estimate soil redistribution rates. Unless the Chernobyl input is considerably greater than the input of ^{137}Cs associated with "bomb fallout", such that the ^{137}Cs measurements will primarily reflect erosion occurring after 1986, the conversion models used must be modified to take account of both bomb fallout and the additional Chernobyl fallout input in 1986;

c) The technique is primarily suited to estimating medium-term (i.e. 50 years) average erosion rates and cannot readily provide the information needed to document changes in erosion rates linked to short-term changes in land use and management practice;

d) The commonly used approach involves comparison of measured inventories with a reference value, representing the inventory associated with a point experiencing neither erosion nor deposition. In view of its central importance to the reliability of the estimates of soil redistribution rates obtained, it is important that the reference inventory should be accurately determined. It is important to ensure that the sampling site used to establish the reference inventory provides a representative estimate of the local reference inventory (Sutherland, 1996). This may cause problems in mountain regions, where rainfall may be characterized by high spatial variability, in areas with stony soils, in areas under intensive cultivation where few undisturbed areas exist, in areas where snow represents a substantial proportion of the annual precipitation and where significant drifting may occur, and in arid and semi-arid areas with poor vegetation cover, where redistribution of soil by aeolian processes can cause the accumulation of additional ^{137}Cs labelled sediment at the reference site;

e) There is a need to take account of sampling representativeness and sample variability (see Owens & Walling, 1996). Multiple or replicate samples are normally collected to determine the reference inventory value. A guideline on the number of reference samples required will be discussed in the following section.

2.9 Re-sampling approach

In reference to some points above-named (the first a) and h)), it is possible to describe the re-sampling approach. It has been used by de Jong and Kachanoski (1988), Lobb and Kachanoski (1999), and Tiessen *et al*. (2009). But a degree of confusion has occurred regarding the exact meaning of the terms *Re-samplin*g and *Repeated-sampling*. In the studies reported by Kachanoski and de Jong (1984), de Jong and Kachanoski (1988), Lobb and Kachanoski (1999), Li *et al*. (2011) and Tiessen *et al*. (2009), the term repeated-sampling has been used to describe an alternative approach to establish the reference inventory. The reference inventory is usually established by sampling an area with undisturbed soils and minimum slope, where soil redistribution is not probable to have occurred during the period between the bomb fallout input and the time of sampling, but in some locations it can be difficult to identify appropriate reference sites. The *Repeated-sampling* approach uses an initial sampling campaign to establish the inventory at each sampling point within a study area and the area is sampled again, usually a decade or more later, to document the inventories at the same sampling points. The inventory documented by the initial sampling campaign for each sampling point is used as the reference inventory for that point. Comparison of the inventory measured by the repeat sampling for a given point with the reference inventory for the same point provides the basis for estimating the mean annual soil redistribution rate for that point over the period between the two sampling campaigns. The success of this approach is related to a number of requirements: the period between the two sampling campaigns must be of sufficient length to produce a significant change in the inventories measured at the individual sampling points; soil redistribution rates must be high enough to produce a significant change in the ^{137}Cs inventory at a sampling point, taking account of measurement precision; it is necessary to relocate the original sampling points but there is a need do not collect precisely the same point because they were disturbed by the previous sampling operation; its use has been limited to cultivated areas where erosion rates are commonly higher than in uncultivated areas and where the mixing of the soil by tillage reduces small-scale variability in soil inventories - which could complicate or even preclude direct comparison of the inventories measured on the two occasions; both sets of samples may have high uncertainties due to measurement errors. The repeated-sampling approach described above does not generate estimates of soil redistribution rates for different periods, although the potential to do this could exist if it was possible to establish a local reference inventory and this was used with the results of the first sampling campaign to estimate the soil redistribution rate for the period between the bomb fallout and the time of the first sampling campaign. This soil redistribution rate could then be directly compared with that relating to the period between the two sampling campaigns. Subsequent re-sampling could provide estimates of soil redistribution rates for successive periods (Porto *et al.*, 2014).

Loughran and Balog (2006) reported a study where the *Re-sampling* was used to indicate a different approach with estimates of soil redistribution rates for different periods that could be directly compared. ^{137}Cs sampling campaigns were undertaken in a vineyard (in Australia) firstly in 1984-1985 and then in 2004. This vineyard had been cultivated and planted in 1971. A similar sampling scheme was used for the two campaigns but the sampling points were not collected closely and the two sets of results were treated independently. The measurements of ^{137}Cs inventory given by the two sampling campaigns were compared with an estimate of the local reference inventory to estimate mean annual soil redistribution rates for the periods 1971-1985 and 1971-2004. The values for the time window 1986-2004 were obtained by time-weighted subtraction and these were compared with those for the period 1971-1985. The results showed a substantial reduction in the mean annual net erosion rate during the second time window. This reduction was connected to a progressive reduction in soil erodibility through time because of incorporation of subsoil into the plough layer and the cessation of clean till cultivation in 1998. The *Re-sampling* approach employed by Loughran and Balog (2006) is seen as probably offering the greatest potential for generating information on changing erosion rates, because it provides estimates of soil redistribution rates for two discrete periods. These can be compared and further re-sampling in the future could add further periods. Furthermore, it is not dependent upon direct comparison of the ^{137}Cs inventories measured at identical individual sampling points on the two occasions (Porto *et al.*, 2014). A re-sampling approach using ^{137}Cs in soil erosion and sediment delivery investigations can be employed to subdivide the period between the occurrence of bomb fallout and the present, in order to document changes in rates of soil loss or soil redistribution within this period. Therefore, it is possible to compare the estimate of mean annual erosion from the period 1954 to the first time of sampling with the recent measurements (usually a decade or more later).

Materials and Methods

3.1 The study catchments

The study focuses on three small catchments, W1, W2 and W3, located near Crotone (35 m a.s.l., 39°09′02″N, 17°08′10″E) in Calabria, southern Italy. These catchments are located in the ephemeral headwaters of the larger Crepacuore basin (Fig. 3.1) which are incised into the Upper Pliocene and Quaternary clays, sandy clays and sands underlying the local area (Sorriso-Valvo *et al.*, 1995). The entire Crepacuore basin area is 1860 ha with a mean slope of 11.7% and a mean altitude of 80.51 m a.s.l. (PAI Regione Calabria).

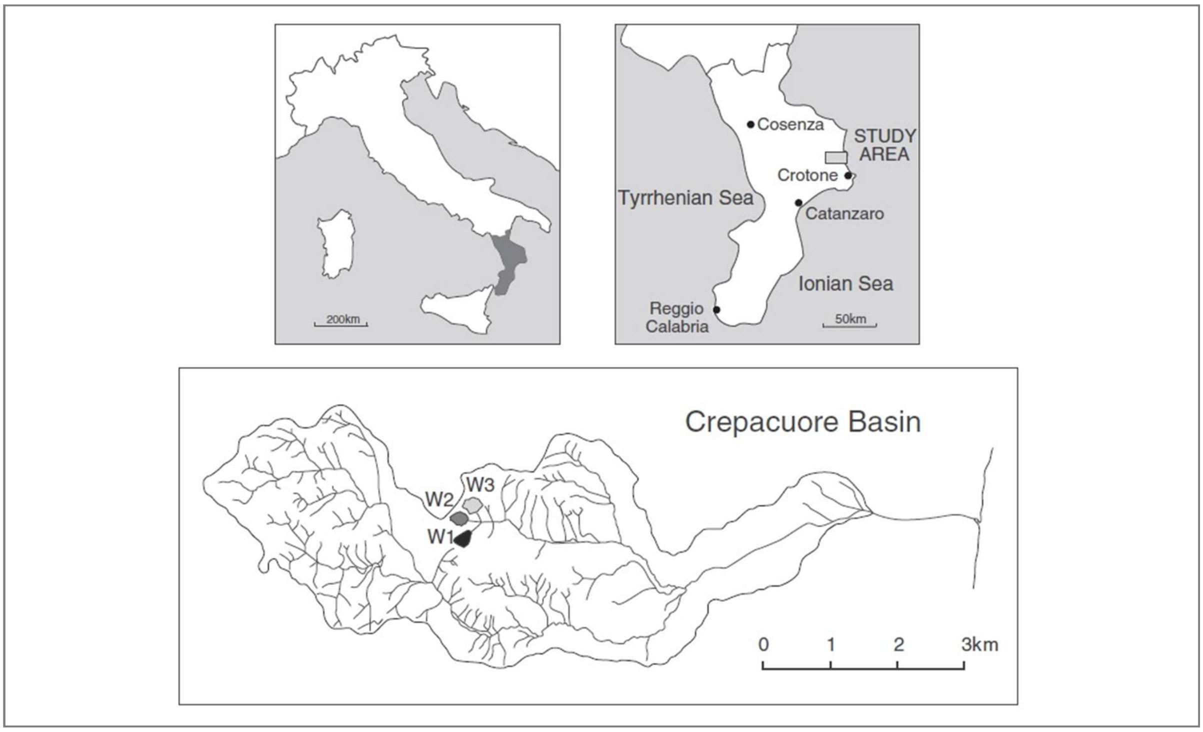


Fig. 3.1 – The study area (Porto *et al.*, 2016a).

These three catchments (W1, W2 and W3) have never been cultivated and they both originally supported a rangeland vegetation cover (Avolio *et al.*, 1980). The details concerning their percentage sand, silt and clay content of the soils are presented in Table 3.1.

Tab. 3.1 – The texiture characteristics of the study catchments.

Mean soil content		W1	W2	W3
Sand	%	14	14.6	20.7
Silt	%	44.5	49.2	45.5
Clay	%	41.5	36.2	33.8

The area of W1 catchments is 1.47 ha with mean altitude of 122 m a.s.l. and a mean slope of 53%. Below a digital elevation model of this catchment is showed (Fig. 3.2a). W1 catchment still supports a rangeland vegetation cover dominated by *Lygeum spartum* and *Atriplex halimus* (Avolio *et al*., 1980) (Fig.3.2b).

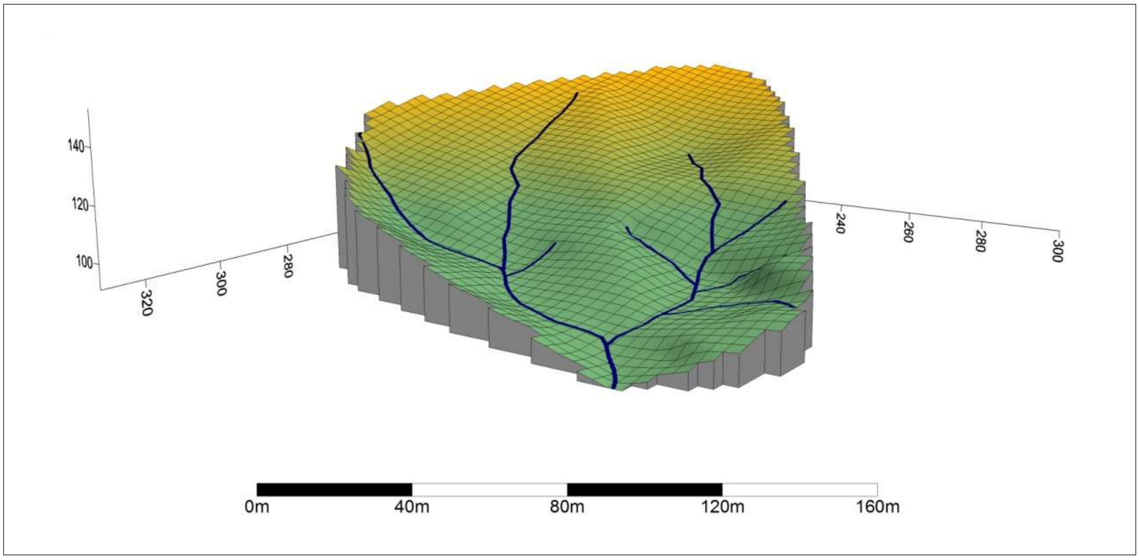


Fig. 3.2a – The DEM of the W1 catchment.

Fig. 3.2b – The W1 catchment vegetation cover.

The W2 catchment area is 1.375 ha with a mean altitude of 103 m a.s.l. and a mean slope of 35%. The W3 catchments areas is 1.654 ha with a mean altitude of 98 m a.s.l. and a mean slope of 24%. Although the two catchments are of a similar size, the relief of catchment W3 is more subdued than that of catchment W2 and the mean slope angles are significantly lower. Below two digital elevation model of both W2 and W3 catchment are showed (Fig. 3.3a and Fig. 3.3b).

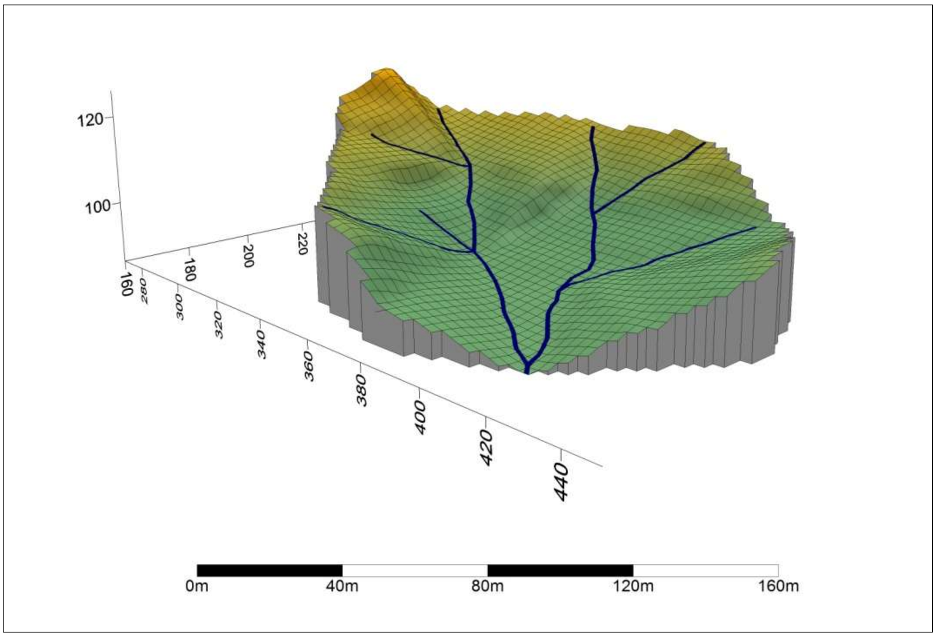


Fig. 3.3a – The W2 catchment DEM.

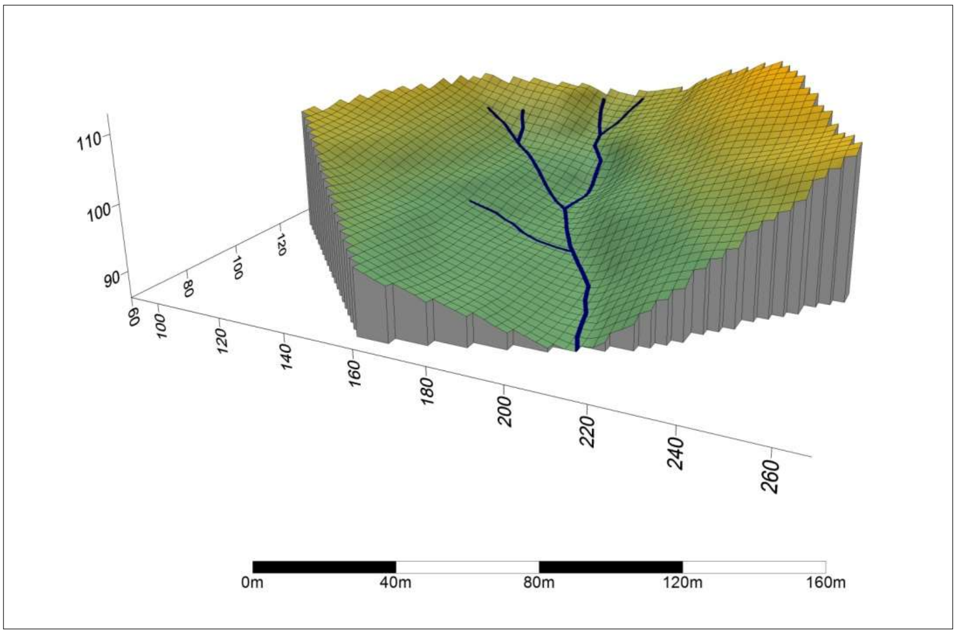


Fig. 3.3b – The W3 catchment DEM.

In 1968, both W2 and W3 were planted with *Eucalyptus occidentalis* trees while the W1 was left unmodified and under rangeland vegetation cover (Cinnirella *et al*., 1998) comprising both grass and shrubs. The trees were cut in 1978 and 1990 in catchment W2 (Fig. 3.4a) and in 1986, 2006, and 2016 in catchment W3 (Fig. 3.4b), respectively, with the tree cover being subsequently restored by natural re-growth.

Fig. 3.4a – The cutting in 1978 and in 1990 in the catchment W2.

Fig. 3.4b – The cutting in 2006 and in 2016 in the catchment W3.

Logging operations took place across the entire area of the catchments and involved the removal of ca. 25–40 m^3 of biomass during each cutting campaign. The harvested timber was extracted from the catchments manually, in order to minimize the impact on the soil surface. In catchment W3 the forest cover is currently almost continuous and only about 2–3% of the area is characterized by a grass cover (Porto *et al.*, 2009). Within catchment W2, the tree cover is not uniform, and about 20% of its area, primarily along its northern margin, is characterized by discontinuous trees and a sparse grass cover. Elsewhere, the tree cover is relatively uniform. The soils under the eucalyptus trees are largely devoid of ground cover and much of the surface beneath the trees and sparse grass cover is therefore bare. The bare soils are exposed to rainsplash and surface runoff during heavy rainfall and significant sheet erosion occurs at such times. The area with discontinuous tree cover is the area of greatest soil loss (Porto *et al.*, 2005).

The climate of the study area is typically Mediterranean, with a mean annual rainfall of about 670 mm for the period 1954–2012 at Crotone (10 km distant) (Porto *et al.*, 2014). Most of the rainfall falls between October and March. The annual potential evapotranspiration in this location is estimated to be 1100-1200 mm (based on the Penman-Monteith formula) (Porto *et al.*, 2016b).

3.2 Catchment instrumentation

The study catchments have been instrumented for measuring rainfall (Fig. 3.5), runoff and sediment yield since 1978 by the National Research Council of Italy (CNR) “Soil Conservation Project” to monitor the effects of afforestation on their hydrological response and sediment yield (Avolio *et al.*, 1980; Iovino and Puglisi, 1991).

Fig. 3.5 – The mechanical pluviograph and the digital pluviograph in catchment W3.

Runoff from the catchments is measured using an H-flume structure (Brakensiek et al., 1979), equipped with a mechanical stage recorder. The sediment load passing the gauging structure is measured using a Coshocton wheel sampler installed below the H-flume (Figg. 3.6a and 3.6b). This sampler collects an aliquot of ca.1/200th of the flow, which is diverted to a tank. After each storm event, samples are collected from the tank to determine the mean sediment concentration in the tank and the sediment yield is calculated as the product of this concentration and the total runoff volume for the event measured by the H-flume. The annual sediment yield is in turn calculated as the sum of the sediment loads for all storm events occurring during the year (Porto *et al.*, 2003).

Fig. 3.6a – The H-flume structure, the stream gauge and the Coshocton wheel sampler.

Fig. 3.6b – A particular of the gauging structure (hydrometer) installed in the catchments.

Due to malfunctioning of the sampling device, full records of sediment yield at the catchment outlet are not available for the years 1995-2005. In December 2005, the sampling equipment was refurbished and the monitoring activity recommenced, with no loss of data to date (Porto *et al.*, 2009). The values of annual sediment yield provided by this instrumentation are estimated to have a precision of ca. ±15% at the 95% level of confidence.

Further, it was decided to refine the procedures of reading and interpretation of rainfall and hydrometric data at the Warsaw University of Life and Science during the period from February to May 2016. At the Faculty of Civil and Environmental Engineering, a rainfall-runoff sediment yield model was used to cover the lack of runoff measurements due to the malfunctioning of the streamgauge stations during the period 2014-2015. In view of the data of the rainfall events available for that period, the SN computer programme developed at the Department of Water

Engineering of WULS-SGGW was applied. This model was able to generate simulated hydrographs to cover this gap, so the database of runoff and sediment yield during this period is complete.

3.3 Soil sampling strategy

3.3.1 Sampling design

Concerning the sampling strategy at the reference site, one of the major issues related to the use of ^{137}Cs as a soil tracer is the choice of the reference site which is used to estimate the total ^{137}Cs fallout input or reference inventory, which is a key component of the conversion models used to estimate erosion and sedimentation rates from the assembled data. Hence, before implementing any study, a suitable reference site should be identified (Mabel *et al.*, 2014b).

The estimation of net erosion from ^{137}Cs measurements is founded on the hypothesis that the local fallout of ^{137}Cs was uniform in space. The ^{137}Cs activities of individual sampling points are compared with that of the control or reference site to estimate the amount of erosion or deposition that has occurred since initial fallout. Measurements undertaken for reference sites usually show some variability in ^{137}Cs inventory (Owens & Walling, 1996). This variability in ^{137}Cs reference values could be related to:

- random spatial variability in soil properties that influence ^{137}Cs micro-variability (e.g. infiltration capacity linked to soil bulk density, cracking and stoniness), the effects of vegetation cover and roots, micro-topography, and human and animal disturbance;
- apparent systematic spatial variability at a regional scale, where variations in rainfall and wind flows may have affected the initial ^{137}Cs fallout;
- sampling variability, a function of the surface area over which the samples are collected;
- measurement precision.

To minimize the variability and to maximize the representativeness of the reference inventory value the following selection criteria have to be applied (Mabel *et al.*, 2008):

a) The reference site should not have been cultivated or disturbed since the beginning of the 1950s. The site should have been stable since the first global occurrence of ^{137}Cs fallout (1954). These undisturbed sites should ideally be selected in protected areas of permanent grassland, abandoned pasture which have not been ploughed for the last 60 years;

b) In order to represent the initial ^{137}Cs input, the site needs to be selected within the study area (field or watershed) or near it (e.g. around 1 km). The stable area should be located as close as possible to the area under investigation;

c) The reference site should not have been influenced by erosion processes, which generally implies the need for a flat area;

d) Highly bio-disturbed areas should be avoided.

The two key objectives of soil sampling at the reference site are to establish the reference inventory (Bq m^{-2}) and to determine the depth distribution of the ^{137}Cs activity to support application of conversion models. In the first case, sampling commonly involves the use of bulk cores, whereas depth incremental sampling is required to establish the depth distribution as well as using of a scraper plate, sectioned cores or the excavation of a soil pit and collection of samples from the exposed face of the pit.

For reference inventory samples, the maximum penetration depth of ^{137}Cs should first be determined to ensure that the complete ^{137}Cs inventory will be represented by the core samples. This is achieved through depth incremental sampling. At undisturbed pasture and forest sites most of the ^{137}Cs should be concentrated within the top 10 cm of the soil profile. At a typical reference site, no ^{137}Cs will be found below 25-30 cm (Walling & Quine, 1995; Owens *et al.*, 1996; Wallbrink *et al.*, 1999). When the depth distribution of ^{137}Cs is known, bulk core samples can be collected. The depth of the core samples should exceed the actual depth of the ^{137}Cs layer by at least 5 cm, in order to ensure that all ^{137}Cs inventory is included in the sample

To express the spatial distribution of ^{137}Cs resulting from post-fallout soil redistribution, the sampling strategy must integrate the micro-scale spatial variability in ^{137}Cs inventories and the spatial variability of soil redistribution processes (Walling & Quine, 1993). In uncultivated sites, where there is a potential for local variability due to bioperturbation or variability in vegetation cover, it is recommended that multiple cores (2 to 3) are collected to represent the individual sample points. The key factor in designing a suitable sampling strategy is to collect representative samples within the field, which reflect the spatial variability of soil redistribution processes. In this context, two approaches are commonly adopted:

1. *Transect approach* - This approach is based on the hypothesis that there is lateral uniformity in ^{137}Cs distribution along parallel transects and is suitable for fields or small areas characterized by simple topography where no significant across-slope curvature exists and where each point along the transect receives flow from only those points immediately upslope. This method is especially suited to relatively steep, short and homogeneous slopes. In these situations, a single transect can represent the variability

of ^{137}Cs inventories. If significant plan curvature exists, a single transect is not sufficient and a series of equally spaced parallel transects should be used. Transects are generally aligned along the axis of greatest slope, and consist of a sequence of sampling points from the upslope to the down-slope boundary. A distance of 10 to 20 m between the sampling points is generally used. The multiple transect approach is particularly convenient if resources are limited. It allows a more rational use of resources than the grid approach, because the variability is usually much higher along the slope than across the slope.

2. *Grid approach* - When the topography is more complex, it is necessary to collect samples from a grid framework, covering the field with regularly spaced sampling points. Usually a square grid is used, but rectangular grids are also sometimes used. It is then possible to define effectively the spatial distribution of ^{137}Cs and the resulting soil redistribution pattern. The sampling density depends on the total area and is conditioned by the resource availability. However, a minimum density is required to establish the spatial distribution of the ^{137}Cs inventories. The number and spacing of the samples will influence the ^{137}Cs pattern obtained and the resulting assessment of the pattern of soil redistribution. Mabit *et al.*, 2002) reported a range of different grid sizes from published studies (e.g. 10 by 10 m; 15 by 20 m; 20 by 25 m; 30 by 30 m; 50 by 50 m; and 100 by 100 m) (Mabel *et al.*, 2014b).

As the use of ^{137}Cs measurements in soil erosion and sedimentation investigation requires information on both bulk ^{137}Cs inventories (Bq m^{-2}) and the ^{137}Cs vertical depth distribution, two methods are commonly used to collect soil samples:

1. *Bulk sampling using cores* - Generally, a steel cylinder is used as the corer. The diameter of the tube is usually 7 to 10 cm (Wallbrink *et al.*, 2002) (Fig. 3.7) with a wall thickness of 2-5 mm. The corer is constructed to cope with stony and compacted soil. To facilitate both the insertion of the core tube and sample extraction, the cutting edge of the tube should have a smaller internal diameter than the tube itself (Walling & Quine, 1993). The tube can be inserted into the soil manually (e.g. using a hammer) or coupled with a mechanical percussion (Fig. 3.8) driver in hard soils.

Fig. 3.7 – Some steel cylinder tube cores used for sampling.

Fig. 3.8 – The motorized percussion hammer.

2. *Depth incremental sampling* - Depth incremental sampling provides important information on the depth distribution (vertical distribution or profile) of ^{137}Cs. This is key information for the reference site as well as the study site. It is needed to determine the appropriate depth of bulk sampling at both reference and study sites, to ensure that the samples include the whole ^{137}Cs inventory. For the reference site, the shape of the ^{137}Cs vertical distribution provides valuable confirmation that the site is undisturbed and thus appropriate for establishing the local ^{137}Cs fallout input.

   A sectioned core provides the simplest means of defining the ^{137}Cs depth distribution. However, in order to provide a sufficient mass of sample from each depth increment for subsequent radiometric analysis, the diameter of the core tube generally needs to be greater than those used to collect bulk cores. A cylindrical steel tube with a diameter of 10 to 15 cm is commonly used (Fig. 3.9). Once collected, the core is then sectioned into incremental samples which can be counted separately. To easily extract the soil core, one of the best options is to use a specially designed core tube which splits into two longitudinal sections to allow access of the contents. Soil increments of 2 to 5 cm are generally used. However, for a precise ^{137}Cs profile shape, it is better to cut the core at 2 cm intervals (Mabel *et al.*, 2014b).

Fig. 3.9 – An example of the sectioning into incremental samples.

3.3.2 Field sampling

A re-sampling approach can be employed to subdivide the period between the events of bomb fallout and the present, in order to document the changes in rates of soil loss or the soil redistribution within this time slots.

The re-sampling approach is seen as probably offering the greatest potential for generating information on changing erosion rates, because it provides estimates of soil redistribution rates for two discrete periods (Loughran *et al.* 2006).

The first soil sampling within the W1, W2 and W3 study catchments involved several campaigns undertaken from 1998 to 2001. Nowadays, the three catchments were sampled again and the samples were collected from points very close to the original sampling points in the campaigns undertaken from 2013 to 2015. In this way, it was possible to compare the estimate of mean annual erosion for the period 1954-1998 with that for the period 1954-2015 (Fig. 3.10). Thus, the difference in erosion rates between the two periods provides the average erosion rate of this short time (1998-2015).

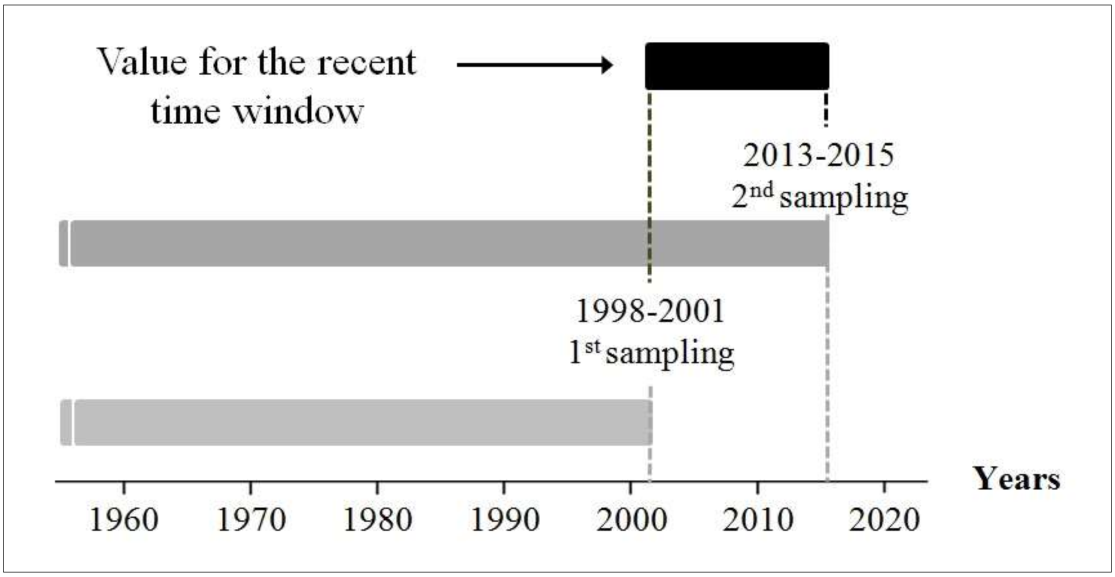


Fig. 3.10 – The time window of re-sampling approach.

The campaigns focused on catchment W1 were undertaken in 1999 and 2001 and involved the collection of 68 bulk cores within the catchment using a steel core tube (6-9 cm diameter) driven into the soil by a motorized percussion corer and subsequently extracted using a hand-operated winch (Porto *et al*., 2011). In September 2014, the catchment W1 was sampled again and the same 68 sampling points used during the previous field campaigns were revisited (Fig. 3.11). Because the actual sampling points could not be relocated precisely, their locations were displaced by about 2 m to avoid the area that was disturbed during the first campaign. Each site was re-sampled using a steel core tube (10 cm diameter) driven into the ground by a motorized percussion corer to a depth of ca. 30 cm.

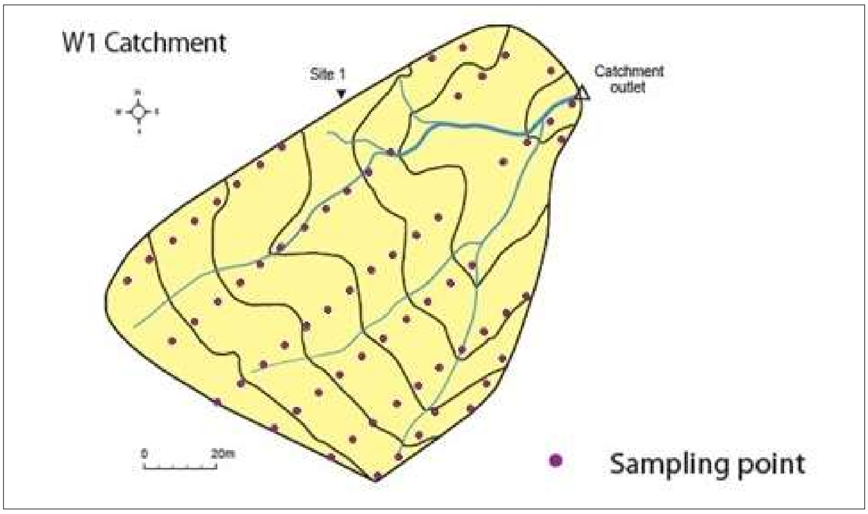


Fig. 3.11 – The sampling points within the W1 catchment.

For the W2 and W3 catchments, a first sampling campaign was undertaken in the study catchment in 1998-2001, in order to collect information on ^{137}Cs inventories within the catchment.

In the campaign undertaken within the W2 catchment, 55 replicate bulk cores were collected at the intersections of an approximately 20 m x 20 m grid, with additional cores collected to take account of topographic variability. The cores were collected using a steel core tube (8.6 cm diameter) driven into the ground by a motorized percussion corer to a depth of 15 cm. A number of additional 45 cm cores was collected and sectioned into 15 cm depth increments, in order to confirm that no ^{137}Cs content was detectable below the depth of 15 cm. The two cores collected for each sampling point were bulked. Where tree cover existed in the vicinity of the sampling points, care was taken to ensure that cores were collected from points located between the trees, rather than close to their trunks (Porto *et al.*, 2014).

The samples used to establish the local reference inventory were collected during a field campaign undertaken in 1999 (Porto *et al.*, 2001). An adjacent area of undisturbed rangeland with minimal slope and similar altitude was selected as the reference site. In this case, one set of samples was collected using a scraper plate (cf. Campbell *et al.*, 1988), in order to provide samples from a larger and therefore more representative area. The sampling frame isolated a surface area of 652 cm^2 for sample collection and depth incremental samples were collected at increments ranging from 1 to 4 cm (progressively increasing with depth) to a depth of 50 cm. In addition, six 8.6 cm diameter soil cores were collected from the reference site and sectioned into 2 cm depth increments, in order to assess the local spatial variability of the ^{137}Cs inventory and the associated depth distributions. Because of the greater surface area associated with the scraper plate (652 cm^2), the areal activity density indicated by these samples was assumed to provide the best estimate of the local reference inventory (Porto *et al*., 2014).

In 2013, the catchment was sampled again and the same 55 sampling points used during the field campaign undertaken in 1998 were revisited (Fig. 3.12). Anyway, in the absence of high precision survey data it was not possible to locate the previous sampling points precisely. Furthermore, the new sampling point was displaced laterally by ca. 2 m to avoid sampling soil that might have been disturbed during the first campaign. Each site was re-sampled using a steel core tube (10 cm diameter) driven into the ground by a motorized percussion corer to a depth of ca. 15 cm, in order to ensure consistency with the previous sampling programme. However, in areas where deposition might be expected, additional 45 cm cores were also collected and sectioned into 15 cm depth increments, in order to confirm that no ^{137}Cs content was detectable below the depth of 15 cm (Porto *et al*., 2014).

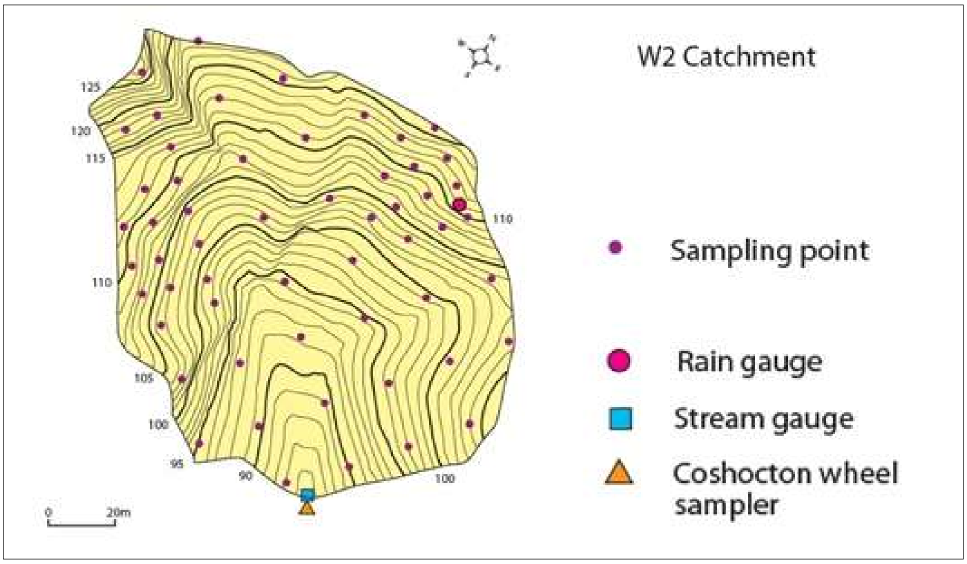


Fig. 3.12 – The sampling points within the W2 catchment.

Soil sampling for ^{137}Cs analysis associated with catchment W3 encompassed two separate sampling programmes. The first was designed to establish the magnitude and spatial distribution of erosion rates within the catchment and involved two campaigns. During the first campaign, undertaken in 1999, 47 bulk soil cores were collected at the intersections of an approximate 20 m x 20 m grid using the procedure outlined above. These cores were supplemented by a further 34 bulk cores collected from sites selected to improve the coverage of topographic variability during an additional campaign in 2001. The second sampling programme, undertaken in 2001, aimed to establish the depth distribution of ^{137}Cs at undisturbed reference sites with minimum slope. One such site was located within a small clearing between the trees (Reference site 2). Because of the difficulty of locating another reference site within the catchment, a second site was selected at a similar altitude to the catchment within an adjacent area of undisturbed rangeland with some scattered oaks (Quercus pubescens) (Reference site 3). In both cases sampling was undertaken using a 652 cm^2 surface area scraper plate and the procedure outlined above. As before, six additional bulk cores were collected in the immediate vicinity of each site in order to confirm the representativeness of the inventory derived from the scraper plate samples (Porto *et al.*, 2003).

On July 2015, this catchment was sampled again and the same sampling points used during the field campaign undertaken in 1999 and 2001 were revisited. Each site was sampled again using a steel core tube (10 cm diameter) driven into the ground by a motorized percussion corer to a depth of ca. 30 cm (Fig. 3.13).

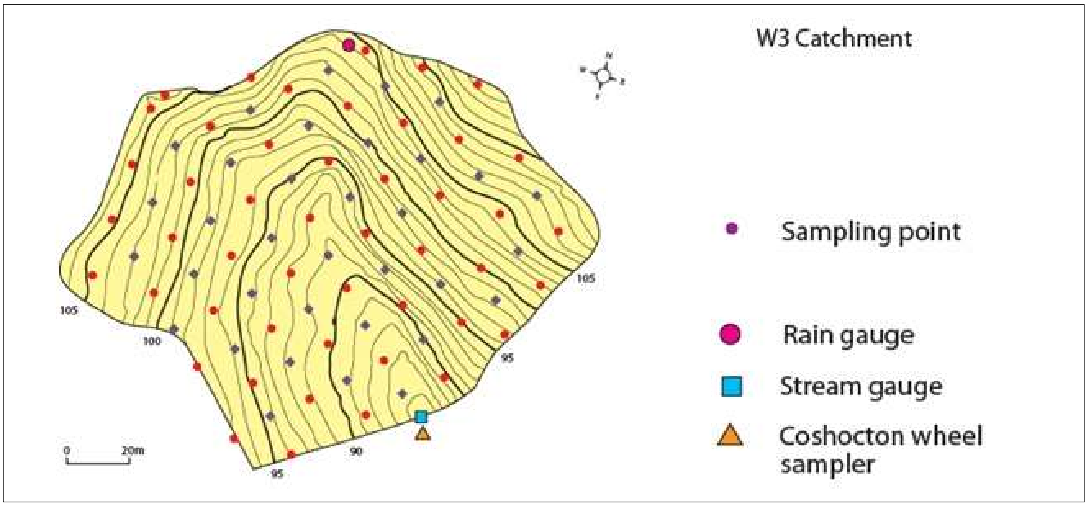


Fig. 3.13 - The sampling points within the W3 catchment.

3.4 Laboratory analyses

3.4.1 Sample preparation and pre-treatment

Based on the protocol proposed (Walling & Quine, 1993; Pennock & Appleby, 2002) sample preparation has been involved the following steps:

a) each soil sample is firstly oven dried at 105°C for 48 hours;
b) the weight of the dried sample is recorded;
c) the sample is lightly ground or hand disaggregated. It is important to disaggregate mineral particles, organic concretions and any porous material;

d) the sample is passed through a 2 mm sieve to separate soil particles (<2 mm) from the coarser rock fragments (>2 mm). The bulk density of the soil sample (fine fraction) is accurately calculated for determining the ^{137}Cs areal activity or inventory;

e) a representative of <2 mm fraction of each samples is then taken and packed for analysis. It is placed in a 500 g "Marinelli" beaker, or in the case of smaller samplers a plastic dish (65 mm diameter, 15 mm high) or a plastic pot (100 g), in order to measures its activity by gamma spectrometry.

3.4.2 Gamma spectrometry measurements

Gamma spectrometry using germanium detectors (HPGe or Ge-Li) is an excellent analytical technique for measuring low level activities of natural (e.g. ^{226}Ra, ^{210}Pb, ^{40}K, etc.) or artificial radionuclides (e.g. ^{137}Cs, ^{60}Co, etc.) in environmental samples (e.g. soil, sediment, vegetation, water, etc.). The method allows both qualitative and quantitative determination of the radionuclides directly in the original sample, through the detection of gamma rays (photons), without the need for chemical separations. The simultaneous detection of several gamma emitters in the same sample material is possible using a shielded high resolution germanium semiconductor detector connected to a multichannel analyzer (MCA). The interaction of gamma rays (photons) with the germanium crystal produces electric signals corresponding to the energy of the incoming photons. Signals are amplified and transmitted to the MCA. A gamma energy spectrum, characteristic of the gamma-emitting nuclides is produced. This is the result of the three effects (Photoelectric, Compton and Pair production effects) due to the interaction of gamma rays with the germanium crystal. The photoelectric effect is characterized by the presence of high resolution peaks (called fullenergy peaks) associated with the γ-rays emitted by the radionuclides while the Compton effect is characterized by a continuum due to the interaction of the gamma rays with electrons of the atom. The conversion of the net peak area (counts per second) of the nuclide of interest, associated with the Photoelectric effect on activity (Bq kg^{-1}) as well as the processing of the overall collected spectral data are performed using a selected software package (Mabel *et al.*, 2014b).

The different components of a gamma ray spectrometry system can be described as follows (see also Fig. 3.14):

(a) *A high purity germanium detector (HPGe) with cryostatic cooling*: A liquid nitrogen Dewar or electrically powered cryostat will be needed. The performance of the detector is given by its relative efficiency, its energy resolution and the Peak-to-Compton ratio. The relative efficiency is a measure relative to that of a 75 x 75 mm NaI (sodium iodide) crystal and is normally based on the measurement of the 1.33 MeV peak of ^{60}Co. Its value depends on the crystal volume and the detector configuration. The energy resolution of the detector which is considered as the full energy peak at half maximum (FWHM) of the full energy peak of 1.33 MeV peak should be between 1.8 and 2.2 keV. The Peak-to-Compton ratio is the ratio of counts in the highest photo-peak channel to the counts in a typical channel just below the associated Compton edge and is conventionally quoted for the 1.33 MeV gamma ray photo-peak of ^{60}Co;

(b) *Shielding*: A detector shield with a cavity large enough to accommodate samples (up to 2 litres) will be needed. It is generally constructed of lead. The wall thickness is usually 10 cm in order to attenuate high-energy photons from all external sources. In addition, to attenuate lead photons coming from the lead shield, it is recommended to install inside the lead shielding a lining of either copper or steel with a thickness of 10 mm;

(c) *A preamplifier*: This is generally an integral part of the detector unit which takes the charge produced from the detector, integrates and amplifies this to produce a pulse, the amplitude of which is proportional to the total energy. The amplifier is located near the detector in order to take advantage of the cooling which is necessary for the operation of the detector with low noise;

(d) *A high voltage power supply*: A bias voltage is applied across the detector volume to collect created charges within the germanium detector. This is usually in the range of ± 1000 to 5000 V;

(e) *A linear amplifier*: This primarily takes the pulse signal from the preamplifier and considerably amplifies it. It also filters and shapes the incoming pulse to enhance the signal-to-noise ratio. High resolution spectroscopy amplifiers with a pile-up rejection and life-time correction are now commercially available;

(f) *An analogue to digital converter (ADC).* The detector produces an analogue signal which is shaped by the amplifier. The ADC converts this analogue signal to a digital signal and can usually only process one incoming pulse at a time (from one detector). When several detectors are used, it is advisable to use a multiplexer or mixer-router which takes the separate counting chains from several individual inputs and routes them through to a single ADC;

(g) *A multichannel analyser (MCA)*. Usually incorporated into the computer system, an MCA registers the pulses emerging from the ADC in one of its channels according to their amplitudes i.e. the energies of gamma rays. It can operate with a minimum of 1024 channels and a maximum of 8192 channels. The number of channels usually used is 4096.

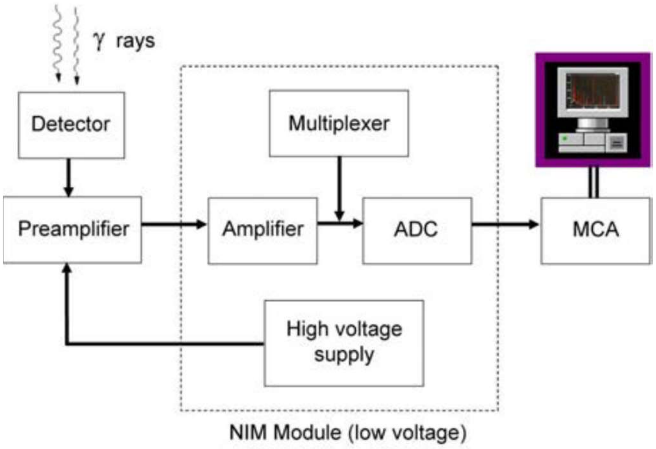


Fig. 3.14 - The electronic components of a gamma spectrometry system.

3.4.3 Laboratory procedures

Measurements of Caesium-137 activity were made on all the samples collected from the three catchments. Each sample was analyzed for ^{137}Cs radioactivity using a gamma spectroscopy. Then the ^{137}Cs radioactivity for each sample was converted to determine the ^{137}Cs inventory for each point.

For the W1 catchment, Caesium-137 activities of the 68 samples were measured using high resolution HPGe detectors in the Laboratories of Geography of the University of Exeter (UK). Count times were typically approx. 80 000 s, providing an analytical precision of about ±10% at the 95% level of confidence. The ^{137}Cs activities in the samples were obtained from the counts at the 662 keV peak in the measured γ-ray spectrum. In 2014 the same 68 sampling point was sampled again and their ^{137}Cs activity was measured using high resolution HPGe detectors provided by the Department of Agriculture of University "Mediterranea" of Reggio Calabria. The ^{137}Cs activities in the samples were obtained from the counts for the 662 keV peak and the count times were typically ~80 000 s, providing results with an analytical precision of ca. ±10% at the 95% level of confidence.

For the W2 catchment, the ^{137}Cs activity of the first set of samples consisting of the 55 replicate cores collected during the sampling campaign undertaken in 1998 was measured using a high resolution HPGe detector in the Laboratory of the Department of Nuclear Engineering at the University of Palermo, Italy. Count times were ca. 30,000 s, providing a precision of ca. ±10% at the 95% level of confidence. The second set of samples collected from the catchment in 1999, which included both the depth incremental samples provided by the scraper plate and the sectioned cores collected from the reference area, was analysed using a high resolution HPGe detector in the Laboratory of the Department of Geography at the University of Exeter, UK.

Count times were again ca. 30,000 s, providing a precision of ca. ±10% at the 95% level of confidence. The third set of samples, collected in 2013, during the repeat-sampling campaign, was analysed using high resolution HPGe detectors in the Department of Physics at the University of Basel and the State Laboratory Basel-City, Switzerland. Because of the expected lower activity of these samples, counting times were increased to ca. 80,000 s providing a precision of ca. ±10% at the 95% level of confidence. The total inventory or areal activity density (Bq m^{-2}) of each bulk core was calculated as the product of the measured ^{137}Cs activity (Bq kg^{-1}) and the dry mass of the <2 mm fraction of the bulk core (kg), divided by the surface area of the core or cores associated with the sample (m^2) (Porto *et al.*, 2014).

For the W3 catchment, Caesium-137 activities were measured using high resolution HPGe detectors in the Laboratories of the Department of Nuclear Engineering at the University of Palermo (Italy) and the Department of Geography of the University of Exeter (UK). Count times were typically c. 30000 s, providing a precision of ca. 10% at the 95% level of confidence (Porto *et al.*, 2003). In the re-sampling campaign undertaken in 2015, the samples were analysed using high resolution HPGe detectors in the Department of Agriculture of University "Mediterranea" of Reggio Calabria and the same analysis process was followed as above.

3.4.4 Particle size analysis

The size distribution of the soil samples was determined using a size analyzer named Laser Particle Sizer *analysette 22* provided by the Department of Agriculture of University "Mediterranea" of Reggio Calabria. A standard procedure for preparing soil samples for absolute particle size analysis is as follows (Walling & Collins, 2000):

a) 5 g of soil is weighed into a beaker;

b) 10 ml of distilled water is added to each sample + 5 ml of Hydrogen peroxide; this reaction may need to be controlled by adding a few drops of IMS (Industrial Methylated Spirit);

c) after 2-3 hours, if frothing has ceased, a further 5 ml of Hydrogen peroxide is added and the sample is allowed to stand overnight;

d) the beakers are warmed on a hotplate (starting the process at 80 °C and gradually increasing the temperature to 100 °C) until the reaction is complete and there is a clear supernatant;

e) the whole contents of the beaker are transferred to a centrifuge tube, using a glass rod to clean the sides of the beaker;

f) the tubes are centrifuged at 2500 rpm for 1 hour;

g) the supernatant is discarded and the samples are washed with 2 ml of IMS, before being recentrifuged at 2500 rpm for 30 minutes;

h) the supernatant is discarded and the contents of the centrifuge tube are transferred to a container, using as little distilled water as possible;

i) as soon as the analytical instrument has been set up for use, 10 ml of Sodium hexametaphosphate (6.7 g Sodium hexametaphosphate and 1.3 g Sodium carbonate to 2 litres with distilled water) is added to each sample, which is then subjected to ultrasonic dispersion for ca. 2 minutes;

j) finally, the samples are then analyzed for their grain size composition.

The Laser Particle Sizer *analysette 22* (Fig. 3.15) is an all-purpose measuring device that can be used to determine the particle size distribution of solids in either fluids or gases (suspensions, aerosol) or in drops of liquid (emulsions). The *analysette 22* is controlled using the MaScontrol software. The exact measurement process is defined using SOPs (Standard Operating Procedures), which permit standardized performance of measurements for frequently recurring sample systems under identical analysis conditions. The analysis of the measurements as well as graphical display of the results can be predefined using templates, allowing standardised reports containing the specific values of interest to be generated as well. MaScontrol is a database-supported program. In other words, all measurements, SOPs, directories and results folders or reports are saved in an SQL database that allows for simple, clear and efficient access to all data.

Fig. 3.15 – The Laser Particle Sizer *analysette 22*.

Laser light encountering particles is deflected from its original direction and scattered at specific angles determined by the size and optical properties of the particles. The scattered light is collected with the help of what is termed a Fourier lens and the angle-dependent intensity distribution of the scattered light is measured by a sensor within the focal plane of the lens. Using the Fourier spectrum obtained in this way, the particle size distribution is calculated in a kind of backward process making use of inversion. Through the application of special algorithms, MaScontrol permits an analysis according to either Fraunhofer or Mie theory (See Fig. 3.16).

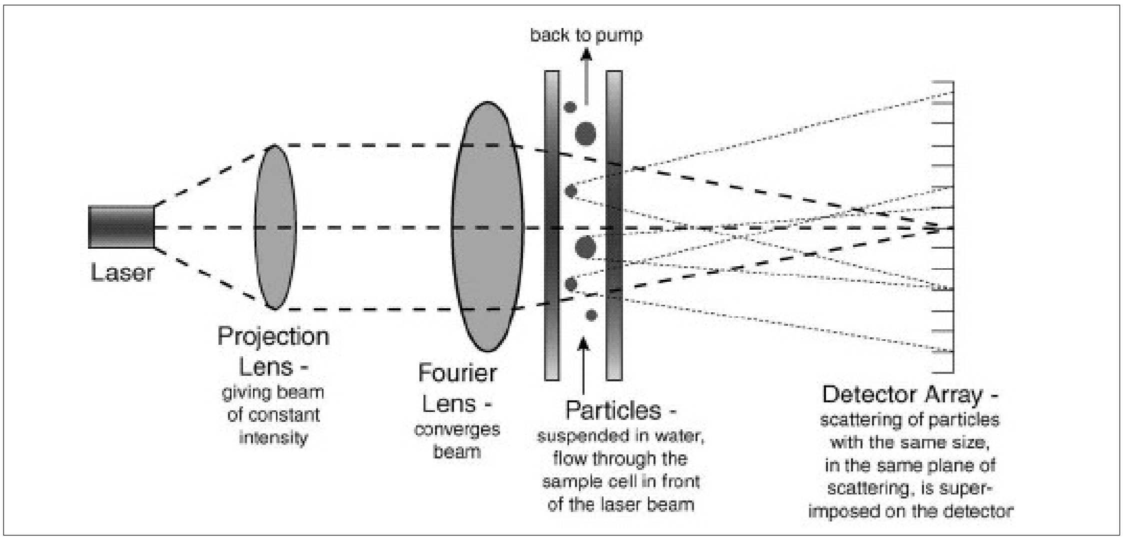


Fig. 3.16 – Example of Fraunhofer or Mie theory.

In other words, when an expanded parallel laser beam illuminates a collection of particles, a specific pattern is projected onto the focal plane of a convex lens situated further down the beam path. The scattered light pattern is the Fourier transform of the diffracting object and clearly describes the shape and size of the particles. Using an appropriate detector for measuring the pattern, a suitable physical model and the right mathematics for solving the resulting system of equations, one arrives at the particle size distribution.

The convex lens is also called a "Fourier transform lens" and is adapted to a single, precisely defined measurement range. As a result, changing of the measurement range also requires changing of the lens. It applied a known fact of Fourier optics to particle measurement and showed that the Fourier transform of the diffracting object can also be obtained by placing a convex lens in front of the object within a convergent laser beam ("reverse Fourier optics"). Moving the measurement cell (see Figg. 3.17 and 3.18) causes an effect similar to a zoom function in the Fourier plane. By simply moving the measurement cell along the laser beam, the measurement range of the *analysette 22* can be optimised and individually adapted to the sample being measured.

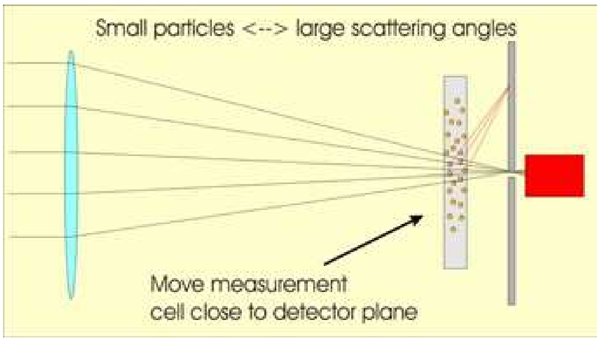


Fig. 3.17 - The position of the sample cell between the Fourier lens and the sensor for the fine measuring range.

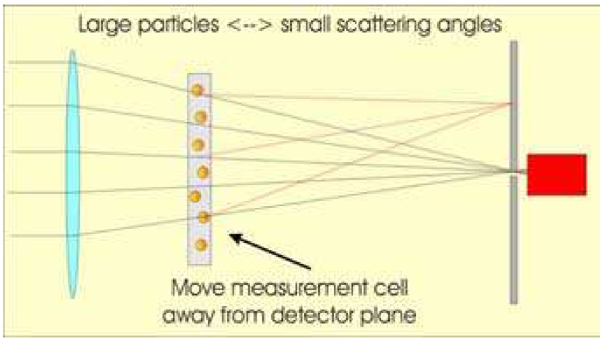


Fig. 3.18 - The position of the sample cell between the Fourier lens and the sensor for the coarse measuring range.

The position of the sample cell between the Fourier lens and the sensor determines the detected particle size range. For the fine measuring range, the measuring cell is located only a few millimetres from the focal point of the Fourier lens, while large measuring cell to detector distances are used for taking measurements in the coarse range.

The particle-size distribution determines the coarseness or the fineness of soils. Soil separates are the size groups of mineral particles less than 2 mm in diameter or the size groups that are smaller than gravel (Foth, 1990). According to the WRB (World Reference Base for soil resources) classification, it’s possible to identify the following classes size: clay, silt, very fine sand, fine sand, medium sand, coarse sand and very coarse sand. The diameters of each separate are showed in Tab. 3.2.

Tab. 3.2 – The diameter limits of Soil Separates.

Name of soil separate	Diameter limits (mm)
Clay	< 0.002
Silt	0.002 – 0.063
Very fine sand	0.063 – 0.125
Fine sand	0.125–0.20
Medium sand	0.20–0.63
Coarse sand	0.63–1.25
Very coarse sand	1.25-2.00

The largest separates are sand particles and are the 2.0 to 0.063 millimeters fraction. Furthermore, large sand particles can be described as very coarse and coarse, intermediate as medium, and the smaller as fine. The next smallest particles are silt particles and have diameters between 0.063 mm and 0.002 mm. At the 0.063 mm particle size separation, between sand and silt, it is difficult to distinguish by feel the individual particles. In general, if particles feel coarse or abrasive when rubbed between the fingers, the particles are larger than silt size. Silt particles feel smooth like powder. Neither sand nor silt is sticky when wet. Sand and silt differ from each other on the basis of size and may be composed of the same minerals, indeed, the sand and silt separates of many soils are dominated by quartz. There is usually a significant amount of weatherable minerals, such as feldspar and mica, that weather slowly and release ions that supply plant needs and recombine to form secondary minerals, such as clay. The greater specific surface (surface area per gram) of silt results in more rapid weathering of silt, compared with sand, and greater release of nutrient ions. The result is a generally greater fertility in soil having a high silt content than in soils high in sand content (Foth, 1990).

The smallest separates are clay particles with diameters of less than 0.002 mm. Clay particles tend to be plate-shaped, rather than spherical, and very small in size with a large surface area per gram. Because the specific surface of clay, is many times greater than that of sand or silt, a gram of clay adsorbs much more water than a gram of silt or sand, because water adsorption is a function of surface area. Their plate-shaped nature contributes to very large specific surface. Films of water between plate-shaped clay particles act as a lubricant to give clay its plasticity when wet. Conversely, when soils high in clay are dried, there is an enormous area of contact between plate-shaped soil particles and great tendency for very hard soil clods to form. Although the preceding statements apply to most clay in soils, some soil clays have little tendency to show stickiness and expand when wetted. The clay fraction usually has a net negative charge. The negative charge adsorbs nutrient cations, including Ca^{2+} Mg^{2+} and K^{+}, and retains them in available form for use by roots and microbes (see Foth, 1990).

Soil texture refers to the weight proportion of the separates for particles less than 2 mm as determined from a laboratory particle-size distribution (USDA,1993). In other words, soil textures are classified by the fractions of each soil separate (sand, silt and clay) present in a soil. Once the percentages of sand, silt, and clay have been determined, the soil can be placed in one of 12 major textural classes, as showed in Fig. 3.19. Each name classification derived from the primary constituent particle size or a combination of the most abundant particles sizes.

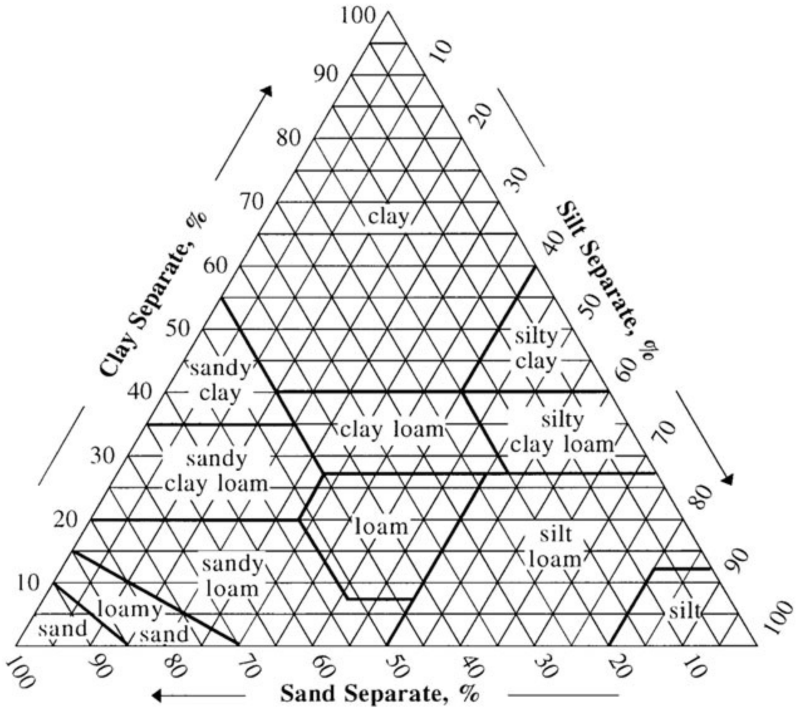


Fig. 3.19 - The textural triangle shows the limits of sand, silt, and clay contents of the various texture classes.

3.5 Conversion model to estimate soil redistribution rates from ^{137}Cs measurements

Two theoretical conversion models applicable to uncultivated soils can be used to derive estimates of soil erosion rates within the three study catchments from the ^{137}Cs measurements. These models are commonly referred to as the exponential profile distribution model and the diffusion and migration model (Walling and Quine, 1990; Walling and He, 1999a). Both models use information on the ^{137}Cs inventory from the depth distribution at the reference site and from the ^{137}Cs inventory of the sampling points within the study area. The profile distribution model assumes an exponential ^{137}Cs depth distribution and the depth of soil removed by erosion over the period covered by the ^{137}Cs measurements is estimated by establishing the reduction in the ^{137}Cs inventory, relative to the reference inventory, and calculating the depth of soil which would need to be removed to produce the calculated reduction in inventory. One limitation of the depth distribution model is that the exponential depth distribution is assumed to be time-invariant over the period covered by the ^{137}Cs measurements. In reality the inventory will have increased over the period of fallout and the depth distribution can be expected to have changed through time as a result of downward diffusion and migration of the ^{137}Cs fallout input to the soil surface. The diffusion and migration model takes account of the time dependent record of fallout input and the evolution of the ^{137}Cs depth distribution through time in response to downward diffusion and migration (Porto *et al.*, 2003). Further details of the two conversion models are given below.

The first of these models is the *Profile Distribution Model*. It is the simplest and take into account the erosion rate at a given sampling point estimated by considering the reduction in inventory relating to the reference inventory. This model is mainly used to estimate soil loss but estimates of deposition can be obtained for sampling points too. Porto *et al.* (2001) employed this model to estimate soil redistribution rates in the study catchment based on the samples collected in 1998 and 1999. That investigation proved a similarity between the measured sediment yield from the catchment and the estimated mean annual net soil loss and. However, one potential limitation of the Profile Distribution Model is that it assumes that the ^{137}Cs fallout occurred as a single input in 1963 and that the depth distribution documented at the reference site was in existence from that time. In reality, the depth distribution will have been time-dependent, in response to the temporal pattern of ^{137}Cs fallout and the ongoing post-fallout redistribution of the ^{137}Cs fallout in the soil. The latter would have resulted in a slow downward movement. In addition, during the period of active fallout fresh fallout can be rapidly removed by erosion before moving down into the soil. In contrast, when fallout ceases and the ^{137}Cs moves progressively deeper into the soil, removal of a given amount of ^{137}Cs will involve removal of a greater amount of soil. As a result, the Profile Distribution Model is likely to overestimate the mean annual rate of soil loss (Porto *et al.*, 2014). This limitation assumes more importance in a re-sampling study where two overlapping periods are considered. In fact, even if actual erosion rates during the entire period were to be essentially constant, the mean annual erosion rate estimated for the longer period is likely to be lower than that for the shorter period. This could potentially prompt misleading conclusions concerning changes in soil redistribution rates.

The second conversion model to estimate soil redistribution rates for areas with uncultivated soils is the *Diffusion and Migration Model* (see Walling and He, 1999a; Walling *et al.*, 2002). For the three study catchments, this conversion model was used to derive estimates of soil erosion and redistribution rates from the ^{137}Cs measurements because it overcomes the limitations of the Profile Distribution Model summarized above. This model takes account of the temporal distribution of fallout input, the progressive post-depositional redistribution of ^{137}Cs in the soil profile and the contrast in the interaction between fallout and soil loss associated with periods of active fallout and later periods with no new fallout.

According to Walling and He (1993), by treating the soil as a semi-infinite homogeneous porous medium, so that it can be characterized by a constant effective diffusion coefficient and a constant migration rate, and assuming that fallout inputs are initially uniformly distributed at the soil surface, the vertical distribution of ^{137}Cs concentration from the surface downwards can be represented by the following partial differential equation (Crank, 1975):

$$\frac{\partial C(x,t)}{\partial t} = D\frac{\partial^2 C(x,t)}{\partial x^2} - V\frac{\partial C(x,t)}{\partial x} - \lambda C(x,t) \tag{3.1}$$

where:

D = the effective diffusion coefficient (kg^2 m^{-4} yr^{-1});
V = the downward migration rate (kg m^{-2} yr^{-1});
λ = the decay constant for ^{137}Cs (0.023 yr^{-1});
x = the mass depth from the soil surface downwards (kg m^{-2});
t = the time since the first deposition of ^{137}Cs (years);
$C(x,t)$ = the activity of ^{137}Cs at time t and cumulative mass depth x (Bq kg^{-1}).

The initial condition for equation (3.1) for an instantaneous input $I(t')$ can be approximated by the following equation (He - unpublished thesis, University of Exeter - 1993):

$$C_i(x,t') = \frac{I(t')}{H}e^{-x/H} \tag{3.2}$$

where:

$I(t')$ = a pulse input (Bq m^{-2} yr^{-1}) at time t'
H = the relaxation depth expressed as a mass depth (kg m^{-2}).

If the post-depositional movement of ^{137}Cs in the soil is restricted to the soil profile itself, mass conservation requires the following boundary conditions for equation (3.1) for an initial input $I(t')$:

$$\text{-}D\frac{\partial C(x,t)}{\partial x}\Big|x = 0 + VC(x,t)\Big|_{x=0} = 0$$

$$\lim_{x\to\infty}\frac{\partial C(x,t)}{\partial x} = 0 \tag{3.3}$$

A solution $C(x,t,t')$ of equation (3.1) for an undisturbed soil, for any cumulative mass depth and time under initial condition equation (3.2) and boundary conditions equation (3.3), and after t–t', can be expressed (Lindstrom and Boersma, 1971; Officer and Lynch, 1982; Walling and He, 1993) as:

$$C(x,t,t') = e^{-\lambda(t-t')}\int_0^\infty \frac{I(t')}{H}e^{-y/H}\left\{e^{\frac{V(x-y)}{2D}-\frac{V^2(t-t')}{4D}}\left[e^{-\frac{(x+y)^2}{4D(t-t')}} + e^{-\frac{(x-y)^2}{4D(t-t')}}\right]\right.$$

$$\times \frac{1}{\sqrt{4\pi D(t-t')}} - \frac{V}{2D} e^{\frac{Vx}{D}} erfc\left[\frac{x+y+V(t-t')}{\sqrt{4D(t-t')}}\right]\Bigg]\Bigg\} dy \tag{3.4}$$

where *erfc(u)* is the error-function complement defined as (Crank, 1975):

$$erfc(u) = \frac{2}{\sqrt{\pi}} \int_{u}^{\infty} e^{-y^2} dy \tag{3.5}$$

Taking account of a continuous input *I(t')*, the ^{137}Cs concentration distribution *C(x,t)* (Bq kg^{-1}) in the soil profile at time t can be obtained by integrating *C(x,t,t')* over time *t'* (Walling and He, 1993):

$$C(x,t) = \int_{0}^{t} C(x,t,t')dt \tag{3.6}$$

The ability of equations (3.4) and (3.6) to model the ^{137}Cs concentration distribution in the soil profile has been demonstrated elsewhere (Walling and He, 1993; He and Walling, 1997).

For a soil affected by sheet erosion, with a constant annual erosion rate *R* (kg m^{-2} yr^{-1}), the concentration of ^{137}Cs in the eroded sediment $C_u(t)$ (Bq kg^{-1}), assuming no enrichment, may be approximated by that of the surface soil $C_u(0, t)$ obtained from equation (3.6). According to Walling and He (1999), the erosion rate *R* may be estimated from the reduction in the ^{137}Cs inventory $A_{u,ls}(t)$ (Bq m^{-2}), defined as the ^{137}Cs reference inventory A_{ref} less the measured total inventory A_u, and the ^{137}Cs concentration in the surface soil $C_u(t')$ according to:

$$A_{u,ls}(t) = \int_{0}^{t} R C_u(t') e^{-\lambda(t-t')} dt' \tag{3.7}$$

The same authors tested the validity of equations (3.6) and (3.7), with a wide range of values assumed for the parameters *D*, *V*, and *H*. This exercise (Walling and He, 1999) showed that the predicted erosion rates are positively related to these parameters, although the model is more sensitive to *D* than to *V* and *H* (Porto *et al.*, 2003).

According to Walling and He (1999), equations (3.4) and (3.6) are applicable only to soils where the erosion rate is relatively low. In this situation, the concentration of ^{137}Cs in the eroded sediment $C_u(t)$ (Bq kg^{-1}) can be approximated by that of the surface soil $C_u(0, t)$ obtained from equation (3.6) and erosion rates can be estimated using equation (3.7). Where erosion rates are higher the concentration of ^{137}Cs in the eroded sediment $C_u(t)$, cannot be approximated by that of the surface soil obtained from equation (3.6), since the decline in concentration with depth must be taken into account (Porto *et al.*, 2003).

Assuming a constant rate of lowering of the surface by erosion E (kg m^{-2}) and diffusional transport, the vertical distribution of ^{137}Cs within a soil (equation 3.4), for any cumulative mass depth and time t', can be expressed as:

$$C_e(x,t,t') = \mathrm{e}^{-\lambda(t-t')} \int_0^{\infty} \frac{I(t')}{H} e^{-\frac{y}{H}} \left\{ \left[e^{-\frac{[(x+E)+y]^2}{4D(t-t')}} + e^{-\frac{[(x+E)-y]^2}{4D(t-t')}} \right] \frac{1}{\sqrt{4\pi D(t-t')}} \right\} \mathrm{d}y \tag{3.8}$$

where:

$C_e(x,t,t')$ = the concentration of ^{137}Cs for any cumulative mass depth x and time t' (Bq kg^{-1});

D = the effective diffusion coefficient (kg^2 m^{-4} y^{-1});

H = the relaxation depth of the initial distribution of fallout expressed as a mass depth (kg m^{-2});

λ = the decay constant for ^{137}Cs (0·023 y^{-1});

x = the mass depth from the soil surface downwards (kg m^{-2});

t = the time since the first deposition of ^{137}Cs (years);

$I(t')$ = the annual fallout deposition flux (Bq m^{-2} y^{-1}) at time t'.

The ^{137}Cs concentration $C_e(x,t)$ (Bq kg^{-1}) in the soil profile at time t can be obtained by integrating $C_e(x,t,t')$ over time t':

$$C_e(x,t) = \int_0^t C_e(x,t,t')dt \tag{3.9}$$

Integration of $C_e(x,t)$ over mass depth x gives the total ^{137}Cs inventory A_u (Bq m^{-2}) for an eroding site at time t:

$$A_u(t) = \int_0^{\infty} C_e(x,t)dt \tag{3.10}$$

Assuming a constant value of H (5 kg m^{-2}), as suggested by Walling and He (1999a), equations (3.8) and (3.10) can be solved simultaneously for E (kg m^{-2}), with A_u (Bq m^{-2}) representing the measured inventory corresponding to an eroding point. The erosion rates R (kg m^{-2} yr^{-1}) may then be estimated by dividing the quantity E by the time t–t' (years) since the commencement of ^{137}Cs fallout (Porto *et al.*, 2014).

For a depositional site, the deposition rate D_R can be estimated from the ^{137}Cs concentration in deposited sediment $C_d(t')$ and the excess ^{137}Cs inventory (defined as the total measured ^{137}Cs inventory A_u less the local reference inventory A_{ref}) using the following relationship:

$$D_R = \frac{A_u - A_{ref}}{\int_{t_0}^{t} C_d(t') e^{-\lambda(t-t')} dt'} \tag{3.11}$$

Assuming that the ^{137}Cs concentration $C_d(t')$ of deposited sediment can be represented by the weighted mean of the ^{137}Cs concentration of the sediment mobilized from the upslope contributing area, $C_d(t')$ can be calculated (Porto *et al.*, 2014) as:

$$C_d(t') = \frac{1}{\int_S R dS} \int_S C_e(t') R dS \tag{3.12}$$

where:

S (m^2) = the upslope contributing area

$C_e(t')$ = the ^{137}Cs concentration of sediment mobilized from an eroding point (Bq kg^{-1}), which can be calculated from equation (3.8), assuming $x = 0$.

3.6 Particle size correction factors *P* and *P'* for selectivity of erosion and deposition processes

The particle size factors are particularly important as water erosion/deposition triggers a selective and preferential loss/increase of small grain size fractions. Usually reference sites show the original grain size composition, while eroded sites are depleted in small particle fractions. Small grain size fractions in soil samples tend to be enriched in ^{137}Cs concentrations and might be preferentially transported during sheet erosion processes (see IAEA, 2014). To take into account the preferential transport of smaller grain size particles with higher specific surface area, and thus higher ^{137}Cs content, a particle size correction factors (*P* or *P'* in case of deposition) is usually proposed by most conversion models. Since information on the ^{137}Cs concentration in soil mobilized by erosion and deposition with the sediments may be very difficult to obtain, estimates of these parameters can be deduced from the information obtained on the grain size composition of the surface soil, eroded sediment and deposited sediment (Arata *et al.*, 2016).

As described in §2.6, because the grain size composition of mobilized sediment is usually enriched in fines compared with the original soil, the correction factor is generally great than 1.0, due to the strong affinity of ^{137}Cs for fine soil particles. Its value is therefore a function of the grain size composition of both mobilised sediment and the original soil. For a depositional site, the correction factor is a function of the ratio of the ^{137}Cs concentration of deposited sediment to that of the mobilised sediment. Because the grain size composition of deposited sediment is frequently depleted in fine fractions compared with the mobilised sediment, the value of the correction factor is generally less than 1.0 (Walling et al., 2006).

In this study, several analyses for all soil samples have been conducted to investigate the transport of the different grain size due to soil erosion/deposition processes. The theoretical *Diffusion and Migration Model* applicable to uncultivated soils was used to derive estimates of soil erosion rates within the three study catchments from the ^{137}Cs measurements. For many of the analysis, the eroded soil has a similar grain size composition to the surface soil, being slightly coarser for some soil samples and slightly finer for others. However, the size distribution of the sediment eroded from the three catchments is consistently coarser than that of the surface soil. Thus, a particle size correction factor of 1 for the selected conversion model was assumed and also the *Diffusion and Migration Model* with no correction factor was used. It's important to compare both the measurements because many workers have highlighted the need to take account of the grain size selectivity of erosion processes when using ^{137}Cs measurements to estimate rates of soil redistribution, since contrasts in grain size composition of eroded sediment relative to the parent soil will influence its ^{137}Cs content (e.g. Sutherland, 1991; Walling and Quine, 1990; Walling and He, 1999a, 2001).

Results and Discussion

4.1 The results of the ordinary sampling and sediment yield at the catchment outlets

The ordinary sediment sampling campaigns, carried out within the three experimental catchments W1, W2 and W3, consisted of several field activities, most of them devoted to the measurements of rainfall, runoff and sediment yield after each natural storm event. These measurements concerned collection of sediment samples, after each storm event, from the tanks located at the catchments outlet. These measurements allowed to establish the mean sediment concentration in the tank at the event scale. The sediment concentration associated with these samples is determined by oven drying at 105°C. The event sediment yield from the catchments is then calculated as the product of the mean sediment concentration and the total runoff volume for the event measured by the H-flume. These measurements (2013-2015), together with the existing dataset available for the previous monitoring period (1978-2012), served for validating the re-sampling technique proposed in this project.

The whole time window of sediment yield data provided by the monitoring programmes in the three catchments relates to the periods 1978-1994 (Porto *et al.*, 2001) and 2006-2015. These measurements are shown in Fig. 4.1. In the catchment W2, due to some periodic malfunctioning of the sediment sampling equipment, values of annual sediment yield are not available for the years 1981, 1988 and 1989. In the catchments W2 and W3, the inter-annual variation of the values of sediment yield reveal both variations in rainfall intensity and changes in the density of the forest cover associated with forest harvesting and regrowth.

In the catchment W2, the dataset for the first monitoring period contains two outliers recorded in 1990 and 1992 associated with some extreme rainfall events occurred when the trees of the catchment have been cut. During the second monitoring period, three years (2009-2011) are characterized by values with above average sediment yield; it is probably linked to high magnitude rainfall events in those years. The other two catchments do not present significant changes in the sediment dynamics for the first monitoring period; during the second monitoring period catchments W2 and W3 also show an increase of values of sediment yield linked to high magnitude rainfall events.

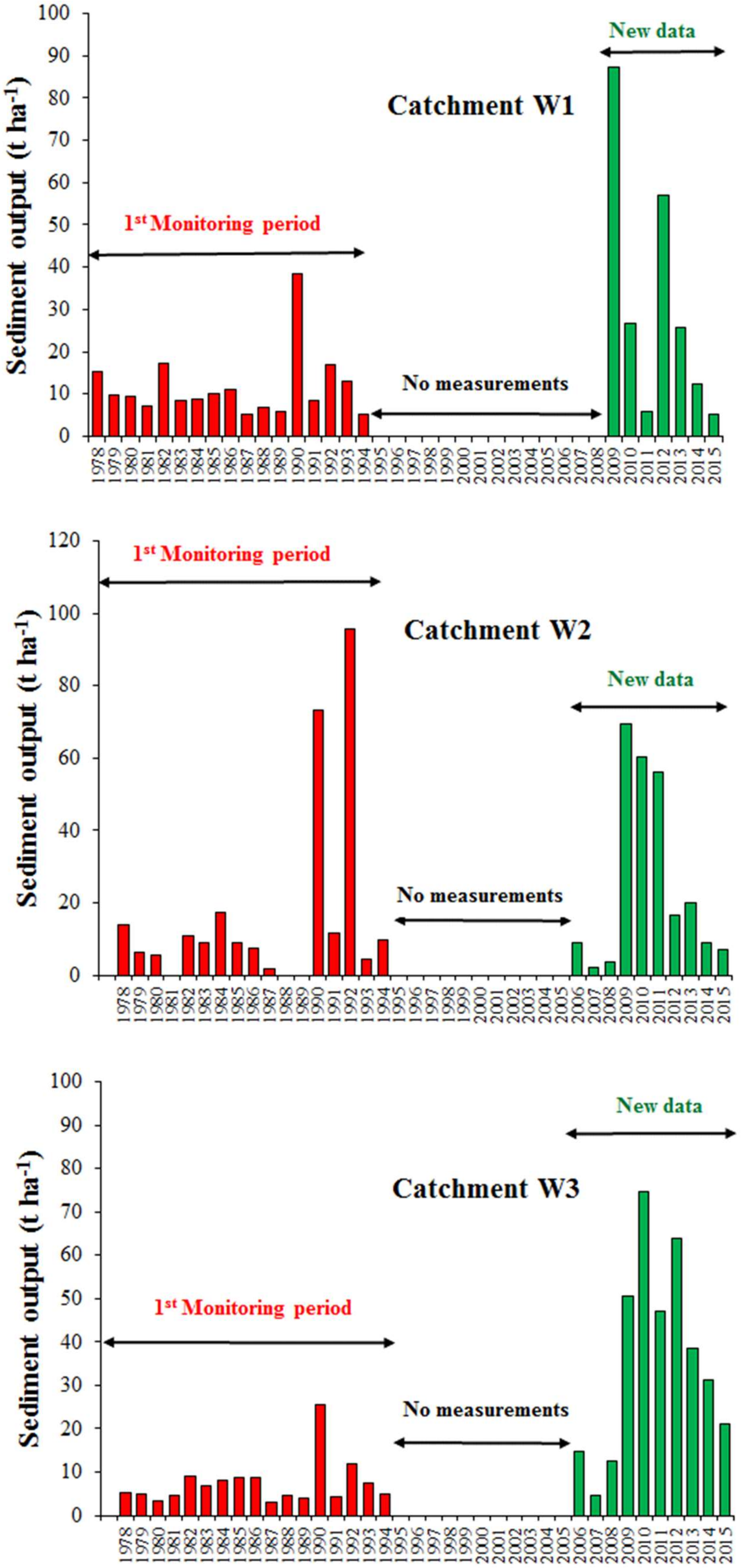


Fig. 4.1 - The records of annual suspended sediment yield available for the three study catchments at the catchments outlet (Porto *et al*., 2016a).

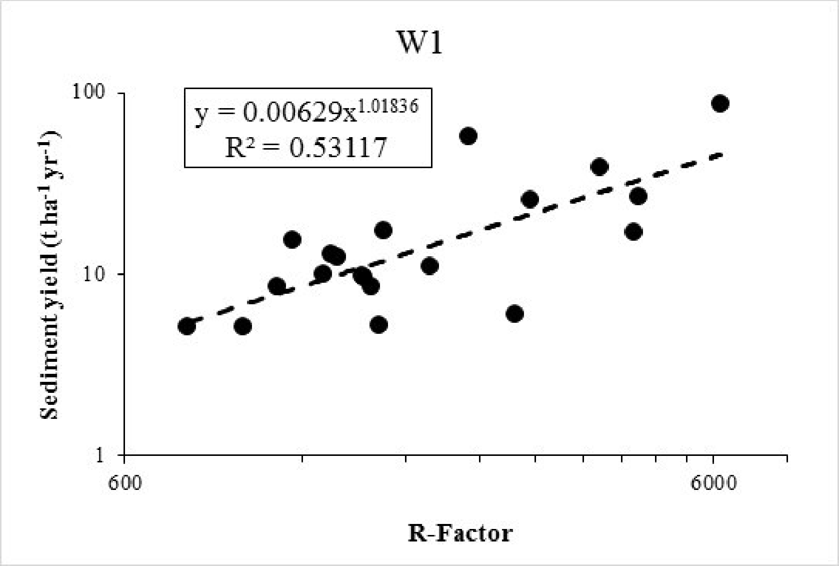
As showed in Tab. 4.1, for the catchment W1, the calculation of the mean annual sediment yield at the catchment outlet for the two periods for which sediment yield data are available gives 11.6 t ha^{-1} yr^{-1} for the period 1978-1994 and 35.9 t ha^{-1} yr^{-1} for the period 2009-2014. For catchment W3, the values of mean annual sediment yield are 7.4 t ha^{-1} yr^{-1} for the period 1978-1994 and 37.5 t ha^{-1} yr^{-1} for the period 2006-2015.

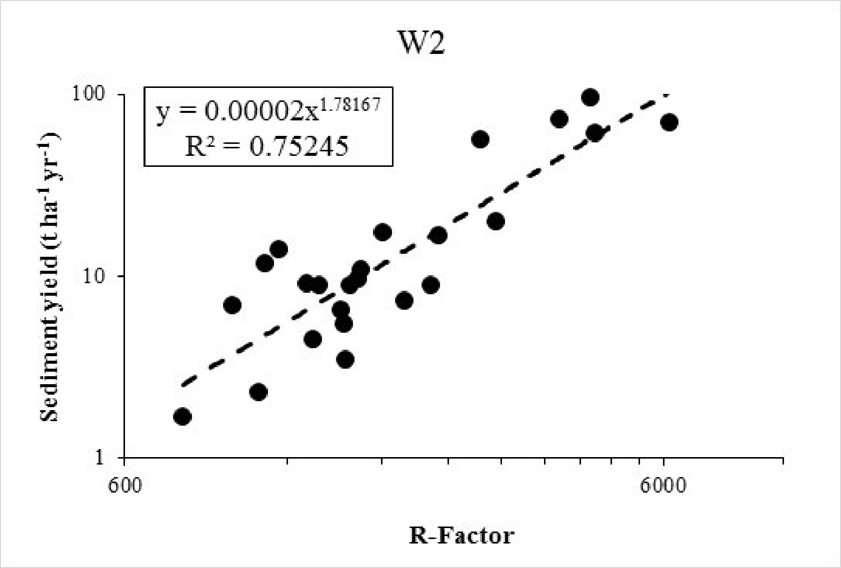
For the catchment W2, calculation of the mean annual sediment yield at the catchment outlet for the first period is complicated by the lack of data for the years 1981, 1988 and 1989. However, annual rainfall in these three years was below average and sediment yields are likely to have been relatively low. The mean annual sediment yield for the period 1978-1994 has therefore been estimated by interpolating values for the missing years based on the annual rainfall and the values of annual sediment yield for adjacent years with a similar rainfall total. Using this approach, the mean annual sediment yield for the period 1978-1994 is estimated to be 17.5 t ha^{-1} yr^{-1}. The equivalent value for the period 2006-2012 is 31.02 t ha^{-1} yr^{-1} (Porto *et al.*, 2014). These mean annual values of sediment yield for catchment W2 have been estimated with a precision of ca. ± 15% at the 95% level of confidence.

Table 4.1: Mean annual sediment yield recorded during the two different monitoring periods from the three study catchments.

Measured values of the mean annual sediment yield at the catchment outlets				
Catchments	First monitoring period		Second monitoring period	
	Years	t ha^{-1} yr^{-1}	Years	t ha^{-1} yr^{-1}
W1	1978-1994	11.6	2009-2014	35.9
W2	1978-1994	17.5	2006-2012	31.2
W3	1978-1994	7.4	2006-2015	37.5

One of the aims of this study is to compare the estimates of mean annual net soil loss from the study catchments, provided by the ^{137}Cs measurements, for the period from 1954 to the year of the first sampling campaign (1998-2001) and for the time window between the two sampling campaigns (from 1999-2001 to 2013-2015), with equivalent sediment yields values. Therefore, the sediment yield data for the three catchments have been extrapolated using information on the values of rainfall erosivity for the missing years to include the period from 1954 to 1998-2001. Like this, a relationship between annual sediment yield and annual rainfall erosivity established using the data for the years when sediment yields were measured has been used. This relationship is reported in Fig. 4.2 for each catchment.





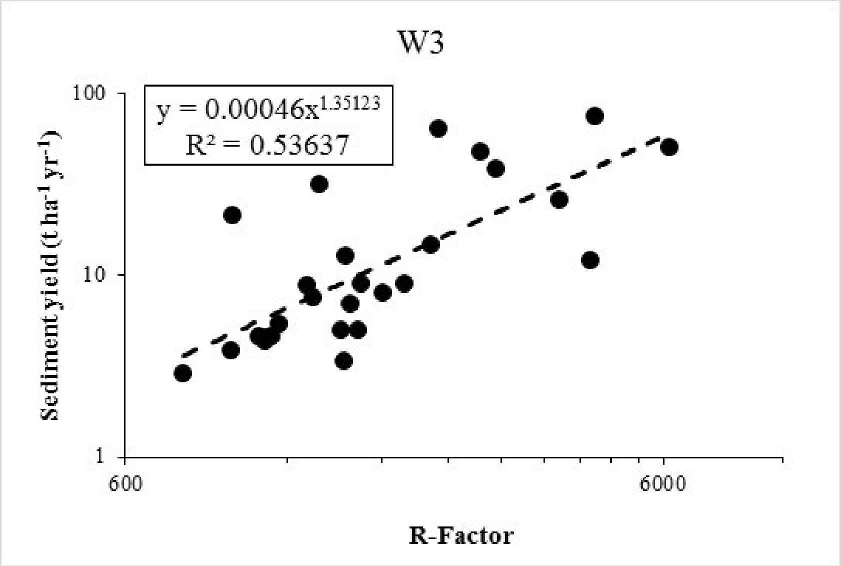


Fig. 4.2 – Relationship between erosivity factor R and the sediment yield for the three catchments.

For catchment W1, the resulting estimates of mean annual sediment yield are 11.6 t ha^{-1} yr^{-1} for the period 1954-2000 and 22.4 t ha^{-1} yr^{-1} for the period 2001-2014. For catchment W2, the estimates are 16.2 t ha^{-1} yr^{-1} for the period 1954-1998 and 20.6 t ha^{-1} yr^{-1} for the period 1999-2013. For catchment W3, the estimates are 9.8 t ha^{-1} yr^{-1} for the periods 1954-2000 and 29.3 t ha^{-1} yr^{-1} for the period 2001-2015 (see Tab. 4.2). The uncertainty attributed to these values includes both the ± 15% related to the methods employed by the measurement programme and the additional uncertainty related to the extrapolation of the data to cover the entire period. For both periods, ca. 50% of the values used to calculate the mean annual sediment yield were extrapolated using the record of annual precipitation and taking account of this and the likely errors introduced by the estimated values, an uncertainty of ca. ± 50% at the 95% level of confidence has been attributed to the resulting values of mean annual sediment yield. When combined the two an estimate of uncertainty of ± 52% at the 95% level of confidence was obtained for the values of mean annual sediment yield associated with each catchment (Porto *et al*., 2016a).

Table 4.2: Estimates of mean annual sediment yield for the two periods.

Catchments	**Estimated values of mean annual sediment yield**			
	Period of first sampling		Time window of Re-sampling	
	Years	t ha^{-1} yr^{-1}	Years	t ha^{-1} yr^{-1}
W1	1954-2000	11.6	2001-2014	22.4
W2	1954-1998	16.2	1999-2013	20.6
W3	1954-2000	9.8	2001-2015	29.3

4.2 The results of the re-sampling approach

4.2.1 ^{137}Cs inventories at the reference site and within the study catchments

Fig. 4.3 shows information concerning the ^{137}Cs depth distribution and the inventories at the reference site for both the first sampling campaign in 1999 and the second one in 2014. The shape of the two ^{137}Cs vertical distributions provide valuable confirmation that the sites are undisturbed and thus appropriate for establishing the local ^{137}Cs fallout input. In addition, both profiles, derived from the depth incremental sampling, depict similar shapes and activities.

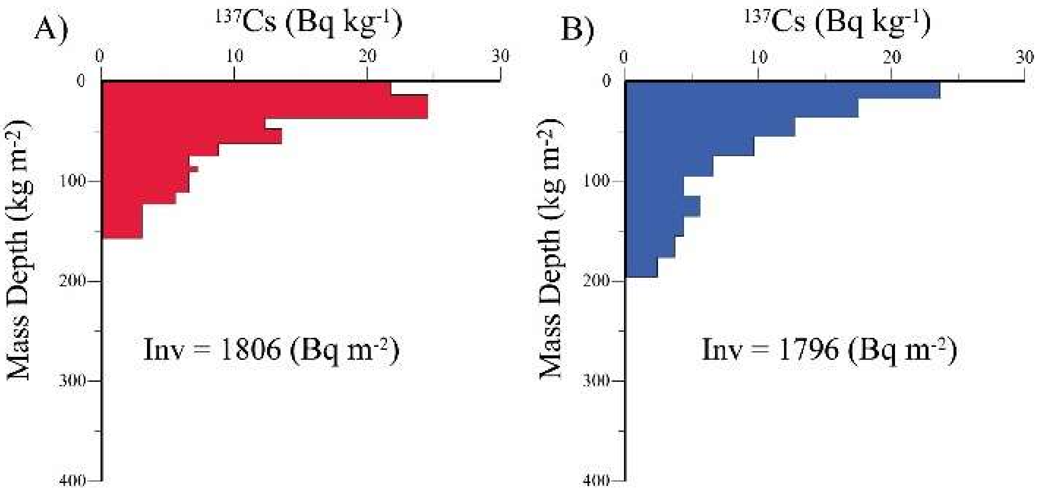


Fig. 4.3: The ^{137}Cs depth distributions and reference inventories documented for the reference site in 1999 (A) and 2014 (B) (Porto *et al.*, 2016a).

For the 1999 profile, ^{137}Cs is found to a depth of about 30 cm in the soil profile and the depth distribution is characterized by an exponential decrease with depth, with the greatest concentration at the surface. The total inventory measured in 1999 was adjusted by taking account of radioactive decay to 2014 in order to confront the values. This reference inventory is estimated to be 1806 Bq m^{-2} (see Table 4.3). The 2014 profile is based on the composite samples for the individual depth increments and the ^{137}Cs depth distribution shows the same exponential decrease with depth, with the greatest activity at the surface too. The total inventory measured in 2014 is estimated to be 1796 Bq m^{-2} which is in close agreement with that of the 1999 profile (after decay correction).

Table 4.3: ^{137}Cs inventories for the reference site and for the study catchments. The values from the 1998-2001 sampling have been decayed to the date of the corresponding resampling campaign (Porto *et al.*, 2016a).

	First sampling 1998-2001				**Re-sampling 2013-2015**			
Catchment	Reference inventory (Bq m^{-2})	Inventory across the catchments			Reference inventory (Bq m^{-2})	Inventory across the catchments		
		Mean (Bq m^{-2})	Range (Bq m^{-2})	SD (Bq m^{-2})		Mean (Bq m^{-2})	Range (Bq m^{-2})	SD (Bq m^{-2})
W1	1806	1035	3.1-3008	565	1796	768	21.2-3127	592
W2		642	13.0-1844	446		435	6.5-969	280
W3		802	71.1-1795	501		462	14.7-1354	290

Table 4.3 provides a summary of the mean, the range and the standard deviation of the ^{137}Cs inventories obtained for the three catchments by the initial sampling campaigns and the re-sampling campaigns. The values have again been decayed to the date of the resampling campaign. All values of ^{137}Cs inventory for the cores collected from catchment W2 and W3 (with the exception of only 1 point) are less than the local reference inventory, indicating that the surfaces of both catchments have been dominated by net soil loss over the period covered by the ^{137}Cs measurements. In the case of catchment W1, however, the inventories for some cores exceeded the reference inventory, pointing to the existence of depositional areas (Porto *et al*., 2016a).

4.2.2 Comparing the ^{137}Cs inventories of the two sampling campaigns at the sampling points

Fig. 4.4 shows the plots of the ^{137}Cs inventories recorded across the three catchments by the 2013-2015 sampling campaigns for a given sampling location versus those recorded by the 1998-2001 sampling campaigns at the same general locations. These provide a useful comparison of the inventory values associated with the two surveys.

There is a sufficient degree of similarity between the first and the second sampling points. The reason of the lack of any clear relationship between the two sets of inventory values concerns the problems associated with re-sampling precisely the same points because it is impossible to find the exact points sampled in 1998-2001 at the present time (GPS has got a margin of error). Furthermore, the need to avoid the area of disturbance associated with collecting the sample during the previous sampling campaign precluded sampling the same points. For this reason, these relationships between the inventories of the two sampling campaigns occur as a result that soil redistribution rates are locally highly variable. Because of this, in order to document changes in soil redistribution rates, it is better to consider the two sets of inventory values given by the two sampling campaigns as two independent representative samples of the ^{137}Cs inventories existing in the study catchment at the time of the sampling campaigns in 1998-2001 and 2013-2015 - instead of comparing the individual pairs of inventory values. This same approach has been defended by Porto *et al.* (2014) when employing a re-sampling approach with ^{137}Cs measurements in the catchment W2.

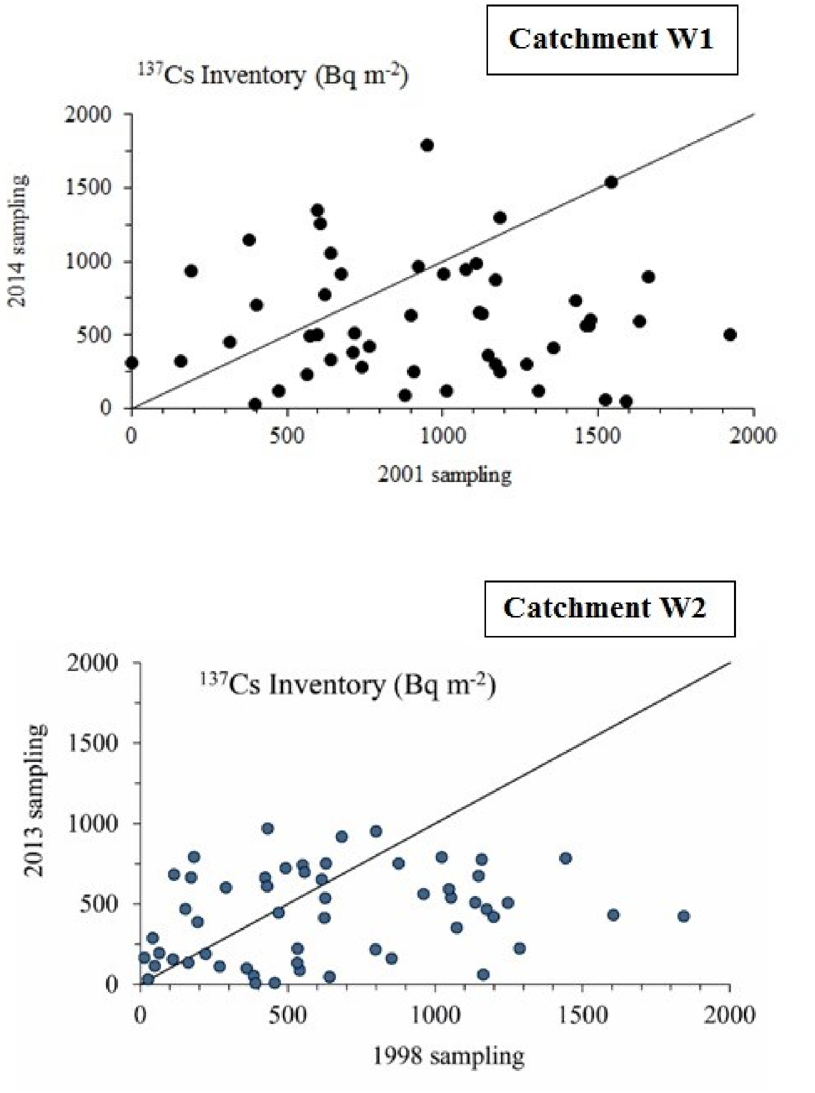


Fig. 4.4: A comparison of the ^{137}Cs inventories measured at the first sampling points in 1998-2001 and 2013-2015 within each three catchments.

4.2.3 Considering particle size and specific surface area selectivity

It is important to take account of the grain size selectivity of erosion processes when using ^{137}Cs measurements to estimate rates of soil redistribution, since contrasts in grain size composition of eroded sediment relative to the parent soil will influence its ^{137}Cs content (e.g. Sutherland, 1991; Walling and Quine, 1990; Walling and He, 1999a, 2001). In this respect, information on the grain size distribution and the specific surface area (SSA) of the sediment eroded and the surface soil for the three experimental catchments was necessary. For each catchment, the SSA is provided in Fig. 4.5 that shows the difference of the SSA frequency distributions derived for the eroded sediment around that of the surface soil. The specific surface area is increased as the particle size becomes small. As a result, it is important to measure the specific surface area to evaluate the coarseness or the fineness of soil and the selectivity of erosion and sedimentation processes.

In Fig. 4.5, the SSA frequency distribution of the surface soil is based on the samples collected from the surface of each catchment, while the SSA frequency distribution of the eroded sediment is based on the measuring of the ordinary sediment sampling. For the three catchments, the values of SSA are consistently lower for the eroded soil than the surface soil. Therefore, the size distribution of the sediment eroded from the catchments is coarser than that of the surface soil. These results indicate a general tendency for the eroded soil to be depleted in fines and the differences between the particle size distributions of the eroded soil and the surface soil likely reflect spatial variability of the grain size composition of the surface soil within the catchments and thus sampling variability associated with the small number of soil samples collected.

It is usually assumed that eroded soil will be enriched in fines, relative to the source material, due to preferential mobilisation of the finer particles (cf. Stone and Walling, 1997) but there are many studies reported in the literature where the eroded soil is coarser than the surface soil (cf. Young, 1980, Porto *et al*., 2003). Young and Onstad (1978) reported similar investigations for loam, silt loam and loamy sand soils in Minnesota, USA. These authors distinguished soil eroded from rill and interrill areas and compared the primary grain size distributions of sediment eroded from rill and interrill areas with those of the bulk surface soil. They found that soil eroded from interrill areas had a higher sand content and a lower clay content than the bulk soil and the sediment eroded from rills. They suggested that these contrasts could reflect the transport processes involved. In the case of interrill areas, the primary transport process is rainsplash, which is capable of transporting larger particles.

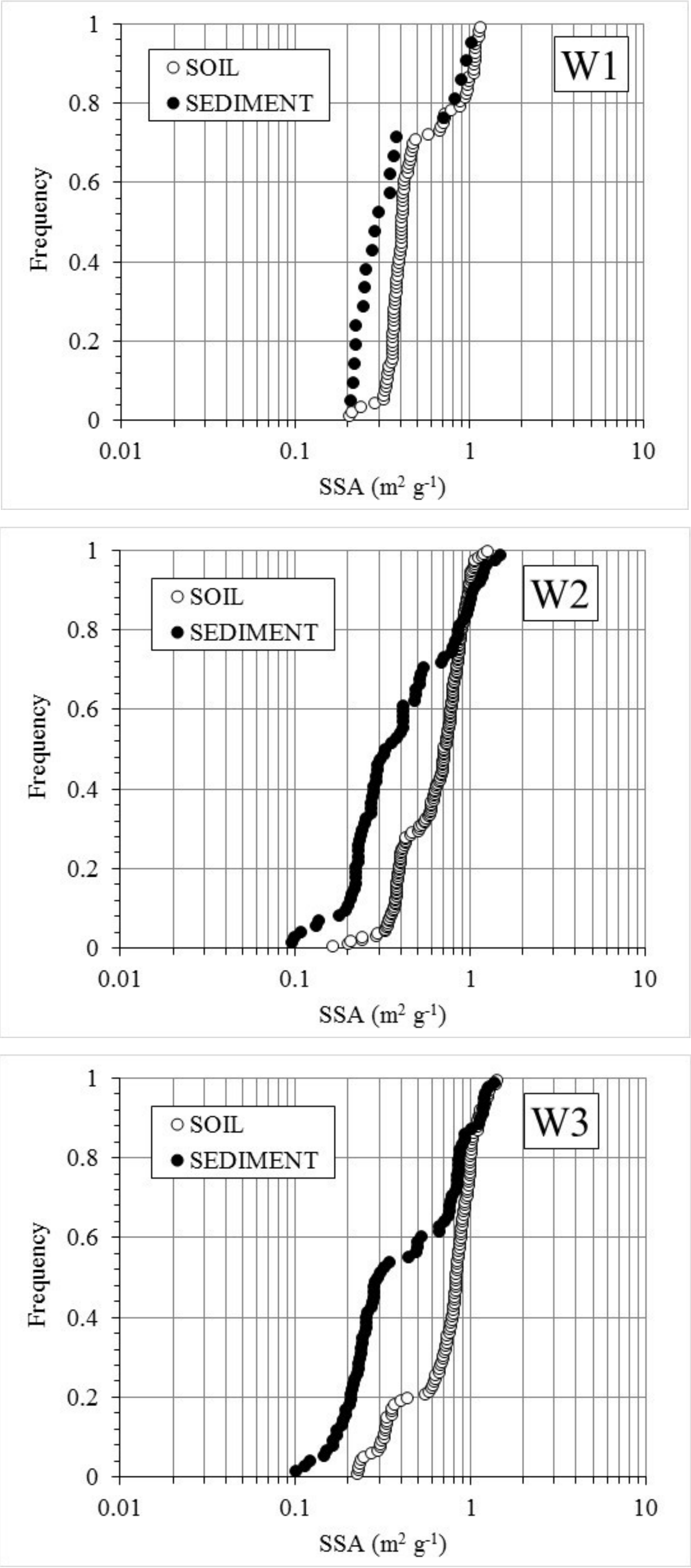


Fig. 4.5: Comparisons between the frequency distributions of specific surface area (SSA) values associated with the samples of eroded soil and the samples of surface soil within the three catchments.

Since much of the soil mobilised by splash erosion will be in the form of aggregates, the primary size distribution of the eroded soil will reflect the size and composition of the eroded aggregates. Available observations indicate that interrill processes dominate sediment mobilisation and transport on the catchments investigated in this study, and the general enrichment of the eroded soil in the coarser size fractions and its depletion in fines is consistent with the findings of Young and Onstad (1978) cited above (Porto *et al.*, 2003).

4.2.4 Estimating mean annual soil redistribution rates

As illustrated in the Section 3 “Materials and Methods” (see § 3.5), in this study the *Diffusion and Migration Model* has been used. Estimates of the soil redistribution rates associated with the individual sampling points for the two sampling campaigns were derived using Eqs. (3.8) and (3.10) to derive estimates of soil erosion and redistribution rates within the catchments from the ^{137}Cs inventories measured at the sampling sites. This model converts the magnitude of the reduction or increase in the ^{137}Cs inventory, relative to the reference inventory, to an estimate of the rate of soil loss or deposition for uncultivated sites. As indicated previously in § 3.6, a particle size effect has been incorporated into the conversion models used to estimate soil redistribution rates from ^{137}Cs measurements by applying the correction factor P, defined as the ratio of the ^{137}Cs concentration in the eroded sediment to that in the original source material. The data obtained from this investigation can be used to test the validity of Eq. (2.22) since the value of P can be calculated for the individual samples of eroded sediment collected from the three catchments. Table 4.4 summarizes the report of values of mean annual soil redistribution rates associated with the two sets of samples with and without correction factor P.

In the case of the first sampling campaigns (1998, 1999, and 2001), the estimates of soil redistribution rate relate to periods from 1954 to 1998-2001, where 1954 is assumed to represent the onset of bomb fallout. For the re-sampling campaigns, the estimates relate to periods from 1954 to 2013-2015. By subtracting the total soil loss for the period 1954 to 2013-2015 from the total soil loss for the period 1954 to 1998-2001, it was possible to estimate the total soil loss, and thus the mean annual soil loss, for the period 1998-2001 to 2013-2015.

For the catchment W1, for the period 1954-2000, the estimated mean annual soil redistribution rates range between 5.0 t ha^{-1} yr^{-1} and -40.7 t ha^{-1} yr^{-1}, with a mean of -8.6 t ha^{-1} yr^{-1} (negative values indicate erosion). For the period 1954-2014, the estimates of mean annual soil redistribution rates range between 10.2 and -25.7 (with a mean of -9.4 t ha^{-1} yr^{-1}). By calculating the total soil loss for the period 1954-2001 as the product of the soil redistribution rate and the period involved in years, and subtracting this from the total soil loss for the period 1954-2014, it is possible to estimate the total soil loss and thus the mean annual soil loss for the period 2001-2014. This latter value was estimated to be 12.3 t ha^{-1} yr^{-1}. Taking in account the values of the correction factor P for each soil samples, the corresponding results are as follows: for the period 1954-2000, the estimated mean annual soil redistribution rate is 10.1 t ha^{-1} yr^{-1}; for the re-sampling period (1954-2014), it is 11 t ha^{-1} yr^{-1} and for the recent time window (2001-2014) it is 14.3 t ha^{-1} yr^{-1}.

For the catchment W2, for the period 1954-1998, the corresponding estimates range between 0 and -34.6 t ha^{-1} yr^{-1} (with a mean of -14.0 t ha^{-1} yr^{-1}), while those for the period 1954-2013 range between -6.4 and -30.0 t ha^{-1} yr^{-1} and provide a mean of 13.6 t ha^{-1} yr^{-1}. The estimate of mean annual net soil loss, calculated as above, for the period 1999-2013 is 12.6 t ha^{-1} yr^{-1}. At the same way, the values of estimated mean annual redistribution rates applying the correction factor P are: 17.2 t ha^{-1} yr^{-1} for the first sampling, 16.7 t ha^{-1} yr^{-1} for the re-sampling and 14.9 t ha^{-1} yr^{-1} for the recent time window.

For the catchment W3, for the period 1954-2000, the equivalent estimates range between -0.1 t ha^{-1} yr^{-1} and -26.5 t ha^{-1} yr^{-1} (with a mean of -11.3 t ha^{-1} yr^{-1}) while those for the period 1954-2015 are respectively -3.2 t ha^{-1} yr^{-1} and -26.8 t ha^{-1} yr^{-1} with a mean of 12.4 t ha^{-1} yr^{-1}. The estimate of mean annual net soil loss, calculated for the period 2001-2015 is 16.4 t ha^{-1} yr^{-1}. If the correction factor P is considered, the corresponding estimates are: 14.4 t ha^{-1} yr^{-1} for the first sampling, 16.2 t ha^{-1} yr^{-1} for the re-sampling campaign and 21.9 t ha^{-1} yr^{-1} for the recent time window (see Porto *et al.*, 2016a).

Table 4.4: Results of estimated mean annual soil redistribution rates for the three catchments.

Estimated mean annual soil redistribution rates						
	Correction factor P = 1					
Catchments	First sampling		Re-sampling		Recent time window	
	Years	t ha^{-1} yr^{-1}	Years	t ha^{-1} yr^{-1}	Years	t ha^{-1} yr^{-1}
W1	1954-2000	8.6	1954-2014	9.4	2001-2014	12.3
W2	1954-1998	14	1954-2013	13.6	1999-2013	12.6
W3	1954-2000	11.3	1954-2015	12.5	2001-2015	16.4
	With correction factor P					
	Years	t ha^{-1} yr^{-1}	Years	t ha^{-1} yr^{-1}	Years	t ha^{-1} yr^{-1}
W1	1954-2000	10.1	1954-2014	11.0	2001-2014	14.3
W2	1954-1998	17.2	1954-2013	16.7	1999-2013	14.9
W3	1954-2000	14.4	1954-2015	16.2	2001-2015	21.9

As indicated on Section 3 "Materials and Methods" (see § 3.4.3), a value of ±10% at the 95% level of confidence, related to the precision of the gamma spectrometer measurements, has been attributed to the individual estimates of mean annual soil loss obtained for each sampling point. However, when summing these values to calculate the mean, these errors made void and they have not been propagated through to the mean values. It is necessary to consider the effects of sampling variability because if the soil core has been collected from a slightly different point it is likely that a different estimate of mean annual soil loss have been obtained. But this source of uncertainty (ca. ± 30% at the 95% level of confidence, reported by Sutherland, 1996) has been ignored when assessing the uncertainty of the three values of mean annual soil loss associated with the different periods. The reason is that the sampling points overestimation at some points is balanced by underestimation at others. However, uncertainty associated with the sampling design has been incorporated into this analysis and the estimates of mean annual soil loss of the two sets of samples have a standard error. Based on their standard deviations associated with the estimates of mean annual soil loss for the first period and the second one, the estimate of mean annual net soil loss for each catchment are assessed with a precision of ± 13% at the 95% level of confidence.

Further uncertainty can be also attributable to the conversion model. The estimates produced could be imprecise but this source of uncertainty can be essentially ignored when comparing the values of mean annual soil loss obtained from the ^{137}Cs measurements for the different catchments and for the two different periods. Instead, it becomes important when comparing the absolute values of the estimates of soil loss provided by the ^{137}Cs measurements for the two periods with the equivalent estimates of sediment yield at the catchment outlets for the same periods. In the absence of detailed guidance for quantifying this source of uncertainty, a nominal ±25% has been used (see Porto *et al.*, 2014). Use of a summation in quadrature procedure to combine these estimates of uncertainty provides an overall uncertainty of ±28% for the estimates of mean annual soil loss for the three periods (Porto *et al.*, 2016a).

4.3 Discussion

The results presented above show both the values of sediment yield and the estimates of soil erosion generally suggest evidence of change in sediment mobilisation within and from the catchment during the different periods.

Figure 4.6 shows the mean annual net soil loss from the three study catchments for the two different periods (from 1954 to 1998-2000 and from 1999-2001 to 2013-2015), estimated using the ^{137}Cs measurements and both without and with the correction factor *P*. As made clear above, these estimates have been expressed as ranges to take account of the estimated ± 13% uncertainty associated with sample size.

Correction factor *P* = 1

With correction factor *P*

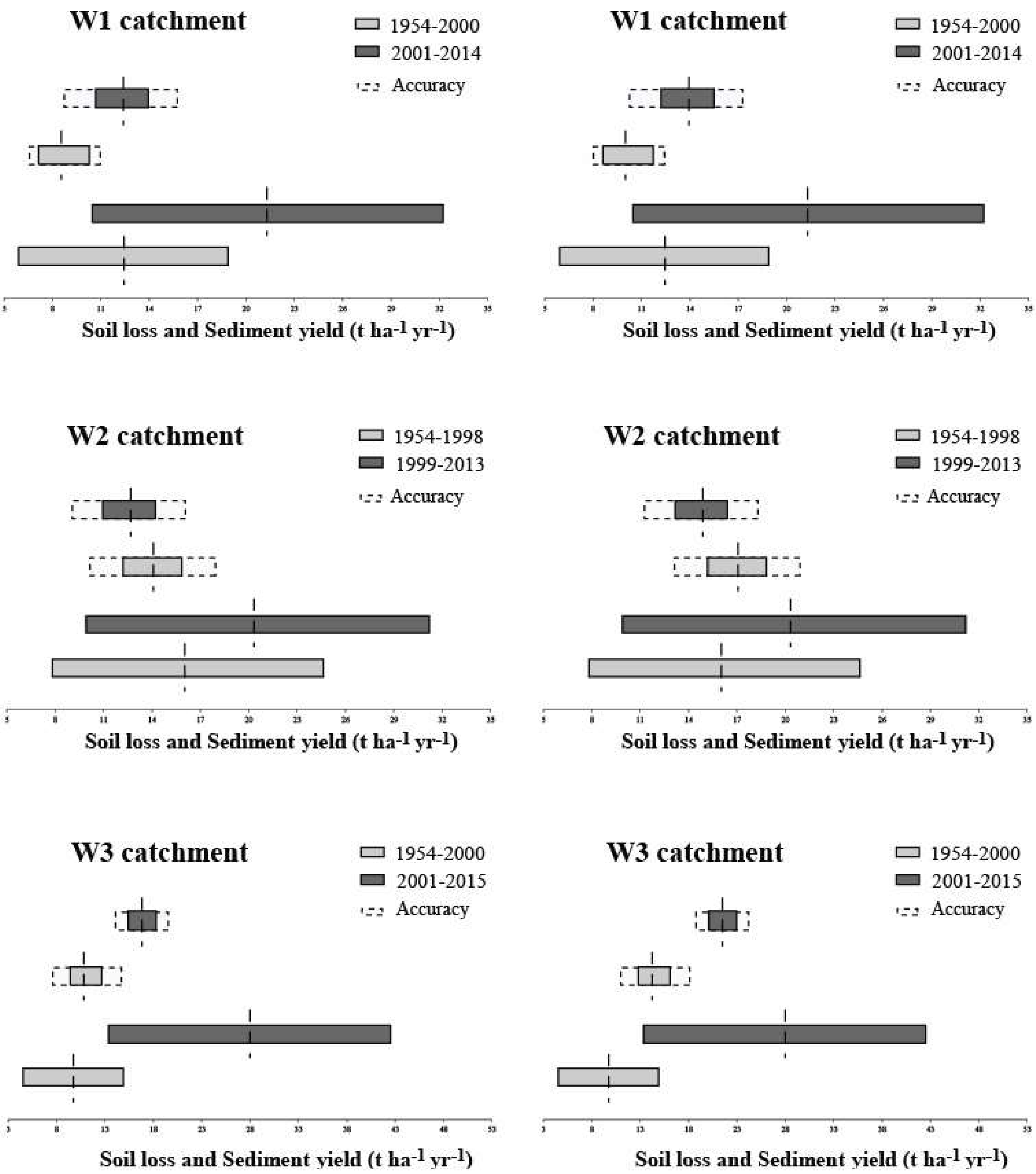


Fig. 4.6: Comparison of the ranges covered by the estimates of mean annual net soil loss and sediment yield for each study catchment and for the periods related to the sampling campaigns, when taking account of their uncertainty.

For the catchments W1 and W3, the results of mean annual net soil loss show a significant increase for the more recent period. Instead, for catchment W2 the results do not evidence a significant increase and the annual net soil loss has conceivably declined in the more recent period.

Reminding that one of the most important goal of this study was to compare the results with the independent estimates of the changes in mean annual sediment yield related to the same periods available for the three catchments, the incomplete records mean that the estimates of mean annual sediment yield for the two periods entail considerable uncertainty. This has been estimated to be ca. ± 52% at the 95% level of confidence (see § 4.1).

A simple comparison of the mean annual sediment yields of the three catchments for the two periods suggests that sediment yields have increased in recent years, but when the uncertainty is taken into account the evidence is less clear. There is strong evidence for an increase from catchment W3 and some evidence for the same trend from catchment W1. However, in the case of catchment W2 there is no evidence of an increase. This is consistent with the results provided by the ^{137}Cs measurements, where catchments W1 and W3 provided clear evidence of increased net soil loss in recent years, but there was no evidence of an increase for catchment W2. When comparing the values of mean annual net soil loss and mean annual sediment yield presented in Fig. 4.6 in absolute terms, the estimates of the latter are consistently greater than the equivalent values of mean annual net soil loss in five out of six cases. The estimates of net soil loss only exceed the estimate of sediment yield for the period 1954-2000 in catchment W3. Close agreement between the estimates of net soil loss and sediment yield would not necessarily be expected. The latter could be expected to be higher if channel erosion represented an important sediment source, or possibly lower if substantial longer-term sediment storage occurred in the catchment channel systems. However, field observations indicate that neither are important processes in the study catchments. When the additional uncertainty associated with the absolute values of the net soil loss estimates noted above is considered and the overall uncertainty is increased to ±28% at the 95% level of confidence, no clear evidence of a significant difference in magnitude between the two measures of soil loss is evident. This provides further general validation of the use of ^{137}Cs measurements to estimate soil redistribution rates. Based on these findings, it can be suggested that there has been an increase in soil erosion rates in catchments W1 and W3 over the past ca. 15 years. On the contrary, the corresponding estimates for catchment W2 show no significant difference in mean annual net soil loss between the two periods. The estimates of sediment yield from catchments W1 and W3 also provide evidence of increasing sediment yield for the second period, which is consistent with the evidence of increasing net soil loss. In the case of catchment W2, the lack of a significant change in sediment yield is consistent with the evidence provided by the soil loss data for the two periods (Porto *et al.*, 2016a).

The increased net soil loss and sediment yield during the second period evidenced by catchments W1 and W3 is in turn consistent with independent evidence provided by the available records of sediment yield from the catchments and the trend of increasing annual erosivity factor R (Wischmeier and Smith, 1978) demonstrated by the records from two rainfall measuring stations in the vicinity of the study catchment, as showed in Fig. 4.7.

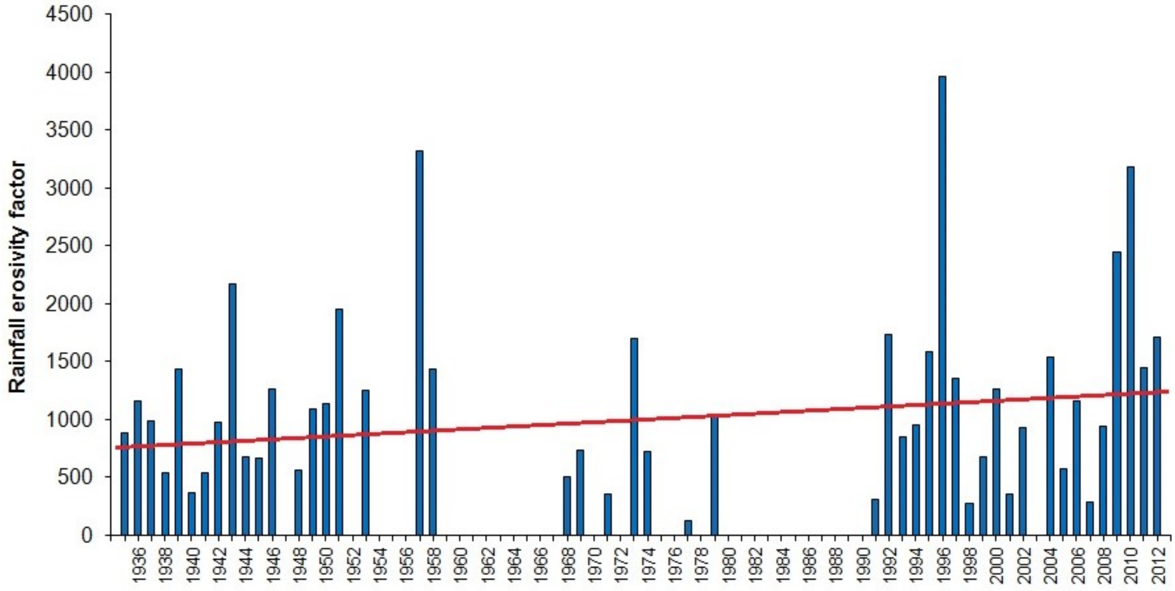


Fig. 4.7: Annual values and trends of the rainfall erosivity factor calculated for two stations in the local area.

Figure 4.7 provides evidence of a statistically significant ($P \leq 0.10$) trend (increase) in rainfall erosivity during recent years, which is again consistent with the increased annual net soil loss suggested by the results of the resampling study and the associated ^{137}Cs measurements. It should, however, be noted that the trend of increasing rainfall erosivity shown in Fig. 4.7 for two rainfall measuring stations in the vicinity of the study catchment, is not reflected by all rainfall measuring stations in the local region (i.e. Calabria). As reported by Capra *et al.* (2015), some stations show a trend of declining erosivity during the period 1992-2012.

Conclusion and Future Prospectives

5.1 Conclusion

The ^{137}Cs technique is traditionally used with a single field visit to estimate retrospectively (from 1950s to the present) the net soil redistribution by the processes of soil erosion and deposition. In this study, the Re-sampling approach, related to the ^{137}Cs measurements, is proposed and compared with the traditional approach. The Re-sampling approach is capable of providing information for different time windows and to obtain information on recent changes in soil redistribution rates. This alternative approach has the potential to allow for more precise results for the much shorter time period than the traditional retrospective application of the ^{137}Cs technique. In fact, the results of this PhD dissertation have demonstrated the potential for using ^{137}Cs measurements and the re-sampling technique to assemble information on soil erosion and redistribution rates within three small uncultivated and forested catchments in southern Italy during a recent period. This information, coupled with the sediment yield measurements given by the monitoring programme at the catchment outlets, provides valuable information on soil redistribution within the catchments in response to Global Change and more particularly changes in land use and precipitation erosivity.

This study represents what is probably the first attempt to apply a re-sampling approach within an uncultivated and two forested catchments. The available data by a suspended sediment monitoring programme provided information on annual suspended sediment yields for the periods 1978-1994 and 2006-2015 in order to confirm both the estimates of net soil loss from the catchment using ^{137}Cs measurements and any contrasts in mean annual erosion rates between the periods first sampling (from 1954 to 1998-2001) and the recent time window 1998-2015 indicated by those estimates.

The catchments were first sampled for ^{137}Cs measurements during the period 1998-2001 and a total of 68 cores was collected from the catchment W1, 55 from the W2 and 81 from the W3, to provide a representative spatial coverage. The collection of these cores from each catchment was seen to provide a meaningful basis for estimating the spatially averaged erosion rates. These same points were re-sampled in 2013-2015 and the ^{137}Cs measurements undertaken on the cores collected from the catchments provide consistent estimates of the erosion and deposition rates.

A *Diffusion and Migration Model* was used as the conversion model, in preference to the more frequently used profile distribution model because the latter is likely to overestimate soil redistribution rates where ambient erosion rates are low. In order to provide more accurate estimates of soil redistribution rates, the choice of conversion model is clearly an important consideration. The more complex diffusion and migration model generally provides more consistent results, although in its basic form it is seen to underestimate erosion rates, particularly where ambient erosion rates are high. The refined version of the model, which takes account of the depth distribution of ^{137}Cs in the eroding layer, provides better results for soils with high erosion rates while also performing satisfactorily for those with lower erosion rates. In conclusion, it can be suggested that the refined migration and diffusion model is likely to provide the most reliable results, although the simple exponential profile distribution model should provide valid results for areas where erosion rates are relatively high (i.e. ca. 20 t ha^{-1} yr^{-1}) (Porto *et al.*, 2003).

Further, as confirming the potential for using a re-sampling approach with ^{137}Cs measurements, the results obtained from the ^{137}Cs measurements suggest an increase in mean annual net soil loss from the catchments W1 and W3 and no significant change in catchment W2. This comparison suggests that mean annual rates of net soil loss had increased during the period between the two sampling campaigns and that this increase was associated with a shift to an increased sediment delivery ratio. This change was consistent with independent information on likely changes in the sediment response of the study catchment provided by the available records of annual sediment yield and changes in the annual rainfall documented for the local area (Porto *et al.*, 2016a).

For the catchment W2, the estimates of mean annual soil loss for the two periods show no evidence of a significant increase or decrease and this lack of change is consistent with the estimates of sediment yield from the study catchment for the two periods. In this context, it is important to recognise that the uncertainty associated with the results of a re-sampling study is likely to mean that the approach will be unable to document small changes in erosion rates (Porto *et al.*, 2014).

This is consistent with the trends demonstrated by independent information on changes in mean annual sediment yield from the catchments. The increased rates of annual net soil loss from two of the three catchments are in turn consistent with recent studies undertaken in Southern Italy which have identified an increasing trend in rainfall erosivity during the last 10-15 years (Porto and Walling, 2012; Porto *et al.*, 2013) comparing with the previous about 45 years. However, the use of these two discrete periods for the comparison could mask some subtle changes, due to the substantial degree of inter-annual variability of soil loss and sediment yield within the study area.

5.2 Future prospectives

Further validation of the re-sampling approach, particularly in different environments and in areas where soil redistribution rates might be expected to be declining could usefully be undertaken. The availability of more complete records of annual sediment yield from the study catchments would permit greater emphasis on validating the extent of the change in the annual soil redistribution rates, as well as its direction (Porto *et al.*, 2016).

More work is required to explore the potential of the re-sampling approach in areas with uncultivated soils and to validate the results obtained with independent data. The study reported in this PhD dissertation addresses this requirement by describing the results obtained from a study of uncultivated catchments (two of them are forested) in southern Italy, where the availability of sediment yield data gave an opportunity to confirm the results obtained. The potential impact of climate change on erosion rates and sediment yields is increasingly seen as a key issue in Mediterranean areas (Garcia-Ruiz *et al.*, 2013) and this study can therefore be seen as possessing both regional and more generic methodological relevance (Porto *et al.*, 2014).

These results highlight an important suggestion of the potential for using ^{137}Cs measurements in association with the re-sampling approach in order to assess recent changes in soil redistribution rates within the study catchments.

References

Anderson JM, Ingram JSI (1993) *Tropical Soil Biology and Fertility. A Handbook of Methods, CAB International*. Wallingford.

Arata L, Alewell C, Frenkel E, A'Campo-Neuen A, Iurian AR, Ketterer ME, Mabit L, Meusburger K (2016) Modelling Deposition and Erosion rates with RadioNuclides (MODERN) - Part 2: A comparison of different models to convert $^{239+240}$Pu inventories into soil redistribution rates at unploughed sites. *Journal of Environmental Radioactivity* **162-163**: 97-106.

Avolio S, Ciancio O, Grinovero C, Iovino F, Mirabella A, Raglione M, Sfalanga M, Torri D (1980) Effetti del tipo di bosco sull'entità dell'erosione in unità idrologiche della Calabria - Modelli erosivi. *Annali Istituto Sperimentale Selvicoltura* **11**: 45–131.

Bagarello V, Ferro V (2006) *Erosione e conservazione del suolo*. MCGraw-Hill.

Bernard C, Mabit L, Laverdiere MR, Wicherek S (1998) Cesium-137 et erosion des sols. *Cahiers Agricultures* **7** (**3**): 179-186.

Blake WH, Walling DE, He Q (1999) Fallout beryllium-7 as a tracer in soil erosion investigations. *Appl. Radiat. Isot.* **51**: 599-605.

Boardman J, Poesen J, Evans R (2003) Socio-economic factors in soil erosion and conservation. *Environ. Sci. Policy* **6**: 1-6.

Brakensiek DL, Osborn HB, Sheridan JM. (1979) Field Manual for Research in Agricultural Hydrology. *Agriculture Handbook No. 224*. US Department of Agriculture Science and Education Administration: Washington, DC.

Brown LR, Wolf EC (1984) Soil erosion: quiet crisis in the world economy. *Worldwatch paper 60* Washington DC, Worldwatch Institute.

Bunzl K, Schultz W (1985) Distribution coefficient of ^{137}Cs and ^{90}Sr by mixture of clay and humic materials. *J. Radioanal. Nucl. Chem.* **90**: 23-37.

Campbell BL, Loughran RJ, Elliott GL (1982) Caesium-137 as indicator of geomorphic processes in a drainage basin system. *J. Aust. Geogr. Stud.* **20**: 49-64.

Campbell BL, Loughran RJ, Elliott GL (1988) A method for determining sediment budget using caesium-137. In: *Sediment Budget* (Proc. of the Porto Alegre Sympos.) *IAHS Publ.* **174**: 171-179.

Capra A, Porto P, La Spada C (2015) Long-term variation of rainfall erosivity in Calabria (Southern Italy*). Theor. Appl. Climatol.* http://dx.doi.org/10.1007/s00704-015-1697-2.

Carter MW, Moghissi AA (1977) Three decades of nuclear testing. *Health Physics* **33**: 55‑71.

Cawse PA, Horrill AD (1986). A Survey of Caesium-137 and Plutonium in British Soils in 1977. *AERE Harwell Report R-10155*, HMSO.

Cinnirella S, Iovino F, Porto P, Ferro V (1998). Antierosive effectiveness of Eucalyptus coppices through the cover management factor estimate. *Hydrological Processes* **12**: 635-649.

Clark EH, Haverkamp JA, Chapman W (1985) Eroding soils: the off-farm impact. The Conservation Foundation, Washington DC.

Coleman NT, Lewis RJ, Craig D (1963) Sorption of caesium by soils and its displacement by salt solution. *Soil Sci. Soc. Am. Proc.* **27**: 290-294.

Crank J (1975) The Mathematics of Diffusion, 2nd ed, Clarendon: Oxford, pp. 414.

Davis JJ (1963) Cesium and its relationship to potassium in ecology. In Schultz V, Klement AW Jr, (Eds) *Radioecology*, New York: Reinhold, pp 539-556.

Dahlman RC, Francis CW, Tamura T (1975) Radiocesium cycling in vegetation and soil. In Howell FG, Gentry JB, Smith MH (Eds) *Mineral Cycling in Southeastern Ecosystems*, *USAEC Symposium Series*, CONF-740513, US Atomic Energy Commission, Washington DC pp 462-481.

De Jong E, Villar H, Bettany JR (1982) Preliminary investigations on the use of ^{137}Cs to estimate erosion in Saskatchewan. Can. *J. Soil Sci*. 673-683.

De Jong E, Begg CM, Kachanoski RG (1983) Estimates of soil erosion and deposition from Saskatchewan soils. *Can. J. Soil Sci.* **63**: 607-617.

De Jong E, Kachanoski RG (1988) The importance of erosion in the carbon balance of prairie soils. *Canadian J. Soil Science* **68**: 111–119.

De Jong E, Wang C, Rees HW (1986) Soil redistribution on three cultivated new Brunswick hillslopes in a Welsh upland lake-catchment based on magnetic susceptibility measurements. *Quat. Res.* **16**: 356-372.

Diaz LJ, Jaffe DA, Kaspar J, Knecht A, Miller ML, Robertson RGH, Schubert, AG (2011) Arrival time and magnitude of air borne fission products from the Fukushima, Japan, reactor incident as measured in Seattle, WA, USA. *J. Environ. Radioact.* **102**: 1032-1038.

Di Stefano C, Ferro V, Porto P, Tusa G (2000) Slope curvature on soil erosion and deposition processes. *Water Resources Research*, **75**(4): 349-364.

Du, P & Walling DE (2011) Using ^{137}Cs measurements to investigate the influence of erosion and soil redistribution on soil properties. *Applied Radiation and Isotopes* **69**: 717-726.

Elliott GL, Campbell BL, Loughran RJ (1990) Correlation of erosion measurement and soil caesium-137 content. *Appl. Radiat. Isot.* **41**: 713‑717.

Elliot WJ, Foster GR, Elliot AV (1991) Soil erosion: Processes, impacts and prediction. In Lal R, Pierce FJ (Eds) *Soil Management for Sustainability. Soil Water Conserv. Soc.*, Edmonton, Alberta, Canada, pp 25-34.

Elwell HA (1984) Sheet erosion from arable land in Zimbabwe: prediction and control. In Walling DE (Ed) *Challenges in African Hydrology and Water Resources. Proceedings of the Harare Symposium*, *July 1984*. IAHS Publication No. 144, Wallingford, UK 429-438.

Evans EJ, Dekker AI (1966) Plant uptake of Cs-137 from nine Canadian soils. *Canadian Journal of Soil Science* **46**: 167-176.

Evrard O, Laceby JP, Lepage H, Onda Y, Cerdan O, Ayrault S (2015) Radiocesium transfer from hillslopes to the Pacific Ocean after the Fukushima Nuclear Power Plant accident: A review. *Journal of Environmental Radioactivity* **148**: 92-110.

Eyman LD, Kevern NR (1975) Cesium-137 and stable cesium in a hypereutrophic lake. *Health Phys*ics **28**: 549-555.

Ferro V (1997) Further remarks on a distributed approach to sediment delivery. *Hydrological Sciences Journal* **42** (5): 633-648.

Ferro V, Porto P, Yu B (1999) A comparative study of rainfall erosivity for South Italy and Southeastern Australia. *Hydrological Sciences Journal* **44**: 3-24.

Food and Agriculture Organization of the United Nations (2005) National Soil Degradation Maps. Soil Degradation Map of Austria. FAO/AGL. GLASOD.

Foster GR, Meyer LD, Onstad CA (1977) An erosion equation derived from basic erosion principles. *Transactions of the ASAE*, **20**(4): 678-682.

Foth HD (1990) *Fundamentals of Soil Science 8th Edition*. Canada: John Wiley & Sons. pp. 22-25 ISBN 0-471-52279-1.

Fredericks DJ, Perrens SJ (1988) Estimating erosion using caesium-137: II. Estimating rates of soil loss. *IAHS Publ*. **174**: 225-231.

Frissel MJ, Pennders R (1983) Models for the accumulation and migration of ^{90}Sr, ^{137}Cs, ^{239,240}Pu and ^{241}Am in the upper layer of soils. In: Coughtrey PJ (Ed.) *Ecological Aspects of Radionuclide Release* (pp 63-72). Special Publication of the British Ecological Society No. 3, Oxford.

Garcia-Oliva F, Martinez Lugo R, Maass JM (1995) Long-term net soil erosion as determined by ^{137}Cs redistribution in an undisturbed and perturbed tropical deciduous forest ecosystem. *Geoderma* **68**: 135-147.

Garcia-Ruiz JM, Nadal-Romero E, Lana-Renault N, Bequeria S (2013) Erosion in Mediterranean landscapes: changes and future challenges. *Geomorphology* **198**: 20-36.

Golosov V, Belyaev V, Kuznetsova Yu, Markelov M, Shamshurina E (2008) Response of a small arable catchment sediment budget to introduction of soil conservation measures. In: *Proceedings of the Sediment Dynamics in Changing Environments*, Wallingford, UK. IAHS Publ. **325**: 106–113.

Goosens D (2002) The on-site and off-site effects of wind erosion. In: Warren A (Ed.): *Wind Erosion in Agricultural Land. Research Results for Land Managers. EC-DG Research, Environment and Sustainable Development Programme*. EU official publication, Luxembourg 29-38.

Greppi M (2004) *Idrologia. Il ciclo dell'acqua e i suoi effetti*. Hoepli **11**: 151-165.

He Q (1993) Interpretation of Fallout Radionuclide Profiles in Sediments from Lake and Floodplain Environments. University of Exeter.

He Q, Walling DE (1996). Interpreting the particle size effect in the adsorption of ^{137}Cs and unsupported ^{210}Pb by mineral soils and sediments. *Journal of Environmental Radioactivity* **30**: 117-137.

He Q, Walling DE (1997) The distribution of fallout ^{137}Cs and ^{210}Pb in undisturbed and cultivated soils. *Applied Radiation and Isotopes* **48**: 677‑690.

IAEA (2006) Assess the effectiveness of soil conservation techniques for sustainable watershed management using fallout radionuclides. Report of the *Third Research Coordination Meeting of the FAO/IAEA Coordinated Research Project Held* (IAEA-311-D1-RC-888) Vienna, pp. 27-30.

IAEA (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. Report of the *Impact of Soil Conservation Measures on Erosion Control and Soil Quality*. Vienna, pp. 11-33.

IAEA (International Atomic Energy Agency), 2014. Guidelines for Using Fallout Radionuclides to Assess Erosion and Effectiveness of Soil Conservation Strategies. IAEA-TECDOC-1741. IAEA Publication, Vienna, Austria, p. 213

Iovino F, Puglisi S. (1991) L'aménagement des reboisements de protection. Un cas d'étude. *Proceedings of 10th World Forestry Congress*, Paris.

Kachanoski RG (1993) Estimating soil loss from changes in soil caesium-137. *Canadian Journal of Soil Science*. **73**: 515-526.

Kachanoski RG, de Jong E (1984) Predicting the temporal relationship between soil cesium-137 and erosion rate. *J. Environmental Quality* **13**: 301-304.

Karlen DL, Mausbach MJ, Doran JW, Cline RG, Harris RF, Schuman GE (1997) Soil quality: a concept, definition, and framework for evaluation. *Soil Sci. Soc. Am. J.* **61**: 4-10.

Kirkby MJ (1969) Infiltration, throughflow and overland flow. In Chorley RJ (Ed) *Water, earth and man*. London: Methuen, pp. 215-27.

Lal R (1994) Soil Erosion. In Lal R (Ed.) Ankeny, IA: *Soil and Water Conservation Society*.

Lal R (2000) Soil management in the developing countries. *Soil Science* **165**:57-72.

Li S, Lobb DA, Lindstrom MJ, Farenhorst A (2008) Patterns of tillage and water erosion on topographically complex landscapes in the North America Great Plains. *J. Soil Water Conservation* **63**: 37-46.

Li S, Lobb DA, Kachanoski RG, McConkey BG (2011) Comparing the use of the traditional and repeated-sampling-approach of the ^{137}Cs technique in soil erosion estimation. *Geoderma* **160**: 324-335.

Lin L, Bao X,Qi W, Lou Y, Du Z, Wen H (1987) Environmental impact of Chernobyl accident on Beijing area. *Chin. J. Radiol. Med. Prot.* **7**(Supplement): 19-22, in Chinese.

Lindstrom FT, Boersma L (1971) A theory on the mass transport of previously distributed chemicals in a water-saturated sorbing porous medium. *Soil Science* **111**: 192-199.

Livens FR, Baxter MS (1988) Chemical associations of artificial radionuclides in Cumbrian soils. *Environmental Radioactivity* **7**: 75-86.

Livens FR, Loveland PJ (1988) The influence of soil properties on the environmental mobility of caesium in Cumbria. *Soil Use and Management* **4**: 69-75.

Livens FR, Rimmer DL (1988) Physico-chemical controls on artificial radionuclides in soil. *Soil Use and Management* **4**: 63-69.

Lobb DA, Kachanoski RG (1999) Modelling tillage erosion in the topographically complex landscapes of southwestern Ontario, Canada. *Soil Till. Res*. **51**: 261-277.

Lomenick TF, Tamura T (1965) Naturally occurring fixation of cesium-137 on sediments of lacustrine origin. *Proc, Soil Sci. Soc. Am*. **29**: 383-387.

Loughran RJ (1989) The measurement of soil erosion. *Progress in Physical Geography* **13**: 216-233.

Loughran RJ, Balog RM (2006) Re-sampling for soil-caesium-137 to assess soil losses after a 19 year interval in a Hunter Valley vineyard, New South Wales, Australia. *Aust. Geogr. Stud.* **44**: 77-86.

Loughran RJ, Campbel BL, Walling DE (1987) Soil erosion and sedimentation indicated by caesium-137: Jackmoor Brook catchment, Devon, England. *Catena* **14**: 201-212.

Loughran RJ, Elliott GL, Campbell BL, Shelly DJ (1988) Estimates of soil erosion from caesium-137 measurements in a small cultivated catchment in Australia. *Appl. Radiat. Isot*. **39**: 1153-1157.

Mabit L, Benmansour M, Walling DE (2008) Comparative advantages and limitations of fallout radionuclides (^{137}Cs, ^{210}Pb and ^{7}Be) to assess soil erosion and sedimentation. *J. Environ. Radioact*. **99**: 1799-1807.

Mabit L, Bernard C (1998) Relationship between soil inventories and chemical properties in a small intensively cropped watershed. *Comptes Rendus de l'Académie des Sciences Série IIa Earth Planet Sci*. **327**: 527-532.

Mabit L, Bernard C, Laverdiere MR (2000) L'étude de l'érosion hydrique au Québec. *Vecteur environnement* **33**:34-43.

Mabit L, Bernard C, Laverdiere MR (2002) Influence of the sampling strategy on soil loss assessments from ^{137}Cs measurements. *Man and Soil at the Third Millenium* (Rubio JL, Morgan RPC, Asins S, Andreu V, Eds). Geoforma Ediciones, Logrono, Spain 2083-2090.

Mabit L, Bernard C, Laverdiere MR (2007) Assessment of erosion in the Boyer River watershed (Canada) using a GIS oriented sampling strategy and ^{137}Cs measurements. *Catena* **71**: 242-249.

Mabit L, Bernard C, Makhlouf M, Laverdière MR (2008) Spatial variability of erosion and soil organic matter content estimated from ^{137}Cs measurements and geostatistics. *Geoderma* **145**: 245-251.

Mabit L, Chhem-Kieth S, Dornhofer P, Toloza A, Benmansour M, Bernard C, Fulajtar C, Walling DE (2014) ^{137}Cs: A widely used and validated medium-term soil tracer. In: *Guidelines for using fallout radionuclides to assess erosion and effectiveness of soil conservation strategies*, IAEA. Vienna, pp. 27-78.

Mabit L, Laverdiere MR, Bernard C (2002) L'érosion hydrique: méthodes et études de cas dans le nord de la France. *Cah. Agri*. **11**: 195-206.

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. *Earth-Sci. Rev*. **127**: 300-307.

Mabit L, Zapata F, Benmansour M, Bernard C, Dercon G, Walling DE (2014) Assessment of soil erosion and sedimentation: the role of fallout radionuclides. In: *Guidelines for using fallout* *radionuclides to assess erosion and effectiveness of soil conservation strategies*, IAEA. Vienna, pp. 3-26.

Masson O, Baeza A, Bieringer J, Brudecki K, Bucci S, Cappai M, Carvalho FP, Connan O, Cosma C, Dalheimerc A, Didier D, Depuydt G, De Geer LE, De Vismes A, Gini L, Groppi F, Gudnason K, Gurriaran R, Hainz D, Halldórsson O, Hammond D, Hanley O, Holeý K, Homoki ZS, Ioannidou A, Isajenko K, Jankovic M, Katzlberger C, Kettunen M, Kierepko MR, Kontro R, Kwakman PJM, Lecomte M, Leon Vintro L, Leppänen AP, Lind B, Lujaniene G, Mc Ginnity P, Mc Mahon C, Malá H, Manenti S, Manolopoulou M, Mattila A, Mauring A, Mietelski JW, Møller B, Nielsen SP, Nikolic J, Overwater RMW, Pálsson S.E, Papastefanou C, Penev I, PhamnMK, Povinec PP, Ramebäck H, Reis MC, Ringer W, Rodriguez A, Rulík P, Saey PRJ, Samsonov V, Schlosser C, Sgorbati G, Silobritiene BV, Söderström C, Sogni R, Solier L, Sonck M, Steinhauser G, Steinkopff T, Steinmann P, Stoulos S, Sýkora I, Todorovic D, Tooloutalaie N, Tositti L, Tschiersch J, Ugron A, Vagena E, Vargas A, Wershofen H, Zhukova O (2011) Tracking of airborne radionuclides from the damaged Fukushima Dai-ichi nuclear reactors by European networks. *Environ. Sci. Technol.* **45**: 7670-7677.

Martz LW, De Jong E (1987) Using caesium-137 to assess the variability of net soil erosion and its association with topography in a Canadian prairie landscape. *Catena* **14**: 439-451.

Matisoff G, Whiting PJ (2011) Measuring soil erosion rates using natural (^{7}Be, ^{210}Pb) and anthropogenic (^{137}Cs, ^{239,240}Pu) radionuclides. In: Baskaran M (Ed.) *Handbook of Environmental Isotope Geochemistry*. Vol. 1, pp. 487-519.

McHenry JR, Ritchie JC (1977) Physical and chemical parameters affecting transport of Cs-137 in arid watersheds. *Water Resour. Res.* **13**: 923-927.

Morgan RPC (1980) Soil Erosion: Topics in Applied Geography 99, 2nd Edition. Longman, New York, USA 113.

Morgan RPC (2005) Soil Erosion and Conservation, 3rd Edition. Longman Group, Harlow Essex, New York, USA 198.

Navas A, Machín J, Soto J (2005) Assessing soil erosion in a Pyrenean mountain catchment using GIS and fallout ^{137}Cs. *Agric. Ecosyst. Environ.* **105**: 493-506.

Officer CB, Lynch DR (1982) Interpretation procedures for the determination of sediment parameters from time-dependent flux imputs. *Earth and Planetary Science Letters* **61**: 55-62.

Owens PN, Walling DE (1996) Spatial variability of caesium-137 inventories at reference sites. An example from two contrasting sites in England and Zimbabwe. *Appl. Radiat. Isot.* **47**: 699-707.

Owens PN, Walling DE, He Q (1996) The behaviour of bomb-derived Caesium-137 fallout in catchment soils. *J. Environ. Radioact.* **32**: 169-191.

Pansak W, Hilger TH, Dercon G, Kongkaew T, Cadisch G (2008) Changes in the relationship between soil erosion and N loss pathways after establishing soil conservation systems in uplands of Northeast Thailand. *Agriculture, Ecosystems & Environment* **128**: 167-176.

Pegoyev AN, Fridman SHD (1978) Vertical profiles of cesium-137. Pochvovedeniye **8:** 77-81.

Pennock DJ, Appleby PG (2002) Site selection and sampling design. *Handbook for the Assessment of Soil Erosion and Sedimentation using Environmental Radionuclides* (Zapata F, Ed). Kluwer, Dordrecht, The Netherlands 15-40.

Pennock DJ, Lemmon DS, De Jong E (1995) Caesium-137 measured erosion rates for five parent material groups in southwestern Saskatchewan. *Can. J. Soil Sci.* **75**: 205-210.

Pimentel D (2006) Soil erosion: A food and environmental threat. *Environment, Development and Sustainability* **8**: 119-137.

Pimentel D, Allen J, Beers A, Guinand L, Hawkins A, Linder R, McLaughlin P, Meer B, Musonda D, Perdue D, Poisson S, Salazar R, Siebert S, Stoner K (1993) Soil erosion and agricultural productivity. In Pimental D (Ed) *World soil erosion and conservation*, Cambridge: Cambridge University Press, pp. 277-92.

Pimentel D, Harvey C, Resosudarmo P, Sinclair K, Kurz D, Mcnair M, Crist S, Shpritz L, Fitton L, Saffouri R, Blair R (1995) Environmental and economic costs of soil erosion and conservation benefits. *Science* **267:**1117-1123.

Pegoyev AN, Fridman SD (1978) Vertical profiles of cesium-137 in soils (translation). *Pochvovedeniye* **8**: 77-81.

Porto P, Walling DE (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 DE (2012) Using plot experiments to test the validity of mass balance model semployed to estimate soil redistribution rates from ^{137}Cs and ^{210}Pb$_{ex}$ measurements. *Applied Radiation and Isotopes* **70**: 2451-2459.

Porto P, Walling DE, Alewell C, Callegari G, Mabit L, Mallimo N, Meusburger K, Zehringer M (2014) Use of a ^{137}Cs re-sampling technique to investigate temporal changes in soil erosion and sediment mobilisation for a small forested catchment in southern Italy. *Journal of Environmental Radioactivity* **138**: 137-148.

Porto P, Walling DE, Callegari G (2004) Validating the use of caesium-137 measurements to estimate erosion rates in three small catchments in southern Italy. In Golosov V, Belyaev V & Walling DE (Eds.): *Sediment Transfer through the Fluvial System* (Proceedings of a symposium held in Moscow, August 2004) International Association of Hydrological Sciences Publication **288**: 75-83. Wallingford, UK: IAHS.

Porto P, Walling DE, Callegari G (2005) Investigating sediment sources within a small catchment in Southern Italy. *IAHS Publ*. **291**: 113-122.

Porto P, Walling DE, Callegari G (2009) Investigating the effects of afforestation on soil erosion and sediment mobilisation in two small catchments in Southern Italy. *Catena* **79**: 181-188.

Porto P, Walling DE, Callegari G (2011) Using ^{137}Cs measurements to establish catchment sediment budgets and explore scale effects. *Hydrol. Process.* **25**: 886-900.

Porto P, Walling DE, Callegari G (2013) Using 137Cs and ^{210}Pbex measurements to investigate the sediment budget of a small forested catchment in southern Italy. *Hydrol. Process*. **27**: 795-806.

Porto P, Walling DE, Callegari G, Catona F (2006) Using fallout lead-210 measurements to estimate soil erosion in three small catchments in Southern Italy. *Water, Air, and Soil Pollution: Focus* **6**: 657-667.

Porto P, Walling DE, Cogliandro V, Callegari G (2016a) Exploring the use of the ^{137}Cs resampling technique to investigate recent changes in soil erosion rates in three small catchments in Southern Italy. Submitted to *Proceeding of the International Association of Hydrological Sciences (PIAHS).*

Porto P, Walling DE, Ferro V (2001) Validating the use of caesium-137 measurements to estimate soil erosion rates in a small drainage basin in Calabria, Southern Italy. *Journal of Hydrology* **248**: 93-108.

Porto P, Walling DE, Ferro V, Di Stefano C (2003) Validating erosion rate estimates by caesium-137 measurements for two small forested catchments in Calabria, Southern Italy. *Land Degradation and Development* **14**: 389-408.

Porto P, Walling DE, La Spada C, Callegari G (2016b) Validating the use of ^{137}Cs measurements to derive the slope component of the sediment budget of a small rangeland catchment in southern Italy. *Land Degrad. Develop*. **27**: 798-810.

Porto P, Walling DE, Tamburino V, Callegari G (2003) Relating caesium-137 and soil loss from cultivated land. *Catena* **53**: 303 - 326

Pourchet M, Pinglot JF, Reynauld L, Holdsworth G (1988) Identification of Chernobyl fallout as a new reference level in Northern Hemisphere glaciers. *Journal of Glaciology* **34**: 183-187.

Povinec PP, Gera M, Holý K, Hirose K, Lujaniené G, Nakano M, Plastino W, Sýkora I, Bartok J, Gažák M (2013) Dispersion of Fukushima radionuclides in the global atmosphere and the ocean. *Applied Radiation and Isotopes* **81**: 383-392.

Quine TA (1999) Use of caesium-137 data for validation of spatially distributed erosion models: the implications of tillage erosion. *Catena* **37**: 415-430.

REGIONE CALABRIA (2001) *Piano Stralcio di Bacino per l'Assetto Idrogeologico (PAI)* http://www.adbcalabria.it/.

Reynolds WD, Gillham RW, Cherry JA (1982) Evaluation of distribution coefficients for the prediction of strontium and cesium migration in a uniform sand. *Can. Geotech. J.* **19**: 92-103.

Ritchie JC, McHenry JR (1973) Determination of fallout Cs-137 and natural gamma-ray emitting radionuclides in sediments. *Appl. Radiat. Isot*. **24**: 575-578.

Ritchie JC, McHenry JR (1975) Fallout Cs-137: a tool in conservation research. *J. Soil Water Conserv*. **30**: 283-286.

Ritchie JC, McHenry JR (1990) Application of radioactive fallout caesium-137 for measuring soil erosion and sediment accumulation rates and patterns: A review. *J. Environ. Qual.* **19**: 215-233.

Ritchie JC, Ritchie CA (2005) Bibliography of publications of 137-cesium studies related to erosion and sediment deposition. USDA-ARS Hydrology and Remote Sensing Laboratory. Occasional Paper HRSL-2005-01.

Ritchie JC, Ritchie CA (2008) Bibliography of publications of 137cesium studies related to erosion and sediment deposition. USDA-ARS Hydrology and Remote Sensing Laboratory Occasional Paper HRSL-2008-02.

Rogowski AS, Tamura T (1970) (a) Erosional behaviour of cesium-137. *Health Physics* **18**: 467-477.

Rogowski AS, Tamura T (1970) (b) Environmental mobility of cesium-137. *Radiat. Bot.* **10**: 35-45.

Schmitter P, Dercon G, Hilger T, Thi Le Ha T, Huu Thanh N, Lam N, Duc Vien T, Cadisch G (2010) Sediment induced soil spatial variation in paddy fields of Northwest Vietnam. *Geoderma* **155**:298–307.

Schulz RK, Overstreet R, Barshad I (1960) On the soil chemistry of caesium-137. *Soil Sci*. **89**: 19-27.

Sepulveda A, Schuller P, Walling DE (2008) Use of ^{7}Be to document erosion associated with a short period of extreme rainfall. *J. Environ. Radioact*. **99**: 35-49.

Soil Survey Division Staff (1993) *Soil survey manual*. United States Department of Agriculture. pp. 62 - 65.

Sorriso-Valvo M, Bryan RB, Yair A, Iovino F, Antronico L. (1995) Impact of afforestation on hydrological response and sediment production in a small Calabrian catchment. *Catena* **25**: 89-104.

Squire HM, Middleton LJ (1966) Behaviour of ^{137}Cs in soils and pastures - a long term experiment. *Radiation Botany* **6**: 413-423.

Steinhauser G, Brandl A, Johnson TE (2014). Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts. *Science of the Total Environment* **470-471**: 800-817.

Stone PM, Walling DE (1997) Particle size selectivity considerations in sediment budget investigations. *Water Air Soil Pollut.* **99**: 63 - 70.

Stocking M (2003) Tropical soils and food security: The next 50 Years. *Science* **302**: 1356-1359.

Sutherland RA (1991) Examination of caesium-137 areal activities in control (uneroded) locations. *Soil Technol.* **4**: 33 - 50.

Sutherland RA (1996) Caesium-137 soil sampling and inventory variability in reference samples; A literature survey. *Hydrol. Process.* **10**: 43-53.

Tamura T (1964) Selective sorption reaction of caesium with mineral soils. *Nucl. Saf.* **5**: 262-268.

Tiessen KHD, Li S, Lobb DA, Mehuys GR, Rees HW, Chow TL (2009) Using repeated measurements of ^{137}Cs and modelling to identify spatial patterns of tillage and water erosion within potato production in Atlantic Canada. *Geoderma* **153**: 104-118.

Torri D (2004) An overview of the current research needs for improving soil erosion control, In: Francaviglia R (Ed.): *Agricultural Impacts on Soil Erosion and Soil Biodiversity: Developing Indicators for Policy Analyses*. Proc. OECD Expert Meeting, March 2003, Rome, Italy 59-74.

Toy TJ, Foster GR, Renard KG (2002) *Soil Erosion: Processes, Prediction, Measurement and Control*. John Wiley & Sons, Inc., New York, USA 352.

Tuo F, Xu C, Zhang J, Zhou Q, Li W, Zhao L, Zhang Q, Zhang J, Su X (2013) Radioactivity analysis following the Fukushima Dai-ichi nuclear accident. *Applied Radiation and Isotopes* **78**: 77-81.

UNEP (1992) *Global assessment of soil degradation*. Wageningen: ISRIC and Nairobi: UNEP

United Nations Environment Programme (2007) *Global Environment Outlook: Environment for Development GEO-4. Summary for Decision Makers*.

United Nations (2009) *UN Millennium Development Goals*.

Valentin C, Poesen J, Li Y (2005) Gully erosion: Impact, factors and control. *Catena* **63**: 132-153.

Van Oost K, Govers G, Van Muysen W (2003) A process-based conversion model for caesium-137 derived erosion rates on agricultural land: an integrated spatial approach. *Earth Surf. Processes Landforms* **28**: 187-207.

Wallbrink PJ, Murray AS (1993) The use of fallout radionuclides as indicators of erosion processes. *Hydrol. Process*. **7**: 297-304.

Wallbrink PJ, Murray AS, Olley JM (1999) Relating suspended sediment to its original soil depth using fallout radionuclides. *Soil Sci. Soc. Am. J.* **63**: 369-378.

Wallbrink PJ, Walling DE, He Q (2002) Radionuclide measurement using HPGe Gamma Spectrometry. In: Zapata F (Ed) *Handbook for the Assessment of Soil Erosion and Sedimentation using Environmental Radionuclides*. Kluwer, Dordrecht, The Netherlands 67-95.

Walling DE (1989) The struggle against water erosion and a perspective on recent researching. In Ivanov K and Pechinov D (Eds) *Water Erosion, UNESCO technical document in hydrology, SC-89/WS-57* Paris: UNESCO, pp. 39-60.

Walling DE (2000) Linking land use, erosion and sediment yields in river basin. *Hydrobiology* **410**: 223-240.

Walling DE (2001) Recent advances in the use of environmental radionuclides in soil erosion investigations. In: IAEA (Ed.), *Nuclear Techniques in Integrated Plant Nutrient, Water and Soil Management.* Proc. FAO/IAEA Int. Symp. Vienna, October 16-20, 2000. IAEA-CSP-11P, Vienna, Austria 279-301.

Walling DE (2006) Tracing versus monitoring: new challenges and opportunities in erosion and sediment delivery research. In: Owens PN, Collins AJ (Eds.) *Soil Erosion and Sediment Redistribution in River Catchments*. CABI, Wallingford, pp. 13-27.

Walling DE (2012) The use of fallout radionuclides in the study of erosion and sedimentation. In: Meyers RA (Ed.) *Encyclopedia of Sustainability Science and Technology* - Springer.

Walling DE, Bradley SB (1988) The use of caesium-137 measurements to investigate sediment delivery from cultivated areas in Devon. In: *IAHS Publication No. 174*, Wallingford, UK 325-335.

Walling DE, Collins AL (2000) *Integrated assessment of catchment sediment budgets: A technical Manual*, University of Exeter.

Walling DE, He Q (1993) Towards improved interpretation of ^{137}Cs profile in like sediments. In *Geomorphology and sedimentology of Lakes and Reservois*, McManus J, Duck RW (Eds). Wiley: Chichester, 31-53.

Walling De, He Q (1997) Models for converting ^{137}Cs measurements to estimates of soil redistribution rates on cultivated and uncultivated soils (including software for model implementation). *A Contribution to the IAEA Coordinated Research Programmes on Soil Erosion (DI.50.05) and Sedimentation (F3.10.01).*

Walling DE, He Q (1999) (a) Improved models for estimating soil erosion rates from cesium-137 measurements. *J. Environ. Qual*. **28**: 611-622.

Walling DE, He Q (1999) (b) Use of fallout ^{210}Pb measurements to estimate soil erosion on cultivated land. *Soil Sci. Soc. Am. J.* **63**: 1404-1412.

Walling DE, He Q (2000) The global distribution of bomb-derived ^{137}Cs reference inventories. Final Report on IAEA Technical Contract 10361/RO-R1. University of Exeter.

Walling DE, He Q (2001) Models for converting ^{137}Cs measurements to estimates of soil redistribution rates on cultivated and undisturbed soils (including software for model implementation). Report to IAEA. Univ. Of Exeter, Exeter, UK.

Walling DE, He Q, Appleby PG (2002) Conversion models for use in soil-erosion, soil-redistribution and sedimentation investigations. In: Zapata F (Ed) *Handbook for the Assessment of Soil Erosion and Sedimentation using Environmental Radionuclides.* Kluwer, Dordrecht, The Netherlands 111-164.

Walling DE, He Q, Whelan PA (2003) Using ^{137}Cs measurements to validate the application of the AGNPS and ANSWERS erosion and sediment yield models in two small Devon catchments. *Soil Tillage Research* **69**: 27-43.

Walling DE, He Q, Zhang Y (2014) Conversion models and related software. In: *Guidelines for using fallout radionuclides to assess erosion and effectiveness of soil conservation strategies*, *IAEA*. Vienna, pp. 125-148.

Walling DE, Quine TA (1990) Calibration of caesium-137 measurements to provide quantitative erosion rate data. *Land Degrad. Rehab.* **2**: 161-175.

Walling DE, Quine TA (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 DE, Quine TA (1992) The use of caesium-137 measurements in soil erosion surveys. In *Erosion and Sediment Transport Monitoring Programmes in River Basins* (Proc. of the Oslo Symposium). *IAHS Publ.* **210**: 143-152.

Walling DE, Quine TA (1993) *Use of Caesium-137 as a Tracer of Erosion and Sedimentation: Handbook for the Application of the Caesium-137 Technique*. University of Exeter.

Walling DE, Quine TA (1995) The use of fallout radionuclide measurements in soil erosion investigations. In: *IAEA* (Ed.), *Nuclear Techniques in Soil-Plant Studies for Sustainable Agriculture and Environmental Preservation*. Proc. FAO/IAEA Int. Symp. Vienna, October 17-21, 1994. IAEA Proc. Series STI/PUB/947, Vienna, Austria 597-619.

Walling DE, Schuller P, Zhang Y, Iroume A (2009) Extending the timescale for using beryllium-7 measurements to document soil redistribution by erosion. *Water Resour. Res*. 45

Walling DE, Wilkinson SN, Horowi AJ (2011) Catchment erosion, sediment delivery and sediment quality. *The Science of Hydrology* **2**: 305-338.

Walling DE, Zhang Y (2010) A national assessment of soil erosion based on caesium-137 measurements. *Adv. Geoecol*. **41**: 89-97.

Walling D, Zhang Y, He Q (2006) Models for Converting Measurements of Environmental Radionuclide Inventories (^{137}Cs, Excess ^{210}Pb, and ^{7}Be) to Estimates of Soil Erosion and Deposition Rates (Including Software for Model Implementation). Department of Geography, University of Exeter, Exeter, UK. Available from the senior author at: DE Walling@ Exeter. ac. uk.

Whitlow R (1986) Mapping erosion in Zimbabwe: a methodology for rapid survey using aerial photographs. *Applied Geography* **6**: 149-162.

Wischmeier WH, Smith DD (1978) Predicting Rainfall-erosion Losses. In *A Guide to Conservation Farming*, vol. 537. US Dept. of Agric., *Agr. Handbook*, p 151.

Young RA (1980) Characteristics of eroded sediment. *Trans. Am. Soc. Agric. Eng.* **23**: 1139 - 1142, 1146.

Young RA, Onstad CA (1978) Characterization of rill and interrill eroded soil. *Trans. Am. Soc. Agric. Eng.* **21**: 1126 - 1130.

Zapata F, Garcia-Agudo E, Ritchie JC, Appleby PG (2003) *Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides.* In Zapata F (Ed), Dordrecht (The Netherlands): Kluwer Academic Publishers.

Zapata F (Ed) (2003) Field Application of the Cs-137 Technique in Soil Erosion and Sedimentation. *Special Issue. Soil & Tillage Research* **69**: 153.

Zapata F, Nguyen ML (2009) Soil erosion and sedimentation studies using environmental radionuclides. In: Froehlich K (Ed.) *Environmental Radionuclides: Tracers and Timers of Terrestrial Processes,* Book series. Radioact. Environ. **16**: 295-322.

Zhang XB, Higgitt DL, Walling DE (1990) A preliminary assessment of the potential for using caesium-137 to estimate rates of soil erosion in the Loess Plateau of China. *Hydrol. Sci. J.* **35**: 267-276.

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.

List of papers produced during the PhD course

1) Porto P., Callegari, G., Cogliandro, V., La Spada, C. (2014). *Misura e previsione dell'erosione idrica superficiale mediante la tecnica del ^{7}Be. Il caso del bacino sperimentale W2 in provincia di Crotone.* Quaderni di Idronomia Montana 32/2, 97-108.

2) Capra C., Cogliandro V., Fosco D., La Spada C., Porto P. (2014) *Understanding climate changes in Southern Italy using the rainfall erosivity factor of the USLE*. Poster al Convegno internazionale: Hydrology in a Changing World, Environmental and Human Dimensions, FRIEND-Water 2014, 7th Global FRIEND-Water Conference, Montellier (France), 07-10 ottobre 2014.

3) Porto P., Walling D.E., Cogliandro V., Callegari G. (2016). *Validating a mass balance accounting approach to using ^{7}Be measurements to estimate event-based erosion rates over an extended period at the catchment scale*. Water Resources Research 52(7): 5285-5300.

4) Porto P., Walling D.E., Cogliandro V., Callegari G. (2016). *Exploring the potential for using $^{210}Pb_{ex}$ measurements within a re-sampling approach to document recent changes in soil redistribution rates within a small catchment in southern Italy*. Journal of Environmental Radioactivity 164: 158-168.

5) Porto P., Walling D.E., Cogliandro V., Callegari G. (2016). *Exploring the use of the ^{137}Cs re-sampling technique to investigate recent changes in soil erosion rates in three small catchments in Southern Italy*. Submitted to: Proceeding of the International Association of Hydrological Sciences (PIAHS).

Scientific Publication in preparation:

1) Cogliandro V., Banasik K., Porto P., Krajewski A., Rutkowska A. *Lag time of small forested catchment before and after a forest fire*. To be prepared for Sylwan

2) Cashman MJ, Gellis A, Gorman Sanisaca L, Noe G, Cogliandro V, Baker A (2017) *Bank-derived sediment dominates in a suburban Chesapeake Bay watershed, Upper Difficult Run, Virginia, USA*