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

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ABSTRACT

Knowledge of spatial structures of the radionuclides ^{137}Cs and unsupported ^{210}Pb (^{210}Pb$_{ex}$) 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}Cs and ^{210}Pb$_{ex}$ 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}Cs and ^{210}Pb$_{ex}$ 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}Cs and ^{210}Pb$_{ex}$ 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}Cs and ^{210}Pb$_{ex}$ 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}Cs and ^{210}Pb$_{ex}$ inventories, except for gravely soils. In general, 15–30 samples are needed to estimate ^{137}Cs reference inventory on reference sites, but may be more for gravely 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}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}Cs technique.

1. Introduction

The fallout radionuclide ^{137}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}Cs ion was rapidly adsorbed by and bound with fine soil particles. The fallout ^{137}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}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}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}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}Cs was not uniform. Zhang (2014) showed that the spatial distribution of ^{137}Cs could be assumed uniform at a large scale so long as long-term average rainfall was uniform at this scale. Since ^{137}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}Cs. Zhang (2014) also reported that spatial distribution of ^{137}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}Cs inventory can be smoothed out by taking more independent samples in the statistically based sampling designs.

To date, random spatial variability of ^{137}Cs inventory on both reference and measuring sites is not seriously considered when applying the ^{137}Cs method to estimate soil erosion rates, although spatial ^{137}Cs variability has been reported in the literature. On reference sites, a typical 20% coefficient of variation (CV) of ^{137}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}Cs inventory on cultivated sites. Lance et al. (1986) reported that the mean sample variance of ^{137}Cs inventories of 17 transects along contour lines in a cultivated slope plot was 99.6 Bq^2 core^{-2}, while that in an adjacent native tallgrass prairie plot was 34.8 Bq^2 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}Cs technique in detecting soil erosion (Kirchner, 2013).

Total variability of the measured ^{137}Cs inventories on a reference site is generally composed of (1) random spatial variability due to small scale variations in soil, vegetation, bioturbidity, and micro-topography; (2) sampling errors; and (3) ^{137}Cs measurement errors (Owens and Walling, 1996). The sampling and measuring errors are random and are inherently included in the measured ^{137}Cs inventories, which are generally < 10% each (Sutherland, 1991; Owens and Walling, 1996). The random spatial variability is the prevalent source of ^{137}Cs variability. Lettner et al. (2000) analyzed the sources of ^{137}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}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}Cs spatial variability (Zhang et al., 2015a). In the presence of large random variations in ^{137}Cs spatial distributions, the ^{137}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}Cs variation is caused by spatial random variation rather than soil erosion (Zhang, 2014; Zhang, 2017b). The random ^{137}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}Cs inventory and consequently soil erosion rates.

While ^{137}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}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}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}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}Cs inventory. Characterizing spatial variability of ^{137}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}Cs distributions at small scales to improve the ^{137}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 to10 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}Cs method.

The objectives of this study are to characterize the spatial structures of ^{137}Cs and unsupported ^{210}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}Cs and unsupported ^{210}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° 42′ 27″N; Long. 110° 35′ 22″W), 16 km north of Sonoita, Arizona. Another is the experimental station at the Grazinglands Research Laboratory (Lat. 35° 32′ 25″N; Long. 98° 02′ 59″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 °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 gravely 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

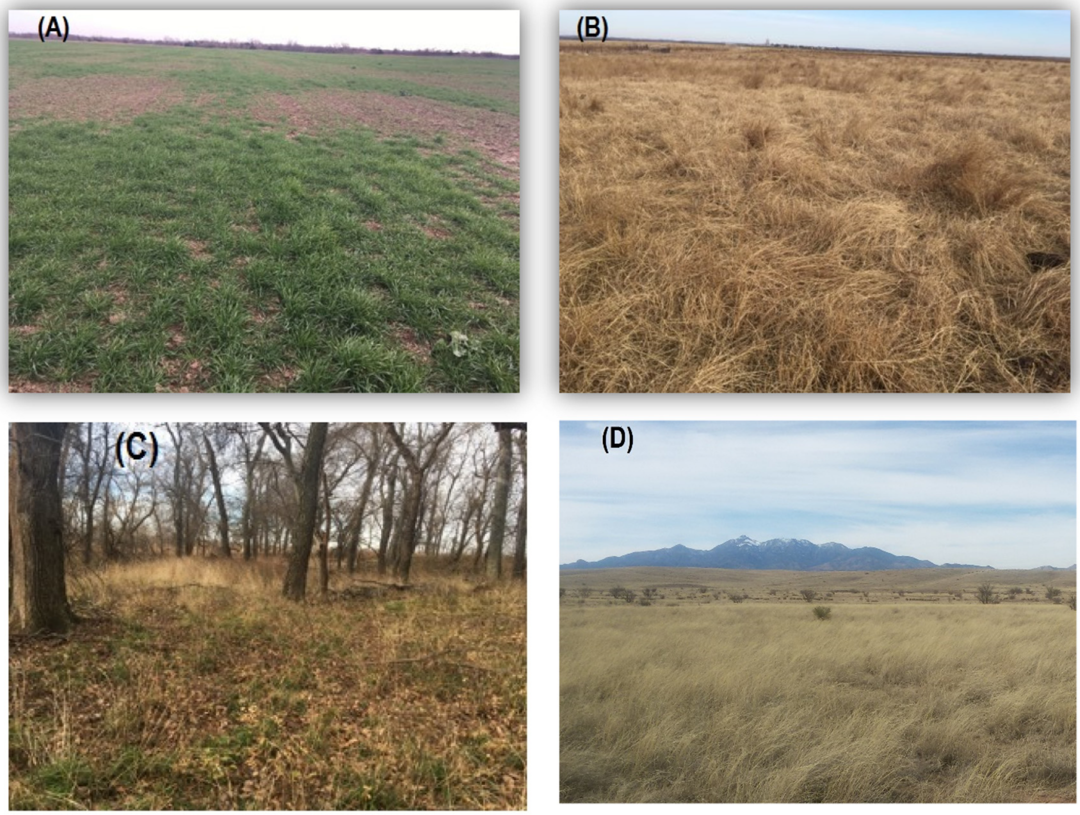

Fig. 1. Cultivated land (A), native prairie tall grassland (B), and forestland (C) at the Grazinglands Research Laboratory near El Reno, Oklahoma; and rangeland (D) on the Empire Ranch near Sonoita, Arizona.

Table 1
Soil particle size distribution on each sampling site at two locations.

Location	El Reno, Oklahoma			Empire Ranch, Arizona
Site	Cropland	Grassland	Forestland	Rangeland
Sand, %	18.6	28.6	62.1	59.5
Silt, %	50.6	49.2	18.8	11.6
Clay, %	23.9	16.8	14.5	11.9
< 2 mm rock, %	6.9	5.4	4.6	17.0
Organic carbon, %	1.2	1.7	1.4	1.0

experimental station near El Reno (Fig. 1). The sampling sites are relatively flat with slopes being < 2%. The long-term average annual precipitation and temperature are 886 mm and 15 °C. The soils (fine, mixed, thermic, Udertic or Pachic Paleustolls) are primarily silt loams on the cropland and grassland sites with approximately 50% silt and 17–24% clay (Table 1). The soil on the forestland site near the North Canadian River bank has 62% sand and 16% clay. The organic carbon contents on the three sites follow the sequence of grassland > forestland > cropland. The cropland site has been cultivated primarily for winter wheat for > 30 years. The grassland site is under native tallgrass prairie and has been grazed periodically with cattle. The forest site, which is under deciduous trees, has not been disturbed since the 1940's.

2.2. Sampling tools and sampling design

A Gidding's hydraulic soil sampler, mounted in the back of a truck, was used to take soil cores at the El Reno station in the fall of 2016. A soil sampling probe composed of a steel tube and a tube bit with a beveled smooth cutting edge was vertically pushed into soil by fluid hydraulics. The sampling depth was at least 300 mm for all three land uses, and samples were taken along straight transects. One soil core size of a 51-mm inner diameter (i.d.) was used on the cropland site. Fifty-one soil cores along a 10-m transect were taken at 200-mm intervals along the transect. Three core sizes of 51, 65, and 86 mm (i.d.) were used to take cores along the three parallel transects (one for each core diameter) spaced 100 mm apart on the native tallgrass site (Fig. 2). The samples were taken at 250-mm intervals for the first 5 m and at 500-mm intervals for the second 5 m. A total of 31 cores for each core size were taken. The same sampling scheme but with different core sizes of 38, 51, and 86 mm (i.d.) was used on the forest site.

Because of the high gravel content in the soil on the Empire Ranch, the straight core sampler with a smooth tube bit as used on the El Reno sites could not be used. A rotating auger with two teeth had to be employed, although the sampling area was somewhat difficult to control due to the interference of large rocks. Three core sizes of 44, 57, and 83 mm (i.d.) were used, and the sampling depth was about 150 mm for all core sizes. A previous study showed that the 150-mm sampling depth was sufficient to catch almost all ^{137}Cs inventory (Nearing et al., 2005). The same sampling scheme in Fig. 2 was used to collect samples in February 2017. All samples from both locations were air dried and sieved through a 2-mm sieve for radionuclides analysis.

2.3. ^{137}Cs and unsupported ^{210}Pb measurement

Radionuclide ^{137}Cs was generated by atmospheric nuclear bomb testing, while ^{210}Pb is a naturally occurring radionuclide derived from the ^{238}U decay series. The atmospheric fallout of ^{210}Pb is referred to as unsupported ^{210}Pb or $^{210}Pb_{ex}$ to distinguish from the ^{226}Ra-supported ^{210}Pb produced in situ, and is calculated by subtracting the supported ^{210}Pb from the total ^{210}Pb activity measured in each sample. Around 250 g of sieved soil sample was packed into 170-ml polypropylene jar with an air-tight lid, and incubated for > 25 days to ensure equilibrium between ^{226}Ra and ^{222}Rn (Polyakov et al., 2017). Radioactivity was

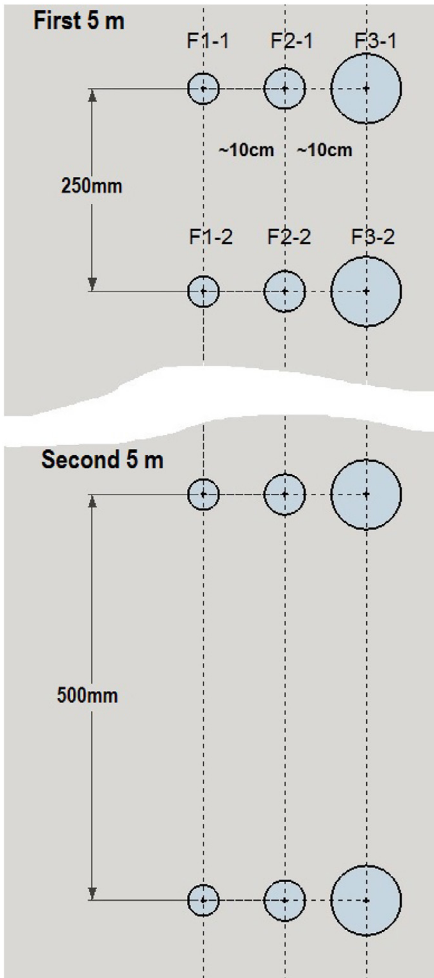

Fig. 2. Sampling scheme for three different core sizes on prairie grassland and forestland near El Reno, Oklahoma; and rangeland near Sonoita, Arizona.

measured at the Southwest Watershed Research Center using a gamma ray spectrometer with two high-purity germanium detectors coupled to a multichannel analyzer (BE5030, Canberra, USA). The relative efficiency of the measurement was > 30%. The system was calibrated using the mixed radionuclide reference material IAEA-447 (Shakhashiro et al., 2012) certified by the International Atomic Energy Agency. The gamma emission spectrum was obtained over 0–2 MeV range with resolution of 0.24 keV (8192 channels). Measurement and spectrum analysis was conducted using Genie-2000 Spectroscopy software (Canberra, 2009). The samples were counted for at least 160,000 s or until < 5% peak area uncertainty was achieved. The ^{137}Cs activity was measured at the 662-keV peak and the total ^{210}Pb activity at 46.5 keV. The supported ^{210}Pb activity was obtained by directly measuring the ^{214}Bi activity at 609.3 keV under the secular equilibrium.

2.4. Data analysis

Basic descriptive statistics including sample mean, sample standard deviation, standard error of the mean, variation of coefficient, and upper and lower 95% confidence limits of the mean were calculated for each core size under each land use. The 95% confidence limits for the mean were calculated with a two-tailed test assuming a normal distribution that is justified by the Central Limit Theorem and the large sample sizes of > 30 on each site. The 95% confidence limits for the sample standard deviation were estimated using a Chi-square distribution with a two-tailed test. Instead of directly testing for the equality of the two means for H_0: $\mu_1 = \mu_2$ (H_a: $\mu_1 \neq \mu_2$), a simplistic approach was used to test for a significant difference between the two means by comparing their 95% confidence intervals (CI). The difference in means was considered statistically significant at $\alpha = 0.05$ if their confidence intervals of the two means did not overlap. Otherwise, it was concluded that there was not sufficient evidence to reject H_0 (i.e., a significant difference was not detected). The difference in two sample standard deviations was inferred similarly by comparing the 95% CI. In addition, F-test was carried out to test the H_0: $\sigma_1^2 = \sigma_2^2$ (H_a: $\sigma_1^2 \neq \sigma_2^2$) with a two-tailed test between the two treatments at $\alpha = 0.05$. Sample number n that is required to yield an estimated mean with an allowable relative error (RE) for the mean at the 95% confidence level ($\alpha/2 = 0.025$) was estimated with:

$$n = \left(\frac{1.96\,CV}{RE}\right)^2 \tag{1}$$

where CV is the coefficient of variation in percent; and RE is the percent relative error of the estimated mean relative to the population mean.

In general, most statistical tests for H_0 only control the Type I error, denoted by α, which is the probability of falsely rejecting H_0 when it is actually true. The Type II error, denoted by β, is not generally considered. The Type II error represents the probability of falsely accepting H_0 when it is actually false. In addition to the Type I error, the Type II error is also useful in correctly determining whether or not to reject or accept H_0. The power of the test is reflected by 1-β, which is the probability of correctly rejecting H_0 when it is actually false. However, the β test requires the knowledge of the population mean that is generally unknown. Thus, to conduct a β test a particular mean value or several values as well as an α value must be specified (Ott, 1988, pp. 143). In this study, the 95% confidence limits were chosen for each dataset for a two-tailed β test at $\alpha = 0.05$ (note the upper and lower 95% confidence limits yield an identical β due to the symmetry of the normal distribution). In addition, $\mu \pm 4$SE (standard error) with $\alpha = 0.05$ were used in two-tailed tests to generate additional information on β for cases in which the confidence intervals of the two means began to overlap.

Under the assumption of isotropic conditions, the empirical semi-variance (γ) can be calculated as:

$$\gamma(h) = \frac{1}{2\,n(h)} \sum_{i}^{n(h)} [z(x_i + h) - z(x_i)]^2 \tag{2}$$

where h is the separation distance; $n(h)$ is the number of paired data at a separation distance of h; and Z is the radioisotope inventory at location x_i or $(x_i + h)$. Empirical semivariograms were obtained by plotting semi-variances with separation distances of < 4 m.

3. Results and discussion

3.1. Spatial dependency of ^{137}Cs and unsupported ^{210}Pb

A single core size of 51 mm was used on the cropland site at the El Reno location. The semivariogram of ^{137}Cs with a 0.2-m separation increment, each lag with > 36 pairs of samples, is shown in Fig. 3 (note $^{210}Pb_{ex}$ was not measured on this site). Except for a zero value at the zero separation as is supposed to be the case for an empirical semivariogram, the semivariances at all other separation distances were comparably large, and close to the variance of the entire transect in the field. This semivariogram indicated that the distribution of ^{137}Cs inventory was nearly stationary in the transect and had little spatial dependence if the separation distance was > 0.2 m. The site was relatively flat with slope being < 1%, and was under conventional tillage for > 30 years. The main tillage tools included moldboard plow, chisel, and disk. The results indicated that the initial ^{137}Cs deposition was spatially random and the long-term tillage operation did not create any discernable spatial dependence in ^{137}Cs distribution at this small spatial scale. The results suggested that samples taken > 0.2 m apart were

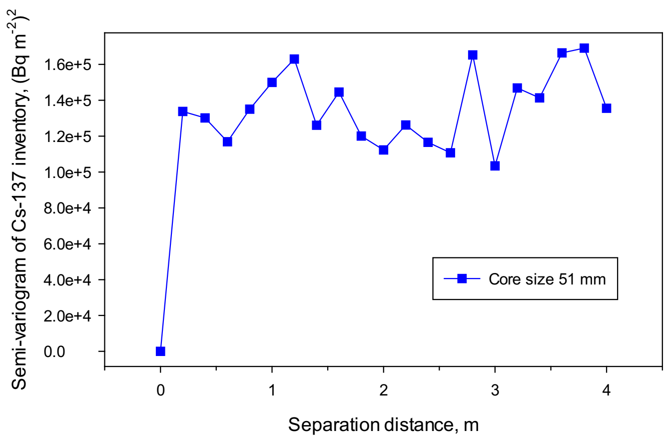

Fig. 3. Semivariogram of ^{137}Cs inventory for the core size of 51 mm on cultivated land near El Reno, Oklahoma.

largely independent, and could be composited to form a representative sample of the sampling location. The representativeness would be compromised if sampled within the 0.2-m radius.

Three core sizes were used on the grassland (tall prairie) site at El Reno. The semivariograms for each core size are shown in Fig. 4 for both ^{137}Cs and unsupported ^{210}Pb. The sizable fluctuations of semivariances at different separation distances might have been partially caused by the variable numbers of data pairs used in the computation, ranging from 10 to 29 depending on separation distances. For ^{137}Cs, semivariances at all separation distances of > 0.25 m except for zero were very large, and were near the ^{137}Cs variance of the field for the core sizes of 51 mm and 65 mm (Fig. 4A), confirming that ^{137}Cs spatial distribution was spatially random with little spatial dependence under the native tall prairie at scales of > 0.25 m. In contrast, spatial dependence was shown for the 86-mm core size up to a separation distance of 1.5 m. It is not clear at this point why spatial correlation relationships were influenced by different core sizes. The result of the 86-mm core size was questionable, because similar spatial patterns of ^{137}Cs distributions of different core sizes were expected given the sampling scheme of Fig. 2. Since the transects of different core sizes were only 10 cm apart, there were no reasons to believe that the patterns of the spatial dependency of ^{137}Cs should differ for different core sizes. This speculation was confirmed by the similarity in the spatial structures of ^{137}Cs for the 51-mm and 65-mm core sizes. It was also indirectly corroborated by the fact that the spatial dependency of $^{210}Pb_{ex}$ was similar for the three core sizes on the same site (Fig. 4B), where little spatial dependency was exhibited if the separation distance was > 0.75 m. If we assume the semi-variogram of the 86-mm core size is acceptable, the correlation distance would be 1.5 m, indicating samples taken > 1.5 m apart would be largely independent. Overall, similar semivariograms for both $^{210}Pb_{ex}$ and ^{137}Cs were shown for the grassland site, signifying little spatial dependence for both radioisotopes. It should be mentioned that the variance of $^{210}Pb_{ex}$ was much greater than that of ^{137}Cs, probably due to much larger measurement error for $^{210}Pb_{ex}$ than for ^{137}Cs. A similar type of semivariograms resembling those of the cropland and grassland sites were also obtained on the forest site at El Reno for both $^{210}Pb_{ex}$ and ^{137}Cs (Fig. 5), again showing that the two fallout radionuclides are randomly distributed in space and little spatial dependence exists at small spatial scales. These results suggest that samples taken > 0.75 m apart are essentially independent, and can be combined to yield a representative composite sample for the sampling location.

Both $^{210}Pb_{ex}$ and ^{137}Cs were measured for the samples from the Empire Ranch, Arizona. The activity levels of both radioisotopes were

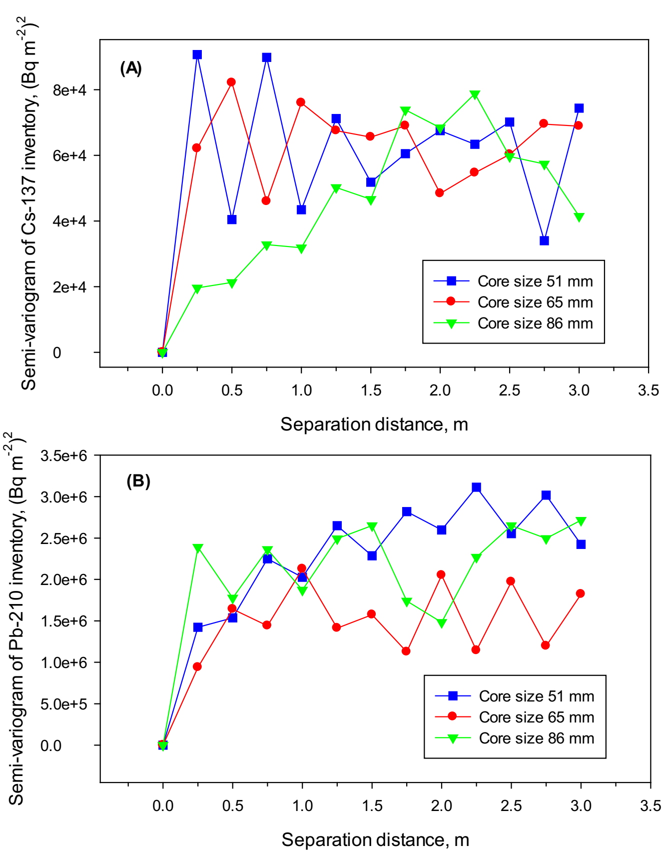

Fig. 4. Semivariograms of ^{137}Cs inventory (A) and unsupported ^{210}Pb inventory (B) for three different core sizes on native prairie grassland near El Reno, Oklahoma.

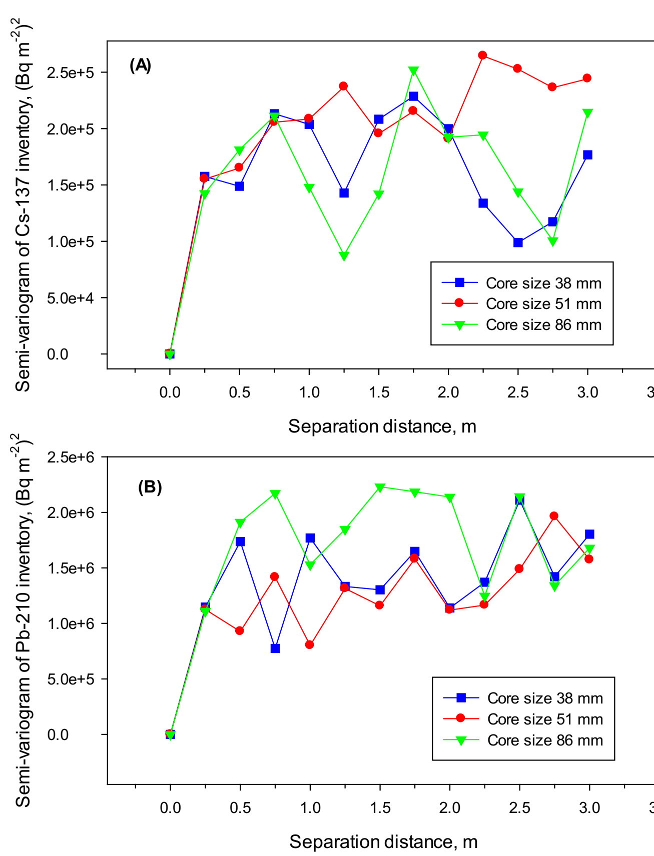

Fig. 5. Semivariograms of ^{137}Cs inventory (A) and unsupported ^{210}Pb inventory (B) for three different core sizes on forestland near El Reno, Oklahoma.

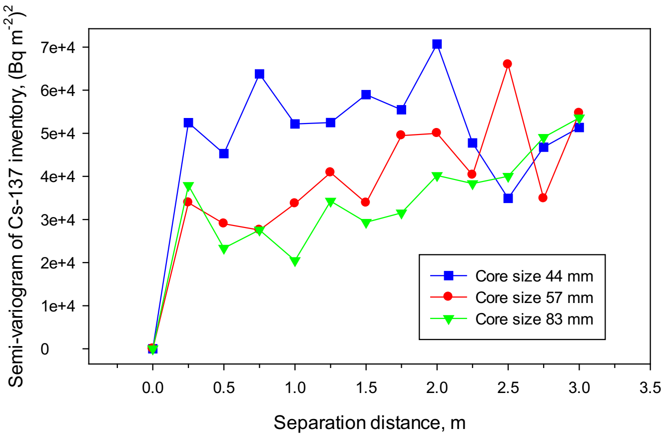

Fig. 6. Semivariograms of ^{137}Cs inventory for three different core sizes on rangeland in a semiarid region near Sonoita, Arizona.

much lower on this site than at El Reno, Oklahoma. The measured activity of $^{210}Pb_{ex}$ was zero for 68% of samples on this site. It was meaningless to characterize spatial correlation with the $^{210}Pb_{ex}$ dataset. Thus, only the semivariograms of ^{137}Cs are shown in Fig. 6 for the three core sizes. The semivariograms exhibited similar patterns to those at El Reno. That is, the semivariances were close to the ^{137}Cs variance of the field at all separation distances except for zero, showing that spatial distribution of ^{137}Cs in the native rangeland in the semiarid region was random with little spatial correlation at spatial scales of > 0.25 m.

To collect spatially independent samples, the spacing between samples must be greater than the semivariogram range or the autocorrelation distance. The development of a reliable semivariogram requires the use of grids with two or more scales in a nested design (Pennock and Appleby, 2010). Considerable resources are needed to fully evaluate the semivariogram range, and consequently sufficient studies have not been conducted to permit a reliable synthesis (Pennock and Appleby, 2010). Grid sizes commonly used in the literature are 10 m to 30 m, with only a few being < 10 m. Based on the limited data, Pennock and Appleby (2010) suggested sampling distances be at least 10 m to ensure the independence between samples. This study complemented the published data and showed that fallout radionuclide distribution was random with little or no spatial dependence at small spatial scales of < 10 m under different land uses and climates. A sampling spacing of > 0.75 m would be sufficient in most cases to yield largely independent samples, which can be combined to cut down measurement cost and time while improving the representativeness of samples by smoothing out profound spatial variation of fallout radionuclides at small scales.

3.2. Estimated mean of radioisotope inventory

Since the samples collected in this study are largely independent, all the samples can be lumped together to characterize the basic statistics of the probabilistic distribution of the fallout radionuclide inventory. Given the relatively large sample number on each site, reliable means and standard deviations may be estimated with confidence (Table 2). Based on whether the 95% confidence intervals are overlapped or not, significant differences between the means can be inferred. To assist the inference of the α test, a β test was also conducted for α = 0.05 for the 95% confidence limits of each core size and land use, and a β value of approximate 0.5 was obtained for all 95% confidence limits. Similarly, an approximate β value of 0.02 was found for the $\mu \pm 4SE$ of all core sizes and land uses. Generally, the corer size, ranging from 38 mm to 86 mm, had no significant effect on the mean estimates within each land use at the El Reno location at α = 0.05. That is, there was no sufficient evidence to reject the null hypothesis that the estimated means are equal, but we also could not accept the null hypothesis given β being > 0.5 (note that the absolute difference of the two means is generally < 1.96SE). However, a significant difference between the 44.5-mm and 82.6-mm core sizes was detected for the Empire Ranch at α = 0.05, with the power of test (1-β) equal to 0.995. For further confirmation, the samples from the 57.2-mm and 82.6-mm core sizes were combined and tested against those of the 44.5-mm core size on the site, and a significant difference was detected at α = 0.05 with (1-β) of > 0.991. The significant difference was caused largely by sampling errors. For gravely soils, it was harder to control sampling area with a smaller auger due to impediment by large rocks. A smaller auger that is more likely to be impeded by rocks had greater tendency to enlarge boreholes when being rotated, resulting in overestimation of the ^{137}Cs inventory. In addition, rocky soils have greater internal sample heterogeneity due to rock content variation as indicated by the larger CV.

Since corer sizes had no significant or apparent effect within each land use at El Reno, a lumped normal test with the increased sample number of 93 between the tall prairie and forest sites was conducted (data not shown). The results showed that the estimated overall mean of ^{137}Cs on the forest site was 14% greater than that on the prairie site at α = 0.05 with the power (1-β) being > 0.952. The slightly higher ^{137}Cs inventory on the forest site than on the prairie site may be explained as follows. First, most ^{137}Cs fallout was deposited in the form of wet fallout, but dry fallout also contributed. The dry fallout might have been disproportionally caught in the woodlands that served as windbreak on the site. Second, removal of ^{137}Cs on the prairie site by grazing and on the cultivated sites due to grain harvest would reduce the ^{137}Cs inventories compared with those on the forest site, where ^{137}Cs taken up by plants and trees was not removed but recycled via organic matter decomposition. In contrast, there was not sufficient evidence to reject the H_0 that the $^{210}Pb_{ex}$ inventories between the prairie and forest sites are equal at α = 0.05, or to accept it with β = 0.91 when tested with the lumped dataset (n = 93). The difference between the two radionuclides might have something to do with their deposition patterns and forms. Fallout $^{210}Pb_{ex}$ is being deposited continuously; whereas ^{137}Cs was mainly deposited during the 1950's and 1960's. Similar to ^{137}Cs, corer sizes didn't have significant or apparent effect on the $^{210}Pb_{ex}$ inventory within each land use at El Reno (Table 3).

3.3. Estimated variability of radioisotope inventory

Total variability of measured ^{137}Cs inventories on a site experiencing neither erosion nor deposition is generally composed of (1) random spatial variability due to small scale variations in soil, vegetation, and micro-topography, (2) sampling errors, and (3) ^{137}Cs measurement errors (Owens and Walling, 1996; Zapata, 2010). Both sampling errors and measurement errors include systematic and random errors such as measuring the cross-section of corer (systematic), photon counting (random), and sample weighing (random) (Lettner et al., 2000). Systematic errors or bias can be removed by careful calibration or bias correction. Random errors can only be minimized by increasing sample number. The sampling and measuring errors are inherently included in the measured ^{137}Cs inventories, which are generally < 10% each (Sutherland, 1991; Owens and Walling, 1996). Random spatial variability is the prevalent source of ^{137}Cs variability. Lettner et al. (2000) analyzed the sources of ^{137}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 random spatial variations of ^{137}Cs on both reference and measuring sites, and the spatial variabilities on both sites are the predominant contributors to overall uncertainty of soil erosion estimation (Zhang et al., 2015a). Given the predominant nature of spatial variability, the results of variability analysis in this work are mainly reflective of the spatial variability.

Variability is commonly expressed in variance, standard deviation, and/or CV. The sample standard deviations of the ^{137}Cs inventories

Table 2
Basic statistical properties of Cs-137 inventories in different land uses at two locations.

Location	El Reno, Oklahoma						Empire Ranch, Arizona			
Land use	Crop	Grassland			Forestland		Rangeland			
Corer diameter, mm	50.8	50.8	65.1	85.7	38.1	50.8	85.7	44.5	57.2	82.6
Mean (m), $Bq\,m^{-2}$	1501	1499	1437	1376	1580	1611	1729	641	499	479
95%U limit-m[a], $Bq\,m^{-2}$	1613	1587	1529	1446	1729	1766	1871	730	570	547
95%L limit-m[a], $Bq\,m^{-2}$	1388	1411	1345	1306	1431	1456	1587	552	428	412
Std deviation, $Bq\,m^{-2}$	408	250	262	198	422	440	404	253	202	192
95%U limit-SD[b], $Bq\,m^{-2}$	507	335	350	264	564	588	539	338	270	257
95%L limit-SD[b], $Bq\,m^{-2}$	342	200	209	158	337	352	323	202	161	154
Sample size	51	31	31	31	31	31	31	31	31	31
Std error, $Bq\,m^{-2}$	57	45	47	36	76	79	72	45	36	35
Coeff. of variation, %	27	17	18	14	27	27	23	40	40	40
Maximum, $Bq\,m^{-2}$	2680	1958	2128	1696	2292	2422	2658	1332	976	1221
Minimum, $Bq\,m^{-2}$	847	979	866	998	585	915	721	291	209	210
Estimated sample no.[c]	13	5	6	4	12	13	9	27	28	27

[a] Upper (U) and lower (L) 95% confidence limits for the mean (m), calculated using a normal distribution.
[b] Upper (U) and lower (L) 95% confidence limits for the standard deviation (SD), calculated using a χ^2 distribution.
[c] Sample number needed to have a relative error of 15% for the estimated mean at the 95% confidence level.

Table 3
Basic statistical properties of unsupported Pb-210 inventories in different land uses at El Reno, Oklahoma.

Land use	Grassland			Forestland		
Corer diameter, mm	50.8	65.1	85.7	38.1	50.8	85.7
Mean (m), $Bq\,m^{-2}$	2406	2582	2483	2237	2407	2572
95%U-m[a], $Bq\,m^{-2}$	2890	3068	2982	2685	2791	3073
95%L-m[a], $Bq\,m^{-2}$	1923	2095	1983	1789	2023	2071
Std deviation, $Bq\,m^{-2}$	1372	1382	1418	1272	1091	1423
95%U-SD[b], $Bq\,m^{-2}$	1835	1847	1895	1701	1458	1902
95%L-SD[b], $Bq\,m^{-2}$	1097	1104	1133	1017	872	1137
Std error, $Bq\,m^{-2}$	246	248	255	229	196	256
Sample size	31	31	31	31	31	31
Coeff. of variation, %	57	54	57	57	45	55
Maximum, $Bq\,m^{-2}$	5325	6077	5644	5448	4556	5676
Minimum, $Bq\,m^{-2}$	0	0	0	0	0	455
Estimated sample no.[c]	56	49	56	55	35	52

[a] Upper (U) and lower (L) 95% confidence limits for the mean (m), calculated using a normal distribution.
[b] Upper (U) and lower (L) 95% confidence limits for the standard deviation (SD), calculated using a χ^2 distribution.
[c] Sample number needed to have a relative error of 15% for the estimated mean at the 95% confidence level.

were not significantly affected by the corer sizes within each land use at both locations (Table 2), and the test results were further substantiated by F-tests at $\alpha = 0.05$. However, a Chi-square test using lumped data ($n = 93$) showed that the standard deviation of the ^{137}Cs inventories on the forest site was greater than that on the prairie site at $\alpha = 0.05$, which was also confirmed by F-test. The lower variability on the prairie site might resulted from the lack of tillage disturbance and the homogeneous grass cover (Fig. 1b) that causes minimal redistribution of rainfall. The larger standard deviation on the forest site was attributed to rainfall interception and redistribution by uneven cover of tree canopy (Fig. 1c) and stem flows. The F-tests showed that the standard deviation on the cultivated site was also significantly greater than that on the prairie site, although it was not different from that on the forest site. Given the level field on the cropland site, the larger spatial variability was caused primarily by tillage operations and uneven soil translocation. This result is in line with those reported in the literature. Sutherland (1994) reported that spatial variability across a gentle slope within the cultivated field was 55% greater than that within a nearby undisturbed field. Lance et al. (1986) reported that the mean variance of ^{137}Cs inventories averaged over 17 transects running along contour lines in a cultivated slope plot was $99.6\,Bq^2\,core^{-2}$; whereas that averaged over nine transects in an adjacent native tallgrass prairie plot was $34.8\,Bq^2\,core^{-2}$, showing 1.9 times increase by tillage operation and localized soil erosion. The CV on the cultivated site, similar to those on the forest site, was much larger than those on the prairie site. These results didn't support the assertion that "soil mixing by cultivation will undoubtedly reduce the spatial variability of ^{137}Cs compared with uncultivated sites" made by Loughran et al. (2010). Compared to the El Reno location, the CV on the Empire Ranch was much greater. The larger variability on the Empire Ranch was caused by large rock contents that required the use of the two-teeth auger. It is harder to control sampling areas with a rotating auger than with a non-rotating tube with a beveled smooth cutting bit. In addition, rocky soil is more internally heterogeneous at a micro scale because rock contents directly influence the ^{137}Cs inventory per unit area. For example, the presence of a large rock in a soil core will lower the ^{137}Cs inventory per unit area. Rocks also affect ^{137}Cs distributions by altering preferential flow redistributions both vertically and horizontally.

Similar to ^{137}Cs, F-tests showed that corer sizes had no significant effect on the standard deviation of $^{210}Pb_{ex}$ inventory (Table 3). Standard deviations of the $^{210}Pb_{ex}$ inventory at El Reno, compared with ^{137}Cs, were much larger, indicating that the measured $^{210}Pb_{ex}$ activity was more variable (Table 3). Since the sampling errors were the same for both radionuclides, the increased variability of $^{210}Pb_{ex}$ over ^{137}Cs might have been attributed to the measurement errors by gamma spectrometers. Radioactivity is derived from peak (histogram) area above background on a spectrum. ^{137}Cs is determined directly from a single peak, while $^{210}Pb_{ex}$ is determined as the difference between peak areas of total ^{210}Pb and ^{214}Bi, which surely yields greater variance ($\approx$sum of the two variances of total ^{210}Pb and ^{214}Bi). The uncertainty of measuring $^{210}Pb_{ex}$ was > 10 times greater than that of measuring ^{137}Cs by the gamma spectrometer used in this study. The maximum inventories of $^{210}Pb_{ex}$ were generally $> 5000\,Bq\,m^{-2}$, while the minimums were zero (Table 3). The larger variability was also indicated by the CV values. The average CV for $^{210}Pb_{ex}$ was 54%, and it was only 22% for ^{137}Cs at the El Reno location. These results indicate that uncertainty of soil loss rates estimated with $^{210}Pb_{ex}$ would be much greater than that estimated with ^{137}Cs due to the larger measurement errors, suggesting $^{210}Pb_{ex}$ would be less reliable than ^{137}Cs in providing quantitative long-term average soil erosion rates unless its measurement accuracy is greatly improved.

3.4. Sample number

Sample number is determined by the significance level of α, sample CV (or variance), and relative error allowable for the objective of a particular study or simply for practical consideration such as work load and resource limitation, as shown in Eq. (1). Sample CV is the major determinant that is mainly determined by spatial variability as discussed earlier. At El Reno, average CV was 16, 26, and 27% for the prairie, forest, and cultivated sites. Soil erosion is believed to be negligible on these sites due to the relatively flat terrains. If they are used as reference sites, approximately 10, 26, and 28 of independent samples are needed to estimate the ^{137}Cs reference inventory with an allowable relative error of 10% at the 95% confidence level for the prairie, forest, and cultivated sites, respectively. As recommended in the literature (Zapata, 2010), flat grassland is the ideal reference site. More samples are required if forest and cultivated lands have to be used as reference sites. A stringent 10% relative error that is chosen for estimating the reference inventory is because it is the most important parameter for soil erosion estimation (Walling and He, 1999; Li et al., 2010; Zhang et al., 2015a, 2015b). The accuracy of a reference inventory directly affects the accuracy of estimated soil erosion or deposition rates at every sampling location in a study area. In contrast, the accuracy of the ^{137}Cs inventory estimate at a particular location merely affects the soil erosion rate estimated for that location. Thus, for effective and efficient application of the ^{137}Cs method, a higher standard for ^{137}Cs inventory estimation is desirable and should be employed for reference sites than measuring sites or points. These results are in line with those reported in the literature on estimation of the reference inventory. It has been reported that a 20% CV of ^{137}Cs inventory was typical on the reference sites (Sutherland, 1996; Bernard et al., 1998; Basher, 2000; Fornes et al., 2005; Mabit et al., 2009). For a 20% CV, approximately 15 samples are needed to quantify reference inventory with an allowable error of 10% at the 95% confidence level. Considering both α and the power (1-β), Kirchner (2013) recommended that 15–20 samples should be used for estimating reference inventory. This is somewhat consistent with the recommendation of Pennock and Appleby (2010) who suggested a minimum of 11 samples be used to estimate the ^{137}Cs inventory on the reference site. Actually, required sample number depends on many factors such as land use type, soil properties, and sampling methods. For gravely soils such as the one on the Empire Ranch, about 61 samples are required to control the relative errors to 10% at the 95% confidence. Compared with ^{137}Cs, a greater sample number is needed to estimate the reference inventory for $^{210}Pb_{ex}$ due to larger measurement errors. Even on the prairie site, about 120 samples are required to estimate the $^{210}Pb_{ex}$ reference inventory with an allowable relative error of 10% at the 95% confidence level. Such a large sample number is infeasible, and it is worth testing whether increasing measurement time or measuring the same sample repeatedly could decrease uncertainty and cut down the required sample numbers for $^{210}Pb_{ex}$ estimation.

Kirchner (2013) established relationships between minimum detectible erosion rates and sample sizes for various CV values, Type I error α, and power (1-β) using a simplified ^{137}Cs erosion conversion model of Zhang et al. (1990). Assuming an equal variance on both reference and measuring sites, a minimum detectible erosion rate of 12.5 $t\,ha^{-1}y^{-1}$ was derived for CV = 20%, $\alpha = 0.10$, power (1-β) = 0.90, and $n_1 = n_2 = 20$ (c.f. Fig. 3 of Kirchner (2013)). The same detection limit could also be achieved for the same conditions if $n_1 = 30$ from the reference site and $n_2 = 15$ from the measuring site were used instead. The method of Zhang et al. (1990) tends to overestimate soil erosion by 15% because it assumes that soil erosion begins in 1963 as opposed to 1954 as widely accepted for the ^{137}Cs technique (Zhang, 2014). The detection limit would be 10.6 $t\,ha^{-1}y^{-1}$ after discounting the 15% overestimation. Given the tolerable soil erosion rate (called T value) of some 10 $t\,ha^{-1}y^{-1}$ as recommended in U.S., an equivalent sample size of 20 for both sites would be sufficient to detect soil erosion rates, which are above the tolerable erosion level and cause soil degradation. If soil cores are taken on a landform unit basis, the same number of samples as on the reference site such as 20 are recommended for the sake of statistical test efficiency. If soil samples are taken in a transect or grid in a study area, it would be infeasible to take as many as 20 individual samples on each location, resulting 2000 samples for 100 grid points. For practical consideration, sample sizes on the measuring site may be reduced and samples may be combined. For example, if 30 samples are taken on a reference site, only 15 samples are needed on each sampling location or point to achieve the same erosion detection limit of the T value. In addition, combining independent samples for each location considerably reduces ^{137}Cs-measurement time and cost, albeit measuring individual samples is statistically preferred. Compositing independent samples can reduce estimation error of the means as well as random sampling error; whereas random ^{137}Cs measurement error would not be reduced, unlike the case if each sample is measured individually.

Generally, spatial variation of ^{137}Cs inventory in measuring areas is greater than that on a reference site (Sutherland, 1994; Lance et al., 1986) due to localized soil erosion such as random rill formation as well as uneven soil translocation by tillage operations. Therefore, more samples are normally needed to estimate mean inventory for a location in measuring areas to achieve the same accuracy as for the reference inventory estimation, especially given the profound spatial variability at small scales as is shown in this work. However, for practical consideration the relative errors for mean ^{137}Cs estimation in measuring areas may be relaxed to 15% due to the decreased importance compared to the reference inventory parameter as demonstrated above. The sample numbers required to have an allowable relative error of 15% at the 95% confidence are shown in Table 2. Approximately 5, 11, and 13 samples are needed for the prairie, forest, and cultivated sites at El Reno, and 27 for the stony rangeland on the Empire Ranch despite the corer sizes. It is strongly recommended that 5 to 15 independent samples should be combined to form a representative composite sample for a sampling location or grid point for most soils and land uses. For the case presented in the last paragraph, combining 15 independent samples is sufficient to yield a detection limit that is near the tolerable erosion rate of 10 $t\,ha^{-1}y^{-1}$ for CV = 20%, $\alpha = 0.10$, power (1-β) = 0.90, and $n = 30$ on reference sites. Because spatial variability of ^{137}Cs within a very short distance (c.f. < 3 m) is profound and independent, multiple cores should be taken and composited to obtain a better estimate of the ^{137}Cs inventory for the location. It is worth noting that composited samples were used in several studies to improve soil erosion estimation (Sutherland, 1994; Owens and Walling, 1996; Ritchie et al., 2009; Porto et al., 2009; Liu et al., 2017; Zhang, 2017a). Unfortunately, this good sampling practice has not been widely adopted and used in most studies published in the literature, partially because correlation distances have not been expressly characterized at small scales to provide the scientific support to the sampling scheme.

4. Conclusions

Owing to profound and random spatial variation of the fallout radionuclides within a short distance (c.f. > 0.75 m), a quantitative soil erosion rate cannot be estimated for a single core sample. The spatial variation of the ^{137}Cs inventory at a measuring location or point is not only caused by localized erosion but also by other random factors as is observed on the reference sites. The use of a single core sample assumes that any variation in ^{137}Cs inventory is caused by soil erosion, which is not the case due to the presence of random variation. The appropriate statistical test for the case with a single soil core from a measuring site and multiple samples from a reference site (case A in Kirchner, 2013) can only correctly classify the location as an erosion or deposition point in a qualitative sense with certainty, because any inventory change caused by random spatial variation at the point is mistakenly taken as caused by soil erosion and thus the corresponding soil loss rate estimated at the point is not the true erosion rate. The 'true' soil redistribution rate can only be quantitatively estimated using the 'true' mean inventory, free of random spatial variation, for the location or the representative area, provided that approximation errors from other assumptions and ^{137}Cs conversion models are acceptable (Zhang, 2014).

For quantitative soil erosion estimation, sufficient independent samples from a particular location or landform unit are required to obtain a reliable estimate of the mean ^{137}Cs inventory. With the true ^{137}Cs inventory, the true soil erosion rate for the location or area can then be estimated using the ^{137}Cs conversion models with confidence. Moreover, given the non-linear nature of the ^{137}Cs erosion models, the true ^{137}Cs inventory is preferred to be used in the models to minimize the estimation error of soil erosion rates, as opposed to a post processing by averaging the estimated soil losses of multiple soil cores. In addition, the new technology of using in-situ gamma detectors to map ^{137}Cs inventory at a high spatial resolution has potential to improve ^{137}Cs inventory estimation and subsequently soil erosion estimation. Given the high spatial variability of ^{137}Cs within a short distance, a relatively large view field or scan area would substantially reduce or average out spatial variability of ^{137}Cs distribution and yield a more representative estimate of the ^{137}Cs inventory for the measuring spot if the measurement accuracy of the in-situ detector can be substantially improved or properly calibrated to overcome the adverse effects of soil bulk density, water content, ^{137}Cs depth distribution, and position-sensitive detecting efficiency on the measurement accuracy.

In this study, spatial structures of the fallout radionuclides ^{137}Cs and $^{210}Pb_{ex}$ at small spatial scales were investigated under different land uses at two locations. The semivariograms showed that the distributions of ^{137}Cs and $^{210}Pb_{ex}$ were nearly stationary and had little spatial dependence at scales between 0.75 and 5 m in the cropland, grassland, forestland, and rangeland. The spatial correlation distances ranged from 0.2 to 0.75 m for most core sizes and land uses. The semivariances at the separation distances of > 0.75 m were very close to the variances of the fields for most cases. The overall results suggested that a sample spacing of > 0.75 m would be sufficient in most cases to yield largely independent samples, which could be composited to form a representative sample for the sampling location or measured individually if neither long measurement time nor high cost is an issue.

Spatial variations of the ^{137}Cs and $^{210}Pb_{ex}$ inventories were profound at small spatial scales of < 5 m. Given the large spatial variability at such a small scale, representative soil erosion rates cannot be estimated using ^{137}Cs and $^{210}Pb_{ex}$ inventories measured with a single soil core, because remarkably different soil erosion rates can be estimated for different soil cores taken < 1 m apart. Thus, reliable soil erosion rates can only be measured with multiple cores or composite samples taken at a sampling spacing of > 0.75 m.

Core size variation between 38 mm and 86 mm has no *apparent* effects on estimating mean and standard deviation of ^{137}Cs and $^{210}Pb_{ex}$ inventories in most soils except gravely soils. That is, given the sample size, there is no sufficient evidence to reject or accept the null hypothesis that the inventories measured with core sizes between 38 mm and 86 mm are equal under each land use. However, for gravely soils, larger rotating corers, which tend to minimize rock impediment, are preferred. A tube sampler with a beveled smooth cutting edge seems to be better than a two-tooth auger for taking soil samples, because the former has a better control of sampling area, and is capable of taking the entire sampling depth in a single core, eliminating the possibility of fall in from the borehole wall.

Except for gravely soils, 15–30 independent samples are generally needed to estimate ^{137}Cs reference inventory on reference sites, depending on land uses and type of corers. Greater sample sizes are required for forest and cultivated sites than for uncultivated grassland sites. More sample number is needed for a two-tooth auger than for a beveled smooth cutting edge corer. On measuring sites, if samples are stratified by uniformly eroded landform unit, the same sample size as on the reference site are recommended to maximize erosion detection sensitivity. If samples are taken in a transect or grid, it is strongly recommended that 5–15 samples be taken and measured individually (statistically preferred) if feasible, or as a combined sample if restricted by resource available, for a reliable estimate of ^{137}Cs inventory for a grid point for most soils and land uses. The general recommendations of sample sizes on both reference and measuring sites are expected to be applicable to most geographical regions because spatial structures of ^{137}Cs inventory at small scales are predominantly affected by land uses rather than fallout amounts.

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