Multivariate geostatistical analysis of fallout radionuclides activity measured by in-situ gamma-ray spectrometry
Case study: Loessial paired sub-catchments in northeast Iran

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A B S T R A C T

Cs-137 and other fallout radionuclides (FRNs) have been used worldwide for more than four decades as powerful techniques to assess the magnitude and spatial pattern of soil redistribution. Cs-137, 210Pbex, and 7Be have successfully been used as tracers of soil redistribution. Their coincident use can frequently provide worthwhile information for different time scales of soil redistribution. These radionuclides can either be measured in the laboratory or in-situ. In the present study in-situ measurements using a portable HPGe detector were carried out in two loessial paired sub-catchments with fairly similar characteristics in northeast Iran. Spatial sampling design based on a minimax approach was used to determine that 60 sites were sufficient for both areas of interest. Geostatistical analysis and the linear model of co-regionalization (LMC) were applied in this study for radionuclides. The spherical model for Sample and Testifier sub-catchments with ranges of 750 m and 500 m respectively was selected as the most suitable model. The highest and the lowest non-captured variability belonged to 210Pbex and 7Be for both studied areas. The patterns of spatial variation of 7Be and Cs-137 for Sample and the patterns of spatial variation of 7Be and 210Pbex for Testifier sub-catchment were very similar. For Cs-137, global uncertainty for both sub-catchments was nearly the same, but different for other radionuclides. The distribution of local uncertainty for all radionuclides in the Testifier sub-catchment was the same. The means of spatial distance between the grid cells with the highest and the lowest uncertainty for 7Be, Cs-137, and 210Pbex are 0.4, 1, and 7 km, respectively.

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1. Introduction

Cs-137, excess or unsupported lead-210 (210Pbex), and beryllium-7 (7Be) are non-exchangeable radionuclides. They have been used world-wide for more than four decades, and their use has proved to be a powerful technique to assess the magnitude and spatial pattern of soil erosion and sedimentation (Ritchie and Ritchie, 2008). They are rapidly and strongly fixed by the surface soil or small sediment particles. For these reasons, they have successfully been used as tracers of soil redistribution (Wallbrink and Murray, 1993; Walling and He, 1999; Matisoff et al., 2002; Zapata, 2002; Walling et al., 2003; Zhang et al., 2003; Mabit et al., 2008; An et al., 2014).

Due to different history and half-life of these radionuclides (Zapata, 2002), their simultaneous use can frequently provide worthwhile information for different temporal scales of soil erosion and sedimentation (Mabit et al., 2008).

Not only soil redistribution data over different temporal scales can be obtained using a single soil sampling, thereby avoiding time consuming and costly installations (Mabit et al., 2008), but
Fig. 1. Location of study area (paired sub-catchments) in northeast of Golestan Province, Iran.
also, the resulting FRNs measurements can provide appropriate spatial data. These data integrate the effects of all processes leading to soil redistribution that frequently cannot be achieved using conventional soil erosion and sedimentation models (Mabit et al., 2008).

Radionuclides can either be measured in the laboratory and in-situ. The advantages and disadvantages have already been discussed in detail (He and Walling, 2000; Mabit et al., 2008). Field measurements using a high purity germanium (HPGe) detector have shown to provide spatially representative estimates of environmental radioactivity across a range of landscapes (Tyler et al., 1996). Thus, it was used in this research. This measurement technique for Cs-137, 9Be, and 210Pbex has been previously employed (Blake et al., 1999; Tyler and Coppipolestone, 2007). Apart from agricultural practices, overland flow (van der Perk et al., 2002) and changes in altitude (Porto et al., 2009) are the main drivers of the redistribution of Cs-137, 9Be, and 210Pbex. Erosion investigations using Cs-137, however, often assume that Cs-137 is relatively homogeneously distributed from field to a small catchment scale (Walling and Quine, 1991; Wu and Tiessen, 2002; Schuller et al., 2004; Heckrath et al., 2005), and for a small catchment, a homogeneous rainfall pattern can be assumed as well (Schaub et al., 2010). Meanwhile, as 210Pbex and Cs-137 both behave similarly and are absorbed strongly by soil clays, their spatial patterns are similar (Zhang et al., 2006). This particular behaviour of the atmospherically derived radionuclides allows the prediction of soil redistribution rates under water erosion (Stefano et al., 2005; Mabit and Bernard, 2007; Mabit et al., 2008).

Based on published results, only a few authors have used geostatistics to characterize the spatial variability of soil redistribution from Cs-137 data (Chappell, 1998; van der Perk et al., 2002; Chappell and Warren, 2003; Mabit and Bernard, 2007; Navas et al., 2011). Variogram models are suitable tools to model the spatial distribution of variables and if they are done with proper care, kriging provides the best possible prediction from data (Goovaerts, 1997; Gong et al., 2014). Their associated uncertainty is of great importance (Goovaerts, 2001), however rarely assessed.

Multivariate geostatistical methods (co-simulation and co-kriging) have been used to analyse the spatial co-variation of soil properties (Goovaerts, 1997), but to the best of our knowledge, no published papers have paid attention to multivariate geostatistical methods and uncertainty in radionuclide measurements to capture soil redistribution. So, in this study radionuclides activity were taken into consideration.

In the case where spatially distributed information is needed, local and global uncertainties of maps should be known in advance. One of the common techniques that can be used for this is geostatistical stochastic simulation (Goovaerts, 1997; Deutsch and Journel, 1998). Geostatistical stochastic simulation is also designed to overcome the smoothing effect of the kriging estimator (Deutsch and Journel, 1998), especially when extreme spatial discontinuities have to be mapped or when interpolation errors need to be reproduced through a non-linear model.

As aforementioned, in the case of radionuclides in soil redistribution, local and global uncertainties are ignored, and the impact of agricultural activities on soil redistribution should be assessed in large scales of sub-basin or micro-watershed. To address this issue, loessial paired sub-catchments with fairly similar characteristics in northeast Iran was selected to reach the following objectives: 1) to use geostatistical stochastic simulation of radionuclide activities and assess their local and global uncertainties, 2) to upscale the area studied of 9Be from plot and field to micro watershed scale and sub-basin, 3) to use other radionuclides as co-variables to reduce prediction error (co-simulate radionuclides activities on different time scales).

2. Materials and methods

2.1. Site description

The studied area is located in the northeast of Golestan Province, Iran. The study includes paired sub-catchments (Sample “enclosed form 1999” and Testifier “an open area”) of representative watershed. They include intensively exploited arable land with commonly cultivated crops such as sunflower, wheat, watermelon, artificial and natural forest. Types of water erosion such as splash, sheet, and stream erosion affect them as well in the same way. The dominant soil texture in this area is silty loam, and soil depth varies from shallow to deep. Range of some soil characteristics in studied area including: soil organic carbon, C/N, bulk density and MWD are (0.75–4.4%), (9.5–12.7%), (1.5–1.6 g cm$^{-3}$) and (0.5–1.6 mm) respectively. The climate is typically semi-arid with a mean annual temperature of about 16.7 °C and annual precipitation of 482 mm (Hematzadeh et al., 2009). The landscape is gently undulating with an average altitude of approximately 800 m above sea level. Some characteristics of the area are given in Table 1. The areas of interest are depicted in Fig. 1.

2.2. Studied radionuclides

Caesium-137 (Cs-137) is an anthropogenic radionuclide (half-life = 30.2 years) most widely used in soil science and has been employed under different agro-environmental conditions around the world (Walling et al., 1986, 1995; Zapata, 2002; Mabit et al., 2007; An et al., 2014). It has been used in soil erosion investigations over a range of different scales, extending from experimental plots to one of the largest scale investigations in Quebec, Canada (217 km$^2$) (Mabit and Bernard, 2007; Mabit et al., 2008). Cs-137 is useful for providing estimates of medium term mean annual soil redistribution rates (i.e. from the beginning of global fallout in the mid-1950s until the time of sampling). Using this radionuclide, retrospective information on soil redistribution rates can be estimated from samples collected at the present time and all processes involving soil particle movements (water erosion, wind erosion, and tillage redistribution) can be integrated. Estimates are based on individual sampling points within the landscape; therefore spatially distributed information on rates and patterns of soil redistribution can be generated.

Lead-210 (210Pbex) is a natural geogenic radiototope (half-life = 22.3 years) that represents part of the uranium decay series. It originates from the decay of 226Ra found in most soils and rocks and produces short-lived gaseous 222Rn (half-life = 3.8 days) as its offspring. Most of this 222Rn decays to 210Pb within the soil producing supported 210Pb that is essentially in equilibrium with the parent 226Ra, some of the 222Rn diffuses upwards into the atmosphere where it rapidly decays to 210Pb. This 210Pb is deposited as fallout, and since it is not in equilibrium with the parent 226Ra, it is commonly termed unsupported or excess 210Pb (210Pbex), as
distinguish it from the supported $^{210}\text{Pb}$ in the soil. In contrast to Cs-137 fallout, the fallout of $^{210}\text{Pb}_{ex}$ is essentially constant through time due to its natural origin (Zapata, 2002). The use of $^{210}\text{Pb}_{ex}$ measurements can provide a retrospective assessment of (100 years) rates of soil redistribution.

Beryllium-7 ($^7\text{Be}$) is a natural cosmogenic radionuclide produced in the upper atmosphere by cosmic ray spallation of nitrogen and oxygen. This radionuclide has a very short half-life (half-life = 53.3 days) in comparison to Cs-137 and $^{210}\text{Pb}_{ex}$ radionuclides. Thus, it offers potential for investigating soil redistribution rates resulted from individual storm events or short periods of heavy rainfall. It has been used since the late 1990s to estimate soil erosion and sedimentation processes associated with individual periods of heavy rain, mainly at scales ranging from plots of a few square meters (Wilson et al., 2003) to fields of a few hectares. $^7\text{Be}$ is generally rapidly fixed by the upper few millimetres of the soil and is rarely found at depths greater than 3 cm. Successful studies employing $^7\text{Be}$ have been reported from Australia, UK, USA, and Chile (Mabit et al., 2008). The potential to document the erosion rates associated with individual events and to investigate the effectiveness of different soil conservation practices represents the major advantage of $^7\text{Be}$ over the assessments of longer-term erosion rates provided by Cs-137 or $^{210}\text{Pb}$. In-situ measurements could provide an effective approach for obtaining the necessary information on $^7\text{Be}$ inventories from a study site without the need for time-consuming soil sampling and associated laboratory measurements.

2.3. Spatial sampling design based on minimax approach

The spatial configuration of the network of sampling sites has a substantial effect on soil erosion and sedimentation assessment.

<table>
<thead>
<tr>
<th>Sub-catchments</th>
<th>Variable</th>
<th>Min</th>
<th>Mean</th>
<th>Max</th>
<th>SD</th>
<th>Skewness</th>
<th>Kurtosis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>$^{210}\text{Pb}_{ex}$</td>
<td>160.1</td>
<td>381.6</td>
<td>872.3</td>
<td>157.2</td>
<td>0.91</td>
<td>0.34</td>
</tr>
<tr>
<td></td>
<td>$^{137}\text{Cs}$</td>
<td>16.8</td>
<td>67.2</td>
<td>119</td>
<td>23.0</td>
<td>0.11</td>
<td>-0.78</td>
</tr>
<tr>
<td></td>
<td>$^7\text{Be}$</td>
<td>7.9</td>
<td>27.8</td>
<td>49.9</td>
<td>10.5</td>
<td>0.06</td>
<td>-0.49</td>
</tr>
<tr>
<td>Testifier</td>
<td>$^{210}\text{Pb}_{ex}$</td>
<td>155.7</td>
<td>354</td>
<td>692.5</td>
<td>136.5</td>
<td>0.70</td>
<td>-0.019</td>
</tr>
<tr>
<td></td>
<td>$^{137}\text{Cs}$</td>
<td>14.9</td>
<td>70.4</td>
<td>131.4</td>
<td>22.0</td>
<td>0.39</td>
<td>0.46</td>
</tr>
<tr>
<td></td>
<td>$^7\text{Be}$</td>
<td>7.1</td>
<td>26.7</td>
<td>48.8</td>
<td>10.4</td>
<td>-0.13</td>
<td>-0.59</td>
</tr>
</tbody>
</table>

Table 3

Featurer correlation of studied radionuclides.

<table>
<thead>
<tr>
<th>Sub-catchments</th>
<th>$^{137}\text{Cs}$</th>
<th>$^7\text{Be}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>0.254</td>
<td>0.134</td>
</tr>
<tr>
<td>Testifier</td>
<td>1</td>
<td>0.144</td>
</tr>
</tbody>
</table>

Here, briefly mentioned, by adopting a model-based approach (Brus et al., 2002), the average kriging variance (AKV) was chosen as a design criterion. In fact, by minimizing the AKV of the area of interest, the optimal sampling design was determined in the case that no directly measured prior information of the primary variable of interest is available. However, AKV is dependent on some unknown parameters, and preliminary estimates of model parameters are not available. Main focus was made on a rich and flexible family for modelling the correlation structure (Banerjee et al., 2004). Thereafter, to overcome the misspecification of model parameters the minimax approach was used. In fact, the optimal design in the minimax sense is the design that minimizes the maximum value of the design criterion over the misspecification of model parameters. For this, a range of plausible values for the parameters were first assumed. Then, the optimal design was chosen when the parameters took the worst possible value within their respective ranges and thus the criterion was minimized. The least favourable parameter values are those that maximize the design criterion. For choosing the optimal design, the parameter space was discretized and then the Simulated Annealing (SA) was applied.

The method is illustrated keeping in mind the ancillary information (slope %) from sub-catchments (Sample and Testifier). A sequential-based method was used for optimizing the sampling networks. It was shown that 60 sites are sufficient for both areas of interest (Fig. 2) (Rivaz et al., 2014).

2.4. In-situ measurement of radionuclides activity and their analysis

In-situ measurements of radionuclide activity in soil are more sensitive and provide more representative data than the data...
obtained by soil sample collection and subsequent laboratory analysis. An unshielded detector placed about one meter above the ground detects gamma rays from an area within about a 10-m radius. This technique can be applied in a small fraction of the time required for laboratory analysis (Beck et al., 1972).

Table 4
Partial sill and nugget effect for studied radionuclides in areas of interest.

<table>
<thead>
<tr>
<th>Sub-catchments</th>
<th>Variable</th>
<th>Log ((210\text{Pb}_{\text{ex}}))</th>
<th>(137\text{Cs})</th>
<th>(^7\text{Be})</th>
<th>Log ((210\text{Pb}_{\text{ex}})-(137\text{Cs}))</th>
<th>Log ((210\text{Pb}_{\text{ex}})-(^7\text{Be}))</th>
<th>(137\text{Cs}-^7\text{Be})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>Nugget</td>
<td>0.829</td>
<td>0.790</td>
<td>0.558</td>
<td>0.164</td>
<td>-0.023</td>
<td>-0.223</td>
</tr>
<tr>
<td></td>
<td>Sill</td>
<td>0.245</td>
<td>0.353</td>
<td>0.623</td>
<td>0.149</td>
<td>0.255</td>
<td>0.452</td>
</tr>
<tr>
<td>Testifier</td>
<td>Variable</td>
<td>(210\text{Pb}_{\text{ex}})</td>
<td>(137\text{Cs})</td>
<td>(^7\text{Be})</td>
<td>(210\text{Pb}_{\text{ex}}-137\text{Cs})</td>
<td>(210\text{Pb}_{\text{ex}}-^7\text{Be})</td>
<td>(137\text{Cs}-^7\text{Be})</td>
</tr>
<tr>
<td></td>
<td>Nugget</td>
<td>1.111</td>
<td>0.968</td>
<td>0.365</td>
<td>0.107</td>
<td>-0.073</td>
<td>0.306</td>
</tr>
<tr>
<td></td>
<td>Sill</td>
<td>0.159</td>
<td>0.128</td>
<td>0.848</td>
<td>0.303</td>
<td>0.349</td>
<td>-0.007</td>
</tr>
</tbody>
</table>

Table 5
Some criteria of cross validation for studied radionuclides in areas of interest.

<table>
<thead>
<tr>
<th>Sub-Catchment</th>
<th>Radionuclides</th>
<th>RMSE (^a)</th>
<th>RSQR (^b)</th>
<th>Bias (^c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>Log ((210\text{Pb}_{\text{ex}}))</td>
<td>153.197</td>
<td>0.041</td>
<td>2.702</td>
</tr>
<tr>
<td></td>
<td>(137\text{Cs})</td>
<td>44.831</td>
<td>0.185</td>
<td>-39.390</td>
</tr>
<tr>
<td></td>
<td>(^7\text{Be})</td>
<td>10.061</td>
<td>0.113</td>
<td>-1.117</td>
</tr>
<tr>
<td>Testifier</td>
<td>(210\text{Pb}_{\text{ex}})</td>
<td>136.77</td>
<td>0.008</td>
<td>-0.0204</td>
</tr>
<tr>
<td></td>
<td>(137\text{Cs})</td>
<td>22.689</td>
<td>0.005</td>
<td>-0.0357</td>
</tr>
<tr>
<td></td>
<td>(^7\text{Be})</td>
<td>9.508</td>
<td>0.181</td>
<td>-0.0327</td>
</tr>
</tbody>
</table>

\(^a\) Root Mean Square Estimation.
\(^b\) R-Square.

Fig. 3. Fitted direct (diagonal) and cross semivariograms (off-diagonal) of log (\(210\text{Pb}_{\text{ex}}\), \(210\text{Pb}_{\text{ex}}\)-\(137\text{Cs}\), and \(^7\text{Be}\) (both sub-catchments).

Fig. 4. Cross validation for \(^7\text{Be}\) for Sample (A) and Testifier (B) Sub-catchments.
as a function of gamma-ray energy and angle of incidence. For field measurements, a portable HPGe detector was used to analyse Cs-137, $^{7}$Be, and $^{210}$Pb$_{ex}$ calibrated in the range 13–1600 keV. Efficiency calibration for in-situ gamma spectrometry is a complicated manner. One can find more detailed explanations in Beck et al. (1972).

2.5. Statistical analysis

The most common application of co-kriging is when the co-variable is cheaper to measure and has been more densely sampled than the target variable (Pebesma and Bivand, 2005). Here, however, we use the same set of sample locations where all variables (co-variables and target: all three radionuclides) have been measured (co-located measurements). A major aim of using co-kriging and co-simulation is to reduce the prediction variances. With so much new information (co-variables) and a fairly good model of co-regionalization, lower prediction errors can be expected. In co-simulation, uncertainties are taken into consideration as well.

All three studied radionuclides have complete space correlation (co-located measurements). The geostatistical package gstat (Pebesma, 2004) and the spatial data package sp (Pebesma and Bivand, 2005) of R environment were used for the analysis.

2.6. Multivariate geostatistical analysis

In practice, we are often interested in modelling more than one property and/or secondary data are often available as additional information for the modelling of a primary variable. When the studied variables are intrinsically correlated, the linear model of co-regionalization (LMC) can be used. LMC assumes all the studied variables are the result of the same independent processes, acting at different spatial scales. One feature of assuming linear model is that it allows the conditional negative semi-definite condition to be expressed in the form of workable criteria.

Multivariate geostatistical techniques must be applied to capitalize on the variable information. In order to perform multivariate geostatistical analysis (co-kriging or co-simulation), a crucial and frequent problem is to find a model of co-
regionalization matrix that fits adequately in a mathematical sense to the empirical cross-variogram matrix. Therefore, first the spatial structure of a co-variable and its covariance should be modelled with the target variable. It is an extension of the theory of a single regionalized variable used for ordinary kriging (Rossiter, 2007). We have to fit models to both the direct (target variable) and cross-variograms (co-variables) simultaneously, and these models must lead to a positive definite co-kriging system. The easiest way to ensure this is to fit a linear model of co-regionalization: all models (direct and cross) have the same shape and range, but may have different partial sills and nuggets (Rossiter, 2007). The modelling in fact treats all variables equally: they are all modelled (by direct and cross variograms) and jointly predicted.

The variograms for all three radionuclides were computed. Their structures and ranges appeared to be fairly similar. The cross-variogram in this case also had a similar structure to the direct variograms. Linear models of co-regionalization to a multivariate sample variogram were fitted for both areas of interest. Direct and cross variograms were fitted for co-kriging and co-simulation. The performance of co-kriging and co-simulation could be evaluated by using cross-validation. The cross-validation was performed for all 60 points with data for radionuclides in the subset. Since the same locations were used for the interpolation, each point is left out in turn, and the prediction at that point is made from the remaining 59 points. For almost all points in the subset, there is a measurement of the co-variables (2 radionuclides). This should give a decided advantage to co-kriging if the feature-space correlation is good and there is a good spatial cross-correlation.

3. Results

3.1. Descriptive statistic

Some statistical characteristics for the above-mentioned radionuclides are presented in Table 2. Their feature correlation coefficients for both study areas are presented in Table 3.

3.2. Linear model of co-regionalization (LMC)

Fitted direct and cross semivariograms, (log (210Pbex) transformed to get normal distribution), Cs-137, and 7Be, for co-kriging and co-simulation, for Sample and Testifier sub-catchments are illustrated in (Fig. 3A and B). For the Testifier sub-catchment, 210Pbex was used. Sample and Testifier sub-catchments have the spherical model, with ranges of 750 m and 500 m respectively that seem comparable to the range of the direct variograms of the target variables. Since LMC requires equal ranges, this is good. Thus, the fitted variograms have the same range but different sills and nugget effects for radionuclides of every sub-catchment. Their partial sills and nugget effects are given in Table 4. The highest and the lowest nugget

![Fig. 7. The global uncertainty for different radionuclides. Red columns: Sample sub-catchment.](image)
(non-captured variability) belong to $^{210}\text{Pb}_{\text{ex}}$ and $^{7}\text{Be}$ for both studied areas.

This ratio is also the highest for $^{7}\text{Be}$ in the other sub-catchment. In case of the cross semivariograms there is negative (cross) correlation between $^{7}\text{Be}$ with Cs-137 and $^{210}\text{Pb}_{\text{ex}}$ in the Sample sub-catchment and between $^{7}\text{Be}$ with $^{210}\text{Pb}_{\text{ex}}$ in the Testifier sub-catchment.

### 3.3. Cross validation

Cross validation, because of its simplicity, is a widespread and popular strategy to perform model selection. Some cross validation results for co-kriging and co-simulation are given in Table 5 and illustrated for $^{7}\text{Be}$ in Fig. 4.

### 3.4. Conditional Gaussian co-simulation

All three radionuclides activities were mapped for both sub-catchments using multivariate geostatistics. The maps of the mean of 1000 simulations (realizations) per grid cell are prepared for every radionuclide (Figs. 5 and 6). The patterns of spatial variation of $^{7}\text{Be}$ and Cs-137 for the Sample sub-catchment are very similar, but they are slightly different for $^{210}\text{Pb}_{\text{ex}}$.

For Testifier sub-catchment, the patterns of spatial variation of $^{7}\text{Be}$ and $^{210}\text{Pb}_{\text{ex}}$ are very similar, but the spatial variation of Cs-137 is completely different (Fig. 6). As shown in (Fig. 3B), direct and cross variograms fitted to Cs-137 are pure nugget effects. It means no spatial dependency for this variable in the Testifier sub-catchment could be captured.

Both multivariate geostatistical methods have the capability to show global uncertainty. It was assessed with co-simulation (Fig. 7). For Cs-137, the global uncertainty for both sub-catchments is nearly the same. In case of $^{210}\text{Pb}_{\text{ex}}$, the global uncertainty is completely different, and for the Sample sub-catchment, it is smaller and narrower than that for the other one. For $^{7}\text{Be}$, the highest global uncertainty for both areas is the same, but the distribution of global uncertainty for the Sample sub-catchment is wider than that for the other area.

Geostatistical stochastic simulation, in this study co-simulation, is the only method that considers local uncertainty. This uncertainty for radionuclides of the Sample and Testifier sub-catchments is illustrated in Figs. 8 and 9(A–C).

The distribution of local uncertainty for all radionuclides in the Sample sub-catchment is not exactly the same (Fig. 8C–A). However, conversely is acceptable for the Testifier sub-catchment (Fig. 9A–C).

The mean of the highest and the lowest local uncertainty for $^{7}\text{Be}$ is exactly the same.

The spatial distances between the grid cells with the highest and the lowest uncertainty for radionuclides for both areas are given in
In the Sample sub-catchment, the grid cell with the lowest uncertainty is the same for $^7$Be and $^{210}$Pb$_{ex}$, however, the grid cell with the highest uncertainty is the same for the other area for these two radionuclides. Means of spatial distance for $^7$Be, Cs-137, and $^{210}$Pb$_{ex}$ is 0.4, 1, and 7 km, respectively.

4. Discussion

In this study, multivariate geostatistical analysis was used to reduce prediction error by using other variables (two radionuclides) as co-variables and to assess the associated uncertainties. However there was poor correlation between all three studied radionuclides, but they are co-located measurements used in multivariate geostatistic. Findings related to non-captured variability showed that measurements of $^{210}$Pb$_{ex}$ need to be done by soil sampling due to self-absorption effects as stated by Mabit et al. (2008), not by HPGe portable measurement detector. However Al-Masri and Doubl (2013) showed not significant difference of $^{226}$Ra between lab and In-situ gamma spectrometry technique. The highest spatial dependency (nugget-sill ratio) in the Sample sub-catchment belongs to $^7$Be as well. So, apart from suitability of HPGe portable measurement, feasibility of $^7$Be also was determined for scales larger than farm size. These findings are not in accordance with (Wilson et al., 2003) but needs more researchs. In this case, rain gauges installation in different places of the study area is highly recommended.

Hence, $^7$Be can be used in micro-watersheds and sub-basins. So, using spatial sampling and geostatistical stochastic simulation of $^7$Be activity, its uncertainty can be measured and considered in assessing soil redistribution rate.

Herein soil redistribution rate by $^7$Be for individual events or short periods of heavy rainfall can be determined in micro-watersheds and sub-basins. Thereafter, different farming practices can be taken into consideration in these scales to reduce soil erosion as well.

By taking cross semivariogram into consideration, $^7$Be has a negative correlation with Cs-137 and $^{210}$Pb$_{ex}$ in the Sample sub-catchment and a negative correlation with $^{210}$Pb$_{ex}$ in the Testifier sub-catchment for short distances. Thus, some factors controlling redistribution of radionuclides are not the same for different time scales (few months, 40–50, and 100 years: in regard to half-life of radionuclides).

The patterns of spatial variation of $^7$Be and Cs-137 for the Sample sub-catchment are very similar, but they are a little bit

![Fig. 9. Local uncertainty of studied radionuclides (Testifier sub-catchment). Red and black columns: The highest and lowest local uncertainty.](image-url)
different for \(^{210}\text{Pb}_{\text{ex}}\). For the other sub-catchment, the patterns for \(^{7}\text{Be}\) and \(^{210}\text{Pb}_{\text{ex}}\) are very similar, but the spatial variation of Cs-137 is completely different.

The different spatial pattern of Cs-137 (no spatial dependency or pure nugget effect) and its completely random behaviour may have resulted from land use change and intensive agricultural practices for time periods of about 40–50 years. This is consistent with (An et al., 2014) that stated the different spatial pattern of Cs-137 is related to agricultural practices and topography. But it is unclear why findings of Mabit and Bernard (2007) and Molchanova et al. (2014), that this may be related to different sizes of study area (farm field and sub-catchment) and different factors (wind direction) controlling Cs-137 deposition. The average nugget effect appeared to be about 0.2 and 0.5 in the Sample and Testifier sub-catchments respectively. Therefore, using radionuclides as co-variables, significantly reduced the nugget sill ratio of target radionuclides (Table 4). This shows that determined multivariate geostatistics is highly fruitful in fallout radionuclides studies. Having nearly the same spatial pattern of \(^{8}\text{Be}\) and \(^{210}\text{Pb}_{\text{ex}}\), means that it is possible to use \(^{8}\text{Be}\) as a co-variable to predict \(^{210}\text{Pb}_{\text{ex}}\), but laboratory measurements would be needed to reduce the uncertainty of \(^{210}\text{Pb}_{\text{ex}}\). Thus, using multivariate geostatistical methods, other radionuclides can be used to reduce prediction errors and obtain more detailed knowledge about factors controlling the redistribution of \(^{210}\text{Pb}_{\text{ex}}\). Since all Cs-137 deposited in both areas and different farming and conservation practices have been done in the past ten years in both of them, global uncertainty for Cs-137 is nearly the same for both areas.

In the case of \(^{210}\text{Pb}_{\text{ex}}\), it is completely different, and for the Sample sub-catchment, it is smaller and narrower than that for the other one. This means that some factors cause uncertainties on the time scale around 100 years. For \(^{8}\text{Be}\), the highest global uncertainty for both areas is the same, but its distribution for the Sample sub-catchment is wider than that for the other one. It means that the different factors controlling the redistribution of \(^{8}\text{Be}\) in time scale less than 6 month are nearly the same for both sub-catchments, but for an explanation of different distributions, more research is needed.

The distribution of local uncertainty as well as the highest and the lowest uncertainty for grid cells for all radionuclides in the Sample sub-catchment are less similar than those in the Testifier sub-catchment. The mean of the highest and the lowest uncertainty are 0.4, 1, and 7 km, for \(^{8}\text{Be}\), Cs-137, and \(^{210}\text{Pb}_{\text{ex}}\) respectively for both sub-catchments. This means that the larger the time scale/span for radionuclides, the larger the spatial distance between the grid cells with the highest and the lowest uncertainty.

5. Conclusion

Concerning stochastic simulation of fallout radionuclides activity in loessial paired sub-catchments in northeast Iran, different results are obtained. In case of knowing soil redistribution rates, conversion models are needed that recommend to do in similar researches (however using portable HPGe, relaxation length should be considered). Anyway, the results show that poor correlation between above-mentioned radionuclides activity in both sub-catchments (0.134–0.254) and their patterns of spatial variation for the Sample sub-catchment are also nearly similar. For both sub-catchments, the highest and the lowest non-captured variability belong to \(^{210}\text{Pb}_{\text{ex}}\) and \(^{8}\text{Be}\), but the highest spatial dependency belongs to \(^{7}\text{Be}\). Hence, \(^{8}\text{Be}\) can be used in micro-watersheds and sub-basins and its uncertainty can be measured and considered in assessing soil redistribution rate. Since different farming and conservation practices have been done in the past ten years in both of sub-catchments, Global uncertainty for Cs-137 is nearly the same for both sub-catchments.

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