Journal of Spatial Hydrology

Volume 2 Article 5

2002

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J. of Spatial Hydrology
Vol.2 No.2

Spatial correlation between radon (^{222}Rn) in groundwater and bedrock uranium (^{238}U): GIS and geostatistical analyses

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ABSTRACT

This study describes approaches to create surface maps of radon in groundwater based on measurements of radon (^{222}Rn) in drilled bedrock wells at unevenly distributed sites and uranium bedrock maps from the South East of Sweden, the Östergotland county (N 58°14' – N 58°56' and E 14°53' – E 16°06'), see figure 1. Geostatistical techniques of inverse distance weighted (IDW), kriging and cokriging were compared in terms of their interpolation power and correlation between the produced radon in the water layer and the bedrock uranium layer. The goal of these analyses and calculations is to improve our understanding concerning the factors influencing the transport of radon. Therefore, these interpolation techniques were investigated by optimizing parameters that are used in the specific interpolation. Using the IDW interpolator method at fixed radius enabled us to determine the linkage or search distances for auto correlation, and linkage between radon in water and bedrock. This method showed good agreement with the cokriging method when using uranium concentration as a secondary variable. Good interpolation layers (with least root mean square errors RMSE=232) were obtained by kriging. However, the kriged radon surface showed poor correlation with bedrock uranium layers. The best radon in water layer that match with uranium in bedrock layer was produced using IDW interpolator (RMSE=377, using all points). The correlation coefficient (R^2) is 0.5 while for the kriging method the best correlation is R^2 = 0.1. A compromise between the two approaches is demonstrated.

Keywords: radon, uranium, groundwater, bedrock, GIS, Kriging, IDW

1. INTRODUCTION

Radon is a radioactive noble gas occurring naturally in the environment. It is produced by the decay of uranium in soil and rock. The release of radon from its source matrix into groundwater, as agreed by many investigators, e.g. Sun and Semkow 1998, is governed by the alpha recoil process and subsequent diffusion. The alpha recoil takes place during radon formation in the decay of its immediate parent (Ra-226). The recoil range is about 40 nm in soil & rock, about 95 nm in water and about 64,000 nm in air (Semkow 1990). Provided that radium atoms are located close to the grain or rock surface, ^{222}Rn may enter air-filled or water-filled pores or cracks, and thereby become accessible for further diffusion and transport. Reported diffusion length of radon is about 300 cm in air and 10 cm in water (Singh 1993). Water increases the absorption of the recoil energy, thus enhancing the chance that the atom will terminate its recoil within the water. The transfer from the rock surrounding to groundwater is mostly by diffusion through crystalline lattices and through and along cracks and the crystal boundaries. However, ^{222}Rn is short-lived (half-life, $T_{1/2}$ = 3.82 days), which will limit the distance of movement from the source before decaying. It is therefore expected that radon in groundwater would be correlated with the local uranium/radium abundance in rock and soil. Theoretically, radon can be transported by diffusion a distance of 5-10 m within its life, but in practice transport distances up to several 100 m were reported (Vàrhegyi et al 1992). Correlation by locations between radon concentration in water and radium or uranium concentrations in bedrock is therefore, expected to vary. It may therefore be useful to convert measurement points to continuous surfaces (raster or girded format) before performing such kind of correlation analysis. The importance of generating these surfaces is that they are used as the basic information to perform further spatial analyses in environmental applications. Based on these surfaces, it is possible to carry out additional analyses to answer questions related to environmental problems, such as health effects due to radon exposure. The accuracy of subsequent analyses directly depends on the accuracy of the surfaces built in the early stage of analyses.

Using the geographical information system (GIS) with geostatistical interpolation tools, it is possible to produce a surface for radon in water by estimating values at unsampled places. However, a problem arises when one attempts to use these tools to produce accurate surfaces from point measurements connected to another surface. This leads to the question: can these methods be used successfully to produce a radon in groundwater layer that is well correlated with bedrock radioactivity?

The kriging method is commonly used to estimate values at non-sampled places. It was applied for example to estimate soil radon concentration (Durrani et al 1997) or to construct indoor radon risk maps (Zhu et al 2001). Comparison between some estimation techniques including kriging and inverse distance weighting was investigated for the estimation of radionuclide concentration in soil, reported by Dowdall and O'Dea 1999.

The aim of this work is to test interpolation methods (kriging, co-kriging & IDW) for the purpose of determining the extent of spatial correlation of radon in water with bedrock radioactivity. This is accomplished by creating maps of radon concentrations in water from point measurements, which are then matched with existing bedrock uranium maps.

The study area is located in the south-eastern Sweden (see figure 1), between (N58°14', E14°53') and (N58°56', E16°06'). The area is characterized by crystalline bedrock with locally very high uranium content. In a central plain the dominant bedrock is oil shells and sand stones that may significantly contribute to radon. Another source of radon in the area are sand eskers from the prehistoric glaciation.

2. **MATERIAL AND METHODS**

2.1 Data

In a recent study we investigated radon in drinking water for 242 residential houses where private wells are used as a primary source of drinking water, unevenly distributed in the study area (figure1), which is located in the Östergotland county in the south-eastern Sweden, with coordinates range between: N 58°14' – N58°56' and E14°53' – E16°06'. Water samples were collected from the wells during 1998-2001, and measured by gamma spectrometry using high-resolution germanium detectors. Results from these analyses (Salih et al 2002) showed radon concentration ranging between 5 and 2750 becquerel per litre ($Bq \cdot l^{-1}$). The study area (150 x 134 km) has a geological structure characterised predominantly by granite formations. Uranium geological bedrock maps were obtained from the Swedish Geological Survey (SGU 2001).

2.2 Interpolation Techniques: descriptions

GIS modules are described to spatially interpolate data of radon concentration in water. Two interpolation techniques, inverse distance weighted (IDW) and kriging (& cokriging), used in the present study are briefly summarised below. Many GIS and geostatistical programs present today includes these modules (e.g. ArcMap "ESRI, 2001").

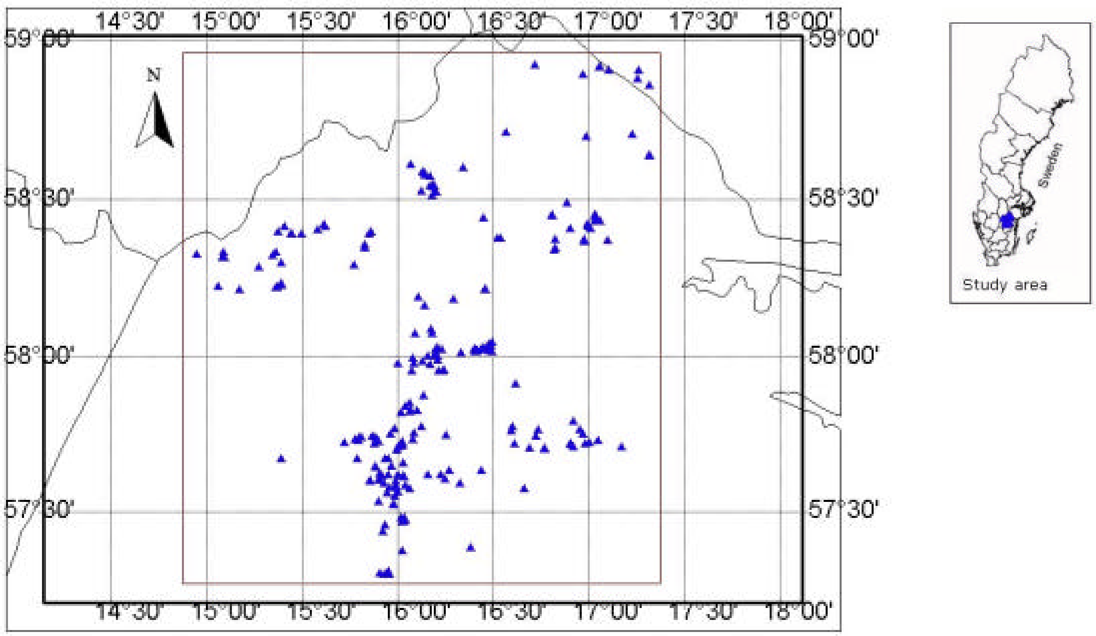

Figure 1: Study area showing 242 sampling locations unevenly distributed

2.2.1 IDW

The points are weighted during interpolation such that the influence of one point relative to another is a function of inverse distance. Weighting is assigned to points through the use of a weighting power and the radius object. The greater power means that the nearby points have the greater influence. Common value of power is two, which was chosen for interpolation in the present study. Most of the software available today allow choice between fixed or variable searching radius. This provides flexibility in performing good interpolation and choice based on the number of samples and their spatial distribution. As a part of this study a fixed radius option was selected to test locality or globalisation of radon transport. The input value of the radius was changed systematically and at each radius the interpolation was performed and the produced layer (radon in water) was compared with the bedrock uranium layer. Finally, cross validation parameters (root mean squares) and correlation coefficients between the generated maps and the uranium map were tabulated.

2.2.2 Kriging and cokriging

Kriging is an advanced interpolation procedure that generates an estimated surface from an x-y scattered set of points with z values (radon concentration). It is a weighted moving averaging method of interpolation derived from regionalized variable theory, which assume that the spatial variation of a property, known as a 'regionalized variable', is statistically homogenous throughout the surface. Kriging derive weights from the semivariogram that measures the degree of spatial correlation among data points in a study area as a function of distance and direction between data points. The semivariogram controls the way that kriging weights are assigned to data points during interpolation, and consequently controls the quality of the results. The semi-variogram γ(h) of a regionalized variable is defined by

$$g(h) = \frac{1}{2N(h)} \sum_{i=1}^{N} \left[Z(x_i + h) - Z(x_i) \right]^2 \qquad (1)$$

where x_i+h and x_i are sampling position separated by a vector h, Z(x_i) is a random variable at fixed position $x_{i,}$ and N(h) is number of data pairs separated by a vector h. Ordinary kriging is a well-known type of kriging interpolation that uses only the sampled primary variable to make estimates at unsampled locations. Cokriging allows one or more secondary variables to be included in the model and assuming that the primary and secondary variables are moderately correlated, the estimation accuracy of the primary variable should increase. Cokriging estimation ensures that the value of a variable estimated, on the basis of the neighboring values of one or several other variables, is the best possible based on the following criteria: a) the absence of bias between the estimated value and the true one and b) the minimization of the variance of the estimations.

3. RESULTS AND DISCUSSIONS

In this study two approaches for mapping radon in groundwater were investigated. The first approach deals with treating the whole dataset as one unit and creating one layer while in the second approach the data is split into sub regions according to the distribution of the samples. In both approaches both inverse distance weighted (IDW) and the kriging method were employed. In the second approach five sub-regions were created and the predicted surfaces were evaluated and correlated to bedrock layer and then merged. The subsections below show results from the two approaches:

3.1 Approach 1

Figure 2a shows the semivariogram fitting used in the kriging interpolation and Fig. 2b shows the kriging cross-validation of the obtained results. It is observed that the kriging method underestimates high values and overestimate low values (RMSE~ 232). In order to perform correlations between layers a layer of virtual points regularly spaced was constructed. Then zonal statistics were performed related to the points in this layer. Correlation analysis between the kriged layers and uranium map data showed poor relationships as can be seen from Fig 3.

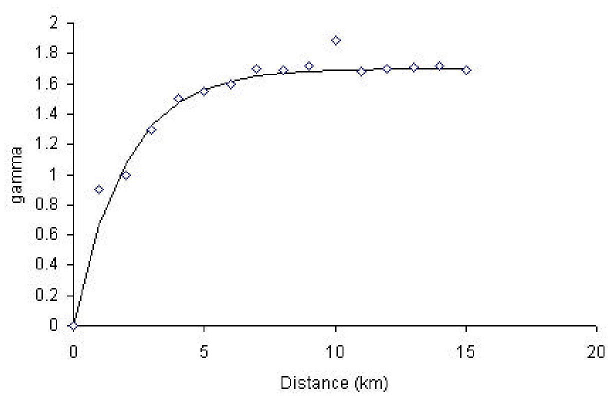

Figure 2a: semivariogram fitting used in kriging for mapping radon in groundwater.

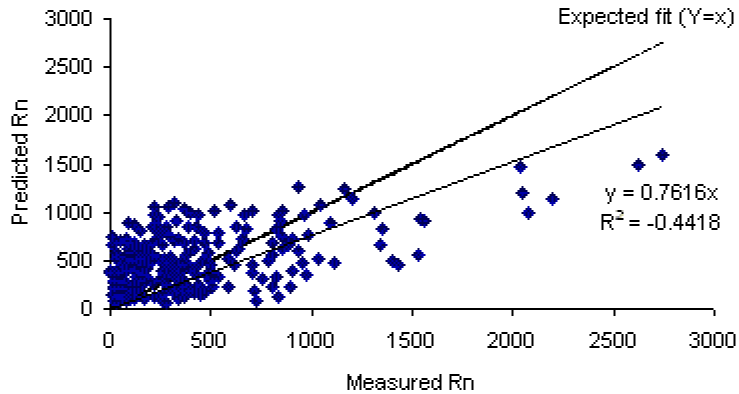

Figure 2b: Kriging cross-validation plot showing predicted Rn concentrations in water as a function of measured Rn concentrations

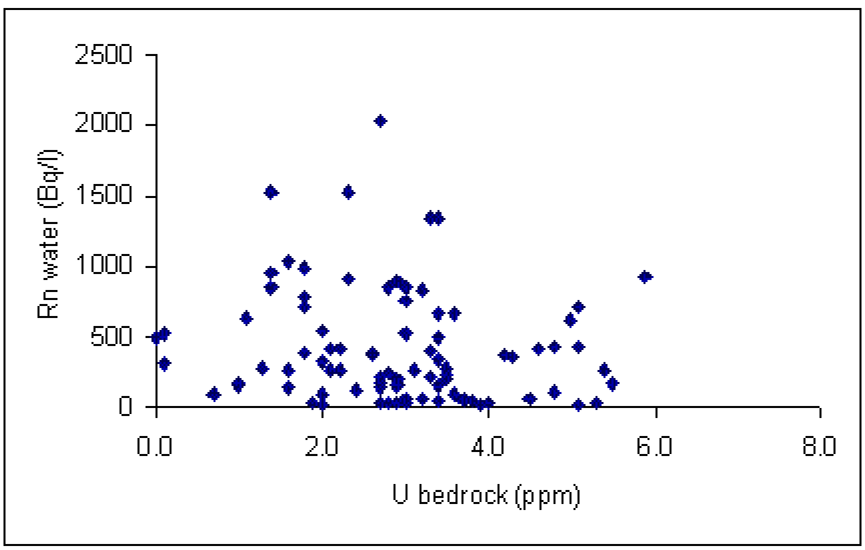

Figure 3: Uranium concentration in bedrock versus radon in water (predicted values via kriging)

The spatial variation of radon concentration in water is mainly attributed to the transport of radon from bedrock to water and mobilisation of groundwater. The aim was to see the extent of this variation i.e. how far (on average) radon is spatially correlated to uranium in bedrocks using geostatistics. The interpolation method of inverse distance weighting (IDW), however, helps to create raster surfaces being interpolated as a function of distance from point data by a rather simple procedure. It can be used to create a surface with fixed radius. It should be emphasised that the prediction characteristics might be scarce since, for example, maxima and minima are always among data points. That is because the inverse distance weighted interpolation is a smoothing technique by definition. On the other hand, it leads to reasonable predictions with no problem with results exceeding the range of meaningful values. In order to perform good correlation analysis a layer of virtual points (more than 1000 points with their coordinates and codes) was inserted between water and bedrock layers as shown in figure 4. The purpose of this was to control the correlation analysis by making use of zonal statistics at all locations in the layers. At these locations the predicted values obtained by the interpolations were compared with the values given from the uranium map. With fixed radius a surface of radon in water was generated and then compared with the bedrock uranium map. This process was repeated several times by varying the input value of the radius. Tabulated results of summary statistics for measured radon concentration and statistics for estimated values produced by each method are presented in Table 1. The table contains also correlation analyses between the produced layer and uranium in bedrock, presented in terms of correlation coefficient (R^2), and overall accuracy of the interpolations. For IDW the table shows only the best results achieved. The best compromise between good interpolation and correlation to the other layer was observed with the IDW procedure with fixed searching radius ranging between 1 and 2.7 km (RMSE ~ 377) when using all points for the interpolation.

The uranium concentration in the bedrock layer was used as a secondary variable to generate a groundwater radon map by the cokriging interpolation method. This method was modelled using cross variogram, which is required between the primary and secondary variables. The experimental models are fitted with a theoretical model and it is the theoretical model that is inserted into the cokriging. The co-kriged map was then compared with maps that were produced by kriging and IDW interpolation methods. A good correlation was obtained with IDW (R^2 ~ 0.6). Figure 4 shows three surfaces produced by overlaying surfaces obtained using Kriging, IDW method on uranium bedrock surface and a surface produced by cokriging.

Table 1: comparison between IDW, kriging and cokriging interpolations of radon concentration in water (in Bq/l). The table shows root mean square error (RMSE), correlation with bedrock uranium and interpolation accuracy and statistics.

	Measured	Predicted IDW	Predicted Kriging
RMSE	-	494.6	441.6
Max	2743.0	2595.9	1706.5
Min	4.7	24.4	221.3
Mean	421.8	400.8	415.5
Median	278.0	327.5	417.5
Std dev.	461.1	297.4	127.9
Overall unc*	-	18.3	51.3
Corr. U**	.01	0.45	0.02

* Overall uncertainty of the interpolation
** Coefficients of correlation to uranium in bedrock (R^2)

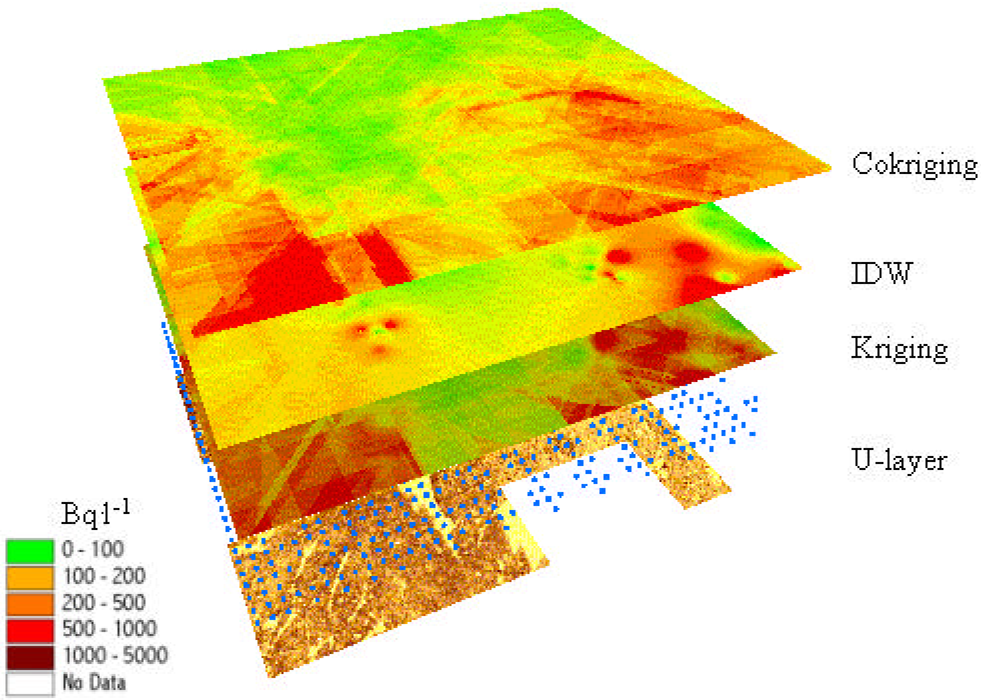

Figure 4: Maps of radon concentration (Bq/l) in groundwater produced, from 242 measurements, via IDW, Kriging and Co-kriging methods overlay on uranium bedrock map (0.0 – 6.0 ppm). The dot layer was inserted to control the statistical correlations between the layers.

2.2 Approach 2

This approach is rather investigates the situations where relatively small data sets are available. The data, as mentioned, were divided into five sub-regions analysed separately. This allows having higher sample density (sample per unit area) compare to the first approach. Based on Figure 1, these sub-regions are displayed in Figure 5, which shows results of one of the interpolation of radon concentration in groundwater. The figure displays also the grid layer (dots) that used for making correlation analysis between the different layers. The result of interpolations and comparisons between IDW and kriging, and their correlation to uranium in bedrock is presented in Table 2. Correlation to uranium in bedrock is expressed in term of coefficients R^2. The variable radius option in IDW was chosen here and the weighting power was optimised for each set. The interpolations were cross validated for both methods where the interpolation errors, root mean square errors (RMSE), were obtained. That will judge the effectiveness of the method in reproducing the original data set. From Table 2 it can be seen that the IDW produces a layer of radon in water that matched better with uranium in bedrock layer as compared to the kriging method. It also shows low interpolation errors. Comparison between the two methods is displayed in Figure 6 where the control layer (the layer of virtual points) was used to match the two results. Estimations obtained via kriging method, as shown in Fig 6, is on the average lower than that obtained via IDW; by 21.2% for radon concentrations above 500 Bq/l and higher; by 4.5% for concentration below 500 Bq/l.

Table2: comparison between IDW and Kriging for creating surface of radon in groundwater: root mean square error (RMSE) of the interpolation and correlation to uranium in bedrock shows the difference between the two methods for the five sub regions.

	IDW		Kriging	
Area	RMSE	Corr to U (R^2)	RMSE	Corr to U (R^2)
1	325.8	0.13	331.1	0.05
2	512.6	0.25	578.8	0.02
3	231.2	0.20	250.5	0.05
4	231.5	0.50	232.8	0.10
5	583.7	0.02	533.4	0.02

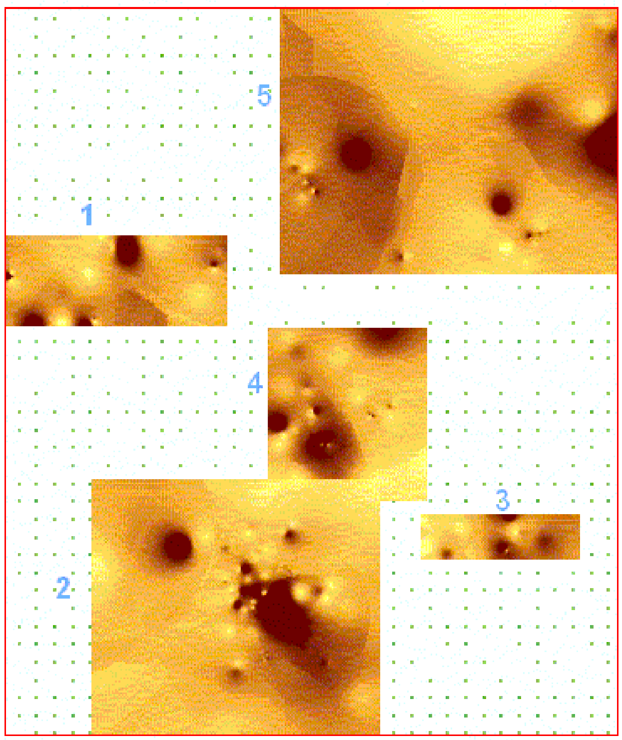

Figure 5: Interpolation results after splitting the data into five sub-regions; the figure also shows the grid layer (dots) that was inserted in order to facilitate calculations of correlation between different layers.

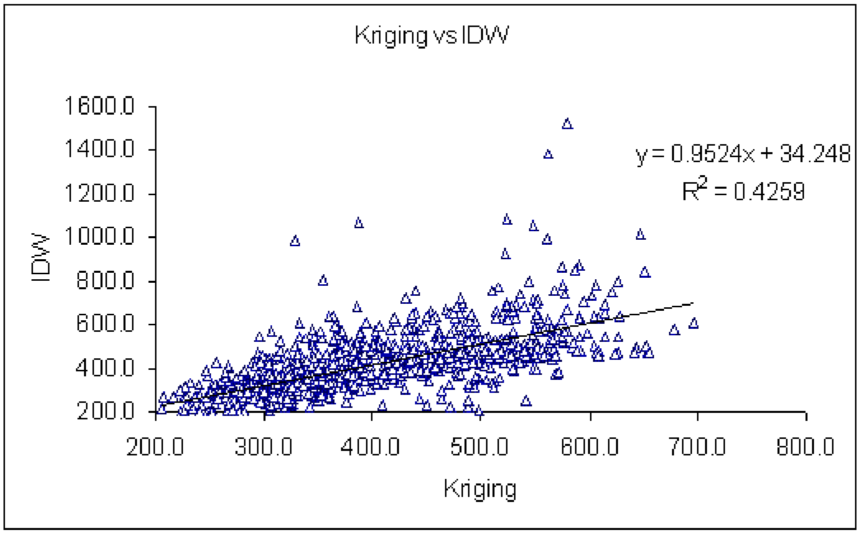

Figure 6: Comparison between IDW and kriging: the locations of virtual points in the dot layer are used for making the comparison.

4. CONCLUSIONS

This work provides a GIS and geostatistical methods for spatially analysing radon distribution in groundwater in connection to bedrock uranium. Two methods (IDW and kriging) were investigated for generating continuous surfaces of radon in ground water based on measured data. The kriging method produced good estimations at unsampled places (RMSE as low as 232), but showed a weak relation to bedrock uranium ($R^2 < 0.1$). This is attributed to several parameters, including groundwater mobilisation, radon transport and other factors that are difficult to incorporate here. Simple interpolation methods, such as IDW, with only one or two variables showed to be useful.

IDW with fixed radius showed to be useful in obtaining search distance that links between two auto-correlated layers (uranium in bedrock & radon in groundwater). The best correlations (R^2 = 0.5) were obtained for input radius between 1 and 2.7 km, i.e. distances far greater than expected for radon transport in water. This implies that there are more parameters involved in carrying radon in groundwater systems that needs investigation.

Therefore, other parameters, such as uranium, radium and about 70 stable elements are under evaluation in samples from the same sites used in the present work. The results of these analyses will be presented shortly. Future studies will include spatial evaluation of disequilibria and transfer factors of uranium and radium (the radon parent) from bedrock into groundwater and the impact of water chemistry.

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