



# Quantifying geostatistical properties of $^{137}\text{Cs}$ and $^{210}\text{Pb}_{\text{ex}}$ at small scales for improving sampling design and soil erosion estimation



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## ABSTRACT

Knowledge of spatial structures of the radionuclides  $^{137}\text{Cs}$  and unsupported  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_{\text{ex}}$ ) is vital for developing sound sampling designs that are crucial for deriving quantitative soil erosion estimates. The objectives are to characterize spatial structures of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{ex}}$  inventories at small spatial scales under different land uses, and to quantify the effects of core sizes on (1) estimated sample means and variances of the  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{ex}}$  inventories and (2) sample numbers required for estimating the mean inventories at a given confidence level. Three different core sizes were used to take soil samples along three 10-m transects at 0.25-m or 0.5-m intervals for each land use. Land uses included cropland, grassland, forestland, and rangeland. 330 samples were analyzed for  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{ex}}$  inventories. Semivariograms were obtained by plotting empirical semivariances with sample separation distances. The spatial correlation distances ranged from 0.2 to 0.75 m for most cases. The semivariances at the separation distances of  $> 0.75$  m were close to the variances of the fields for all four land uses, indicating that the spatial distributions of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{ex}}$  were nearly stationary and had little spatial dependence at scales between 0.75 and 5 m. The overall results suggested that samples taken at a separation distance of  $> 0.75$  m would be largely independent and could be composited to form a representative sample for the sampling location for most cases. Given the large spatial variability at such a small scale, quantitative soil erosion rates cannot be estimated for a single soil core, because remarkably different soil erosion rates can be estimated for soil cores taken within a meter. Core size variation between 38 mm and 86 mm has no apparent effect on estimating sample means and sample standard deviations of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{ex}}$  inventories, except for gravelly soils. In general, 15–30 samples are needed to estimate  $^{137}\text{Cs}$  reference inventory on reference sites, but may be more for gravelly soils. More samples are required for forest and cultivated sites than for uncultivated grassland sites. On measuring sites, it is strongly recommended that 5–15 samples be taken for a grid point and measured individually if feasible or as a combined sample to allow reliable estimation of the mean  $^{137}\text{Cs}$  inventory for the location for most soils and land uses. If samples are taken for each of uniformly eroded land form units, the same number of individual samples as on the reference site are recommended. This work will be useful to improving sampling designs and consequently the accuracy of soil erosion estimation of the  $^{137}\text{Cs}$  technique.

## 1. Introduction

The fallout radionuclide  $^{137}\text{Cs}$  was released to the atmosphere during the atomic bomb testing primarily during the late 1950s and early 1960s, and fell back to the earth surfaces mainly in rain water

during that period. The  $^{137}\text{Cs}$  ion was rapidly adsorbed by and bound with fine soil particles. The fallout  $^{137}\text{Cs}$  has been widely used as a sediment tracer to estimate soil erosion rates in the past 40 years (Walling and He, 1999; Zapata, 2010; Mabit et al., 2009, 2013). The tracing method is based upon the assumption that spatial distribution of

**Abbreviations:** CI, confidence interval; CV, coefficient of variation; IAEA, International Atomic Energy Agency; i.d., inner diameter; RE, relative error; SD, standard deviation; SE, standard error; Std, standard

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$^{137}\text{Cs}$  fallout was initially uniform (Walling and Quine, 1992). Based on that assumption, soil erosion at any sampling point can be estimated by directly comparing the  $^{137}\text{Cs}$  inventory at the point with the mean reference inventory estimated on a reference site that experienced neither erosion nor deposition. The ability to retrospectively estimate average soil erosion rates for individual points is considered an important advantage of the  $^{137}\text{Cs}$  technique (e.g., Walling and Quine, 1992; Walling et al., 1995; Walling and He, 1998; Zapata, 2010). However, the uniform assumption was challenged by Parsons and Foster (2011), who argued that the initial spatial distribution of  $^{137}\text{Cs}$  was not uniform. Zhang (2014) showed that the spatial distribution of  $^{137}\text{Cs}$  could be assumed uniform at a large scale so long as long-term average rainfall was uniform at this scale. Since  $^{137}\text{Cs}$  was deposited in wet fallout over the period of 20 years, it would be expected that long-term total rainfall depth was uniform at a relatively large scale, and hence so was the input flux of  $^{137}\text{Cs}$ . Zhang (2014) also reported that spatial distribution of  $^{137}\text{Cs}$  was not uniform at a small scale due to rainfall redistribution by plants and surface roughness. The non-uniformity was largely caused by random spatial differences in vegetation interception, vegetation type and cover, surface residue cover, soil properties, water infiltration rates, and micro-topography. Such random variation in  $^{137}\text{Cs}$  inventory can be smoothed out by taking more independent samples in the statistically based sampling designs.

To date, random spatial variability of  $^{137}\text{Cs}$  inventory on both reference and measuring sites is not seriously considered when applying the  $^{137}\text{Cs}$  method to estimate soil erosion rates, although spatial  $^{137}\text{Cs}$  variability has been reported in the literature. On reference sites, a typical 20% coefficient of variation (CV) of  $^{137}\text{Cs}$  inventory has been reported (Sutherland, 1996; Bernard et al., 1998; Basher, 2000; Fornes et al., 2005; Mabit et al., 2009). A 20% CV means that about 15 samples are needed to quantify the mean reference inventory with an allowable relative error of 10% at the 95% confidence level. However, a review by Sutherland (1996) showed that only one third of the studies used sufficient number of samples to determine the mean reference inventory. Compared with spatial variability on reference sites, there are additional variation sources of  $^{137}\text{Cs}$  inventory on cultivated sites. Lance et al. (1986) reported that the mean sample variance of  $^{137}\text{Cs}$  inventories of 17 transects along contour lines in a cultivated slope plot was  $99.6 \text{ Bq}^2 \text{ core}^{-2}$ , while that in an adjacent native tallgrass prairie plot was  $34.8 \text{ Bq}^2 \text{ core}^{-2}$ . Sutherland (1994) reported that spatial variability in a cultivated field was 55% greater than that in an adjacent undisturbed field. The increased variance on an eroding site would substantially reduce the sensitivity of the  $^{137}\text{Cs}$  technique in detecting soil erosion (Kirchner, 2013).

Total variability of the measured  $^{137}\text{Cs}$  inventories on a reference site is generally composed of (1) random spatial variability due to small scale variations in soil, vegetation, bioturbation, and micro-topography; (2) sampling errors; and (3)  $^{137}\text{Cs}$  measurement errors (Owens and Walling, 1996). The sampling and measuring errors are random and are inherently included in the measured  $^{137}\text{Cs}$  inventories, which are generally < 10% each (Sutherland, 1991; Owens and Walling, 1996). The random spatial variability is the prevalent source of  $^{137}\text{Cs}$  variability. Lettner et al. (2000) analyzed the sources of  $^{137}\text{Cs}$  variability and reported a total CV of 21.5%, most of which was caused by the intrinsic spatial variability. An in-depth sensitivity and uncertainty analysis shows that soil redistribution estimates are most sensitive to both reference and sample inventories of  $^{137}\text{Cs}$ , and that spatial variability on both reference and measuring sites is the predominant contributor to overall uncertainty of soil erosion estimation, showing that close attention must be paid to  $^{137}\text{Cs}$  spatial variability (Zhang et al., 2015a). In the presence of large random variations in  $^{137}\text{Cs}$  spatial distributions, the  $^{137}\text{Cs}$  technique cannot be used to quantitatively estimate point soil erosion rate using a single soil core as is widely perceived in the literature, simply because part of the  $^{137}\text{Cs}$  variation is caused by spatial random variation rather than soil erosion (Zhang, 2014; Zhang, 2017b). The random  $^{137}\text{Cs}$  variation can only be reduced by increasing

independent sample numbers on both reference and measuring sites. Given substantial spatial variations on both reference and erosion sites, large sample numbers are often required to obtain reliable mean estimates of  $^{137}\text{Cs}$  inventory and consequently soil erosion rates.

While  $^{137}\text{Cs}$  variability is not usually considered in erosion studies, it should be pointed out that there were efforts made in several studies in which multiple samples were taken in close vicinity and were composited to obtain representative  $^{137}\text{Cs}$  inventory for the sampling location (e.g., Sutherland, 1994; Owens and Walling, 1996; Ritchie et al., 2009; Porto et al., 2009; Liu et al., 2017; Zhang, 2017a). However, sample spacings or separations in those studies were haphazardly chosen without considering the spatial structures of  $^{137}\text{Cs}$  inventory distributions due to lack of the spatial correlation data at small scales. This is because statistically based sampling designs that allow estimation of spatial structure of  $^{137}\text{Cs}$  distributions at small scales are not commonly employed in most studies. Typically grid spacings of 10 to 30 m are used in the literature. Pennock and Appleby (2010) analyzed the available spatial data and suggested that the separation distance for taking independent samples should be at least 10 m. Nevertheless, closer sampling distances must be explicitly studied for better sampling designs and for determining minimum distances for taking independent samples at small spatial scales (Zhang, 2014; Zhang et al., 2015a).

Sample number largely depends on the magnitude of spatial variability of  $^{137}\text{Cs}$  inventory. Characterizing spatial variability of  $^{137}\text{Cs}$  as well as its spatial structure at small scales is crucial to developing statistically sound experimental designs and therefore to obtaining reliable soil erosion estimation. This kind of information is essential to determine where to take independent samples and how many replicates are needed to obtain a representative sample for a location. However, such information including semi-variograms at small scales is almost nonexistent in the published literature. Thus, exploratory studies are needed to characterize spatial features of  $^{137}\text{Cs}$  distributions at small scales to improve the  $^{137}\text{Cs}$  technique. In addition, sample number may also be influenced by sampling area (i.e., corer size). Steel cylinder corers with inner diameters of 5 to 10 cm are commonly used for sampling. It is intuitive that fewer replicates may be needed for larger corers to obtain a representative composite sample due to presumed strong spatial correlation within 10 cm. Nevertheless, there is no information available in the literature about the effects of corer sizes on sample number, including for studies that used composited samples. This information is needed for better sampling designs and more accurate erosion estimation with the  $^{137}\text{Cs}$  method.

The objectives of this study are to characterize the spatial structures of  $^{137}\text{Cs}$  and unsupported  $^{210}\text{Pb}$  inventories within 5 m under different land uses at two locations in southern U.S., and to quantify the effect of corer sizes on calculated sample variances of  $^{137}\text{Cs}$  and unsupported  $^{210}\text{Pb}$  as well as on sample numbers required to obtain an practically acceptable inventory estimate and consequently soil erosion rate at a given confidence level.

## 2. Materials and methods

### 2.1. Site description and soil property

Two locations were chosen in this study. One is the Empire Ranch (Lat.  $31^\circ 42' 27''\text{N}$ ; Long.  $110^\circ 35' 22''\text{W}$ ), 16 km north of Sonoita, Arizona. Another is the experimental station at the Grazinglands Research Laboratory (Lat.  $35^\circ 32' 25''\text{N}$ ; Long.  $98^\circ 02' 59''\text{W}$ ), 7 km west of El Reno, Oklahoma. The long-term average annual precipitation and temperature near the Empire Ranch are 480 mm and  $15^\circ\text{C}$  with a semiarid climate. The landscape is formed on an inter-mountain area in basin and range physiography, and the vegetation is a typical semiarid rangeland (c.f. Fig. 1). The soil is a gravelly loam (fine, mixed, superactive, thermic Ustic Haplargids) with 17% rocks of > 2 mm. Sand and clay account for 60% and 12%. Three different land uses of cropland, grassland (tallgrass prairie), and forestland are selected at the

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