A method for determining sediment budgets using caesium-137

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Abstract Caesium-137 is a globally distributed product of nuclear weapons tests which has widespread but under-utilized applications as a tracer of soil and sediment movements. Contrary to such equations as the Universal Soil Loss Equation, which are calibrated under limited conditions, the ^{137}Cs method provides real data which are specific to the site.   It may be used to identify sediment sources and storages, which are the most difficult components to quantify in a sediment budget.   This paper shows the simplicity of the technique and describes technical aspects often neglected in scientific papers.   Starting with the basin, the criteria for site selection, including reference sites, and techniques for sampling are described.   In the laboratory, simple treatment of the samples, followed by gamma-ray spectrometry enables calculation of ^{137}Cs concentration in various units (g^{-1}, cm^{-2}).   Comparison of ^{137}Cs values relative to a reference site shows the gain or loss of ^{137}Cs time-averaged for the period 1954–1988.   Construction of iso-caesium lines may reveal patterns of soil loss but nevertheless soils can be ranked for apparent soil degradation.   The authors have calibrated, within confidence limits, the measured ^{137}Cs loss *vs.* measured net soil loss ($kg\ ha^{-1}\ year^{-1}$) and also correlated these variables with productivity yields.   This allows them to produce maps of estimated net soil loss in the basin.   At deposition sites absolute values for the sedimentation rates for the period 1954–1988 have been obtained.   Sediment collected by water sampling at basin outlets can also be retrieved for ^{137}Cs "fingerprinting".

Une méthode pour déterminer la quantité de sédiment au moyen du césium-137

Résumé   Le césium-137 est un produit globalement réparti qui provient des essais d'armes nucléaires et a des applications générales, mais sous-utilisées comme traceur de mouvements du

sol et des sédiments. Contrairement aux équations comme l'équation de la perte universelle de sol, qu'on vérifie sous des conditions limitées, la méthode de ^{137}Cs fournit des données réelles lesquelles sont spécifiques d'un site. Elle peut être employée pour identifier des sources et dépôts des sédiments, qui sont les composantes les plus difficiles à quantifier dans un bilan de sédiments. Cette communication montre la simplicité de la technique et décrit les aspects techniques lesquels sont souvent négligés dans des études scientifiques. Le travail commence par le bassin et décrit les critères pour la sélection du site, y compris les sites de référence, et les techniques d'échantillonnage. Au laboratoire, un traitement simple des échantillons, suivi par la spectrométrie gamma, nous permet de calculer la teneur en ^{137}Cs en différentes unitées (g^{-1}, cm^{-1}). La comparaison des valeurs du ^{137}Cs relatives à un site de référence montre le gain ou la perte de ^{137}Cs, adoptant comme base la moyenne de 1954 à 1988. L'établissement des lignes d'iso-césium peut mettre à jour la distribution de la perte de sol, néanmoins, des sols peuvent être classées pour la dégradation apparente du sol. Les auteurs ont calculé, avec intervalle de confiance, la perte de ^{137}Cs, mesurée en fonction de perte nette de sol (kg $ha^{-1}an^{-1}$), et ont mis ces variables en corrélation avec des rendements de productivité. Cela leur permet de produire des cartes de la perte nette de sol estimée au bassin. Au site du dépôt, les valeurs absolues des taux de sédimentation ont été obtenues pour la période de 1954 à 1988. Le sédiment collectionné par l'échantillonnage d'eau aux sorties du bassin peut être encore recupéré pour "l'examen dactyloscopique" au moyen du ^{137}Cs.

INTRODUCTION

All lands are potential sediment sources. The erosion is accelerated by socio-economic pressures either to bring new lands into production, to increase their existing productivity or to change existing land uses. Conservation and other environmental groups have to compete within this developmental framework. An appropriate basis for decision on land use limitations would, in all cases, be a comprehensive sediment budget in which the spatial and temporal variability of the sediment sources can be measured and their real contributions to the total catchment sediment yield determined. Sediment delivery ratios are usually coupled with estimates of sheet and rill erosion from the Universal Soil Loss Equation (USLE) or a similar equation. The geomorphological diversity of basins being subjected to developmental pressures are such that conventional sedimentological techniques are not always successful. For example, the USLE may be used to predict sheet and rill erosion but the values of the parameters may lie outside the constraints under which it was developed. Radioactive tracers provide an alternative means of evaluating sediment budgets. By monitoring the tracer's major property, the type of radiation emitted, it is possible to determine the nature

of the material in both its static and dynamic states. In addition, the radioactive half-life of the tracer allows a time frame to be assigned to the build-up of the sediment budget.

Since 1954 this capability has been present in all surface soils exposed to precipitation or gravitational settlement of fallout debris from atmospheric nuclear weapons tests. As a result of these tests large amounts of caesium-137 (^{137}Cs, $T_{1/2}$ 30.2 years, $E_{1/2}$ 662 keV) were globally distributed. Chemically and physically, ^{137}Cs is very rapidly and strongly adsorbed onto soil colloids. When the soil particles are redistributed by erosion then real data specific to the site are produced. These data identify the sources of sediment and resolve their spatial and temporal variations, which are the most difficult components of a sediment budget to quantify.

DATA SOURCES

Results of the global monitoring of major radioactive species in fallout were published in reports of the Health and Safety Laboratory of the US Energy Research and Development Administration (Hardy, 1977), the Environmental Measurements Laboratory of the US Department of Energy (Larsen, 1984) and the United Kingdom Atomic Energy Authority (Cambray, 1985). Other countries, principally Denmark, France and Japan, maintain their own national environmental surveillance systems. From these various sources the total input of ^{137}Cs at specific locations may be calculated either directly or from the ^{137}Cs/^{90}Sr data according to the procedure of Stewart *et al.* (1957).

Important characteristics of the patterns of global fallout of ^{137}Cs are (a) the first significant appearance, 1953–1955, (b) a substantial decrease from 1959–1961, (c) accelerating fallout from 1962 with a maximum in 1964–1965, (d) atmospheric depletion from 1965 to present, except for perturbations due to atmospheric tests by France (1966–1974) and the Peoples Republic of China (1963–1980). By 1986 the concentrations of ^{137}Cs and other long-lived fission products in air and rain were near to or below the limit of detection (Cambray, *et al.*, 1985). However, during the uncontrolled phases of the Chernobyl reactor incident in 1986 significant amounts of ^{137}Cs and various other radioactive species were dispersed to parts of the Northern Hemisphere. Thus, relabelling of soil minerals with ^{137}Cs has occurred which will present many new opportunities to study its distribution in soils and sediment transport in the environment.

GAMMA-RAY SPECTROMETRY

All gamma rays emitted from soils and sediments originate from traces of uranium and thorium and their daughter decay products, potassium-40, cosmogenically produced radionuclides (e.g. beryllium-7) and residual radioactive species in fallout from either nuclear weapons tests or the operations associated with nuclear power reactors. These radionuclides all have a characteristic gamma-ray energy spectrum. Resolution of the individual

photopeaks is best achieved with asolid state detector system, such as the hyperpure germanium crystal detector systems which operate at liquid nitrogen temperature. Their relative efficiency for photon detection is usually 15–30%, and, for analytical purposes, the higher efficiency is more advantageous. It should be noted that the cost of the detector increases markedly with efficiency.

Quantitative analysis of the radionuclide is a non-destructive procedure, and is based on the relationship between net peak area and concentration:

$$\frac{\text{concentration in sample}}{\text{concentration in standard}} = \frac{\text{net peak area of sample}}{\text{net peak area of standard}}$$

Radioactive standards are available from the US National Bureau of Standards, Washington, DC (Homogeneous River Sediment for Radioactivity Measurements, Research Material 45b) and the International Atomic Energy Agency, Vienna (Reference Material, Soil-6). Urquhart (1973) has listed all gamma-ray peaks from the uranium/thorium series and Beck (1980) listed gamma emitters in the environment. These data show that the 662 keV gamma-ray of ^{137}Cs is well separated in the energy spectrum from other peaks. The gamma activity of the sample is counted in a Marinelli beaker for up to 8 h for the more active samples or up to 16 h for low activity samples. The exact time taken is that which produces an acceptable statistical error or an answer of geomorphological significance. Radioactivity is a statistical process: the estimate of uncertainty at the 1 σ level is calculated using standard procedures described in Faires & Parks (1973), Currie (1968), and Debertin (1980). The unit of radioactivity is the becquerel (1 nuclear transition per second) and a convenient expression for the concentration of ^{137}Cs in soil is millibecquerels per gram (mBq g^{-1}). Following the practice of Ritchie *et al.* (1975), we express the activity per gram of clay, or per gram of clay and silt, since these are the fractions most associated with ^{137}Cs. Measurements of the photopeak area can be made in various ways (Covell, 1959; Baedecker, 1971). We use the total peak area (TPA) method coupled with a five point cubic smoothing of the data to reduce statistical fluctuations (Savitzky & Golay, 1964). Personal computers are readily available with program cards and software that convert them to a powerful multichannel analyser and at the same time provide rapid, economical means for the necessary data reduction. The total activity per unit area (mBq cm^{-2}) is determined from the expression $A = c\,m\,a^{-1}$, where c is the concentration (mBq g^{-1}), m is the total weight of dried sample, and a is the area of the frame or tube sampler (cm^{-2}).

SAMPLING METHODS FOR SOILS

Soil samples are scraped at depth increments using a hardened aluminium blade, bevelled on the leading edge to 60°. Down each side of the blade are holes at 1 cm intervals to index a 12 mm diameter steel rod that acts as a

depth guide (Fig. 1A). The area sampled is defined within a 200 × 500 mm steel frame made from 30–45 mm wide angle iron.   The frame is hammered into the soil until its upper edge is level with the soil surface. In the final scraping of each depth increment the blade is held vertical.   A modified version has the scraper blade held at a fixed cutting angle of either 45° or 60°, depending on the soil conditions (Fig. 1B).   At sites where only the total ^{137}Cs is required, a strong steel cylinder (100 mm i.d.) is inserted into the soil to a depth of 200 mm on uncultivated land, or 300 mm on cultivated land. The depth of sampling can be varied but it must always intersect the entire ^{137}Cs distribution.   Over-sampling into soil which contains no ^{137}Cs lowers the ^{137}Cs concentration in the sample; this may necessitate a longer counting time to improve the counting error ($\sigma = \sqrt{N}$ where N is the total number of counts in the net peak area). At depositional sites samples can be taken at 5 cm or other convenient increments by trenching or coring. The soil sample is prepared for gamma-ray spectroscopy by drying at 105°C and either sieving, or crushing and sieving to a predetermined standard.   This achieves a packing geometry in the Marinelli beaker similar to that used in calibration.
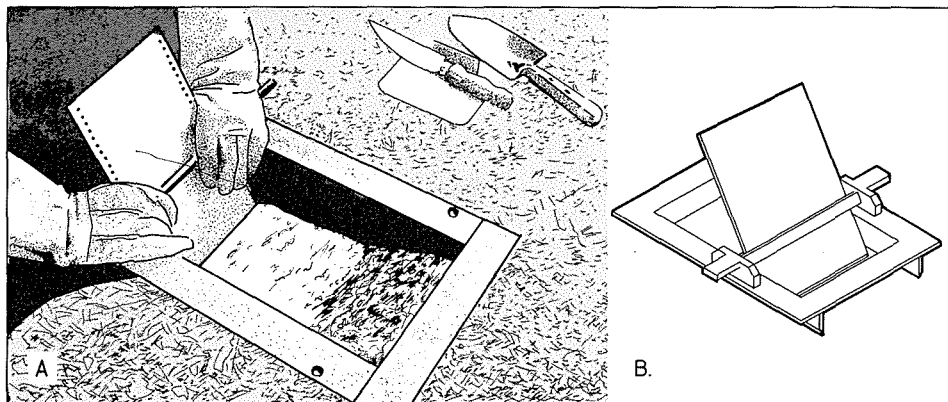

Fig. 1   Incremental soil samplers: (A) free angle (B) fixed angle.

CRITERIA FOR SELECTION OF REFERENCE INPUT SITES

Caesium-137 input data from meteoric measurement sites (see DATA SOURCES) can only be a useful guide to reference values for ^{137}Cs within drainage basins. They should not be a substitute for the actual measurement of wet and dry fallout on soils within the study area if suitable, undisturbed, sites can be found.   Reference input sites should generally receive little or no run-on, such as typically occurs at interfluves and plateau areas of hilltops. Areas with a high surface stone content are to be avoided. Evidence of soil stability can be established by (a) the presence of lichens on the surface, (b) the presence of a thick, friable topsoil, (c) the presence of a stable

vegetative cover, for example, a forest or perennial grass. Indicators of an unstable site are (a) evidence of deflation; e.g. the occurrence of grass and stone pedestals, (b) tree root or tree bole exposure, (c) the presence of weeds or annual grasses, (d) accumulation of sediment on the upslope side of fallen branches or tree boles, and (e) evidence of soil loss around rock outcrops. Anomalous areas to be avoided are macro/meso faunal features, e.g. termites in no-till areas or stockcamps.

CHARACTERISTIC DISTRIBUTIONS OF ^{137}Cs

Stable soil sites

Typically, the ^{137}Cs is concentrated in the surface layers and decreases exponentially with depth (approximately 60% per cm of depth, Campbell *et al.*, 1982). Truncation of this profile will indicate erosion, even of a relatively thin layer of surface soil (Campbell *et al.*, 1986). Land stability, time-averaged over the last 34 years, is reflected in the total amount and a characteristic distribution of ^{137}Cs in the soil profile (Loughran *et al.*, 1981, site 3 of Table 3).

Land use impacts

Grazing Degradation of land due to overgrazing, drought, soil acidification, soil salinization, loss of organic matter and soil loss from sheet and rill erosion may be obvious. However, discrimination of the effects of soil erosion is generally not possible. Caesium-137 concentrations provide real and effective data to describe the land condition. Caesium-137 values ranging from 26 to 137 mBq cm^{-2} have been measured in New South Wales (Campbell *et al.*, 1982, 1986, 1987; Loughran *et al.*, 1981: Table 3, site 1 – stable forest, sites 2 and 6 – grazed slopes). These values represent ^{137}Cs losses of 70% to zero, respectively, compared with reference input values.

Cultivation Cultivation mixes ^{137}Cs down to the plough or working depth with various degrees of efficiency and the ^{137}Cs distributions generally approximate a rectangular form (Loughran *et al.*, 1987). Lines of equal ^{137}Cs content have been used qualitatively to map areas of erosion and accumulation (Longmore *et al.*, 1983; Mitchell *et al.*, 1981; Campbell *et al.*, 1986; Martz & de Jong, 1987). The relabelling of exposed soil surfaces, loss of soil in successive erosion events and re-mixing from land use are not problems when a reference site can be obtained to index the local ^{137}Cs input amount. Values in the Maluna drainage basin, in the Hunter Valley region of New South Wales, ranged from zero to 90 mBq cm^{-2}, compared with a reference value of 104 mBq cm^{-2} (Campbell *et al.*, 1986, Fig. 1). High variability between sites is a frequent feature of ^{137}Cs measurements. It should be emphasized that each of these values is a useful expression of the geomorphic or topographic response to the magnitude of the erosion forces at

that location.   For this reason, the mean or other value of the central tendency are less useful than the actual values.

APPLICATION OF ^{137}Cs IN SEDIMENT BUDGET STUDIES

A method of applying ^{137}Cs measurements to sediment budget studies is shown in Fig. 2.   To identify sediment sources within the basin, levels of ^{137}Cs can be used to rank soil erosion status.   For example, the amount of ^{137}Cs revealed the stability of a hillslope after clear cutting (McIntyre *et al.*, 1987).   Where the ^{137}Cs levels can be calibrated against measurements of net soil loss, sediment sources may be quantified.   At sedimentation sites, ^{137}Cs can provide direct information on rates of accumulation since 1954 and other useful temporal variations linked to the fallout pattern.   For example, Loughran & Campbell (1983) identified more than 1 m of sedimentation as a result of viticulture on a steep hillslope.
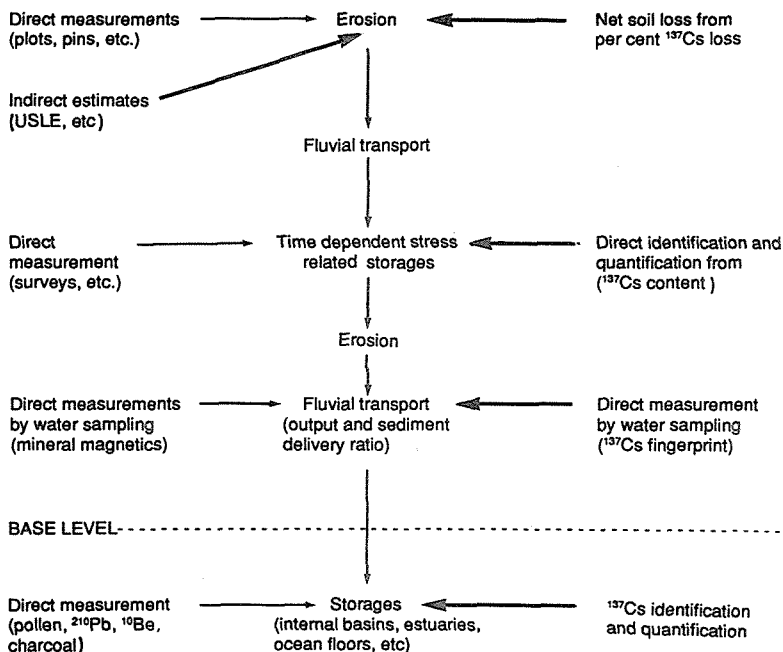

Fig. 2   Schematic representation of methodologies for sediment dynamics in a drainage basin system.

The ^{137}Cs technique is sufficiently precise to define the effect of erosion on productivity (Elliott *et al.*, 1988) and therefore to define the socio-economic effects of land use management or its change in addition to the physical effects.   We have used this ^{137}Cs method to obtain sediment budget in the Maluna drainage basin (Loughran *et al.*, in preparation).

CONCLUSIONS

The ^{137}Cs technique for measuring net soil loss has an advantage over plot and pin measurements, since there is no artifact of measurement. This universal tracer has the additional capability of being able to measure sedimentation and to "fingerprint" sediment sources. Even though specialized equipment is required for ^{137}Cs analyses, the field and laboratory methods for sediment budget construction are straightforward as outlined in this paper.

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