

GEOSTATISTICAL MODELING OF HEAVY-METAL STREAM-SEDIMENT SAMPLES

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Abstract. Stream sediment samples present problems when studying its spatial variability as compared to the study of other spatially regionalized variables. Their continuity may be linked to their position along the stream paths and not so much to their absolute coordinates. This paper investigates this problem using two different metrics to analyze the spatial correlation of heavy-metal sediment concentrations measured on streams in a specific catchment. The first approach adopts non-Euclidean metrics and computes heavy-metal semi-variograms using distances measured along the water courses. The second approach uses the Euclidean distance for semi-variogram calculations. The first approach, which is more difficult to implement and more time consuming than the second one, yields variograms with a large nugget plus a short-scale structure, whereas the second approach shows clearly nested structures, including short range ones, of the same magnitude observed in the first approach, and long range ones, representative of some geological control. These results suggest that, for this particular dataset, Euclidean metrics are more effective in capturing the spatial phenomena underlying the sediment spatial distribution than non-Euclidean metrics, and, as a conclusion, the origin of the heavy metal contamination in the river sediments maybe due to the background geology rather than of anthropogenic origin.

Keywords. heavy metal pollution, stream sediment, semi-variogram calculation, non-Euclidean distance

1 Introduction

Improving prediction results is commonly a principal concern of geostatistical studies. Semi-variogram modeling is the first step of any geostatistical project. However, the standard way to compute semi-variograms may be questionable when spatial variability presents some particular pattern, beyond anisotropy. A good example is given in the paper by Dagbert *et al.* (1984), dealing with samples taken in a folded geological layer. In such case, the classical Euclidean distance between sample points is no longer suitable to compute the semi-variogram. According to these authors, historically this problem was first addressed in the early 1980's by T. Barnes and J. M. Rendu. Other authors,

discussed below, faced identical situations when applying geostatistical theory to possibly non-Euclidean spatial variability.

Dagbert *et al.* (1984) propose two methods to compute semi-variograms in folded geological layers, both methods are much complex to implement than the standard semi-variogram calculation and limited to layered geological deposits. Audergon *et al.* (1993) focused on theoretical aspects to propose three other metrics in addition to the Euclidean distance, when carrying out a geostatistical analysis of fruits in a tree. The application showed good results, while the main difficulties were measuring the different metrics proposed and interpreting what these metrics meant; besides the fact that the fruits in a tree are more amenable to be modeled as a discrete rather than a continuous variable. Gardner *et al.* (2003) tested three metrics for the distance between temperature loggers in a stream; they found that the more complex the metric used, the better its predictive capability. Skøien *et al.* (2005) proposed “Top-kriging”, a method “*for spatially interpolating a range of stream flow-related variables including mean annual discharge, flood characteristics, low flow characteristics, concentrations, turbidity and stream temperature*”. Ganio *et al.* (2005) worked on the geostatistical characterization of spatial patterns in stream networks. Dubois (2006), discussed the question of non-Euclidean geostatistics and argued that, although existing theoretical and practical problems “*exploratory non-Euclidean variography may provide valuable information*”. More recently, Peterson *et al.* (2010) proposed a moving-average approach that leads to a mixture of autocovariance functions (Euclidean and stream based) to be incorporated into a single geostatistical model.

However, none of these papers discusses the type of variable that should be analyzed using these complex metrics. For instance, its application may be evident for variables like fruit size, water temperature, water flow or fish growth, which are indeed linked to the stream paths; however, this is not so obvious for variables sampled within the streams but probably strongly linked to some process occurring at a larger scale, such as geological deposition. The latter variables may be partly influenced by the existence of the stream, but their spatial variability will be controlled by the underlying geology. To resolve this question a good knowledge of the variable regionalisation is necessary .

2 Case Study

The studied area is a hydrographic network sampled for heavy metals (Fig. 1). The aim of the study is to answer the following question: what is really controlling the heavy metal spreading? Is it the water, which flows across old and recent industrial areas? Or is it the geological background, which is known, in this case, to contain disseminated metal mineralizations?

The two options are very different. Their consequences for decision makers are paramount since, unless the heavy metal spreading obeys the first alternative, any remedial opera-

tion concentrated on the streams becomes useless and unnecessary. In order to compare these two hypotheses, we investigated two different approaches for semi-variogram computing. A one-dimensional (1D) approach, in which the continuity is determined by the stream, and a two-dimensional (2D) one, in which continuity is determined by the geological background. The two approaches have been tested and compared on real stream sediment data, sampled for six heavy metals (Zn, Co, As, Ni, Cu, Pb).

2.1 Data set

The Wallonian region (south Belgium) has been widely investigated for mineral exploration during the early 1980's. The campaign consisted in geochemical sampling of stream network to detect any potential sign of minerals upstream. Non navigable rivers were mainly concerned by this operation. According to Charlet *et al.* (1983), samples were collected within the recent alluvial, by means of soil auger at the intersection of the lower edge of the bank and the active bed, with a density of 1 sample/km² (nearly 1 sample every 200 m along stream). About 10.000 samples were collected and analyzed for 20 chemical elements.

In the late 90's, a new soil pollution law was approved in Belgium. This has led the governmental agency (DGRNE) in charge of pollution control in water resources to draw detailed charts of pollution risk using geostatistical techniques (Benamghar and Sonnet 1999, Benamghar 2002). These charts should then be compared with the underlying geology to try to understand whether the origin of the pollution was anthropogenic or geological. The Wallonia data set was used for this purpose.

Due to the great spatial extent of the Wallonia data set, the Amblève watershed in the eastern part of Wallonia, has been chosen as a testing area in this case study (Fig. 1).

3 Methodology

Usually, semi-variograms are computed using the Euclidean distance as the separation distance between samples; however, when analyzing variables which may be linked to one-dimensional, non-rectilinear features such as stream paths, it maybe appropriate to consider a different metric to measure distance. In our case, we have heavy metal concentration along streams and we wish to consider the length as measured along the stream path as the separation distance between samples, and compare the resulting variograms with those obtained using the Euclidean distance. In the first approach, semi-variograms are one-dimensional, whereas in the second approach semi-variograms are two-dimensional and may exhibit spatial anisotropy. We could interpret for the first approach that the “anisotropy” is so strong that continuity only happens along stream branches.

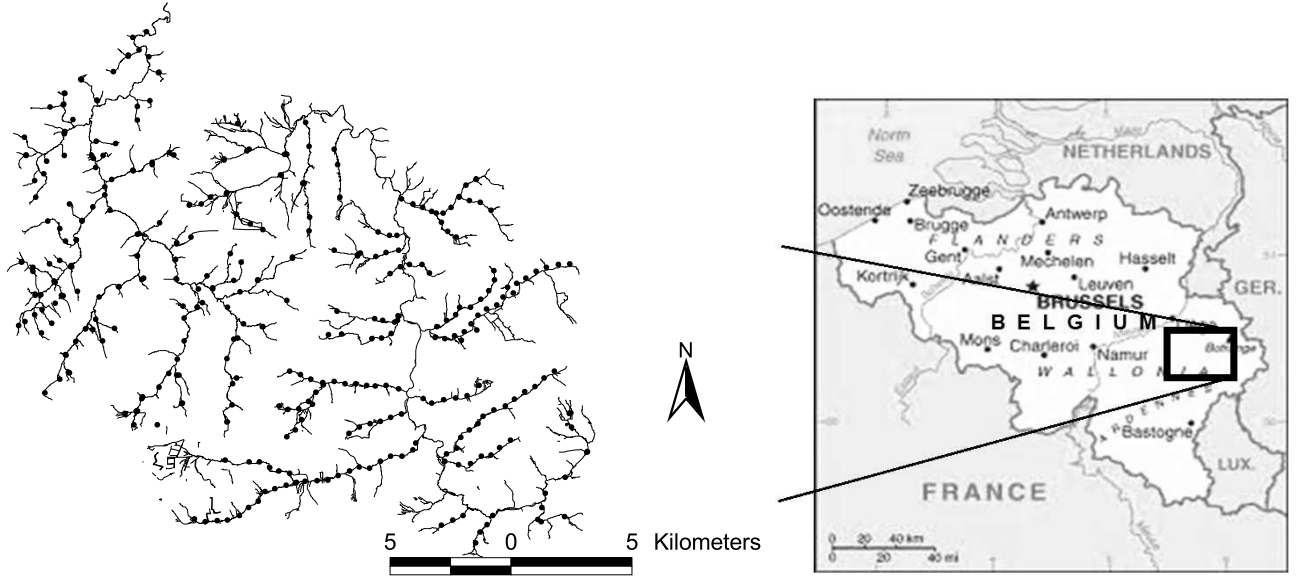


Figure 1: Left side: western part of Amblève watershed, studied area with samples (dots) and natural streams (curved lines). Right side: Amblève area south-eastern Belgium.

4 Theory

We consider that the heavy metal concentrations as a regionalized variable z that has been sampled at a finite number of locations $\{z(x_i), i = 1 \dots N\}$. We model this regionalized variable by a second-order stationary random function $Z(x)$, the spatial variability of which is characterized by a semi-variogram function $\gamma(h)$:

$$\gamma(h) = \frac{1}{2} E\{(Z(x) - Z(x+h))^2\}, \quad (1)$$

which is estimated from the experimental semi-variogram as

$$\gamma_e(h) = \frac{1}{2N(h)} \sum \{z(x_i) - z(x_i+h)\}^2, \quad (2)$$

where h is the metric that measures the distance between two samples, and $N(h)$ is the number of pairs found within the sampling domain that are a distance h apart (plus/minus some tolerance defined by the user).

The value of h is going to depend on whether we use as metric the distance along the streams or the Euclidean distance. There will be a need to perform some pre-processing of the sample coordinates to compute the semi-variograms for the first approach using standard semi-variogram computing codes. (This pre-processing basically amounts to stretching all river branches and to map them as parallel lines along which the 1D variogram will be computed.)

5 Results and discussion

On one hand, experimental semi-variograms were computed using stream distance as the separation distance (1D); they are displayed on the left columns of Fig. 2 and Fig. 3. The semi-variograms for As, Ni, Cu and Pb show an important nugget effect, some structure for the shorter lags, and a chaotic behavior for longer lags. This small structure that can be guessed for the shorter distances may indicate some localized dependency of the spatial variability on the stream network geometry. The remaining elements, Zn and Co, merely displayed chaotic structures, suggesting no spatial continuity along the watercourses.

On the other hand, and for the same elements, experimental omnidirectional semi-variograms were computed using the Euclidean distance as the separation distance (2D); they are displayed on the right columns of Fig. 2 and Fig. 3. The semi-variograms show a well structured pattern for all the elements, with some elements displaying two nested structures, with two distinct ranges.

It is important to remark that, the same four heavy metals (As, Ni, Cu, Pb) that show a slight spatial continuity with the 1D approach, displayed two nested structures for the 2D approach. Consequently, for these metals, it can be argued that the spatial behavior is controlled by a local range, limited to the stream extent, and by a regional one, reflecting a geological control as shown by Benamghar and Gómez-Hernández (2013).

For the other elements, Zn and Co, 2D model semi-variograms are composed of a nugget effect with a spherical model for Co and two nested structures for Zn. For Zinc, the small range is difficult to attribute to some watercourse control since the corresponding semi-variogram for the 1D approach was a pure nugget effect.

In the light of these results, one can already observe that the 2D semi-variograms reveal a regional spatial phenomenon as well as a local dependency when it exists. For this case study, the conventional approach to semi-variogram computation is sufficient to characterize the spatial variability of samples collected along the river network, and drives us to conclude that the origin of the contamination is due to the background geology.

6 Conclusion

The Amblève data set, which is part of the Wallonia geochemical data, has been used in this paper as a testing area. Six heavy elements were analyzed in this study: As, Ni, Cu, Pb, Zn and Co. The aim of this work was to examine whether the encountered heavy metal concentration in the stream sediments was related to the flow of water along the streams (and therefore it could be of anthropogenic origin) or merely reflecting the natural background. By analyzing two approaches for computing the semi-variograms we could conclude that much of the heavy metal concentrations is controlled by the geological background.

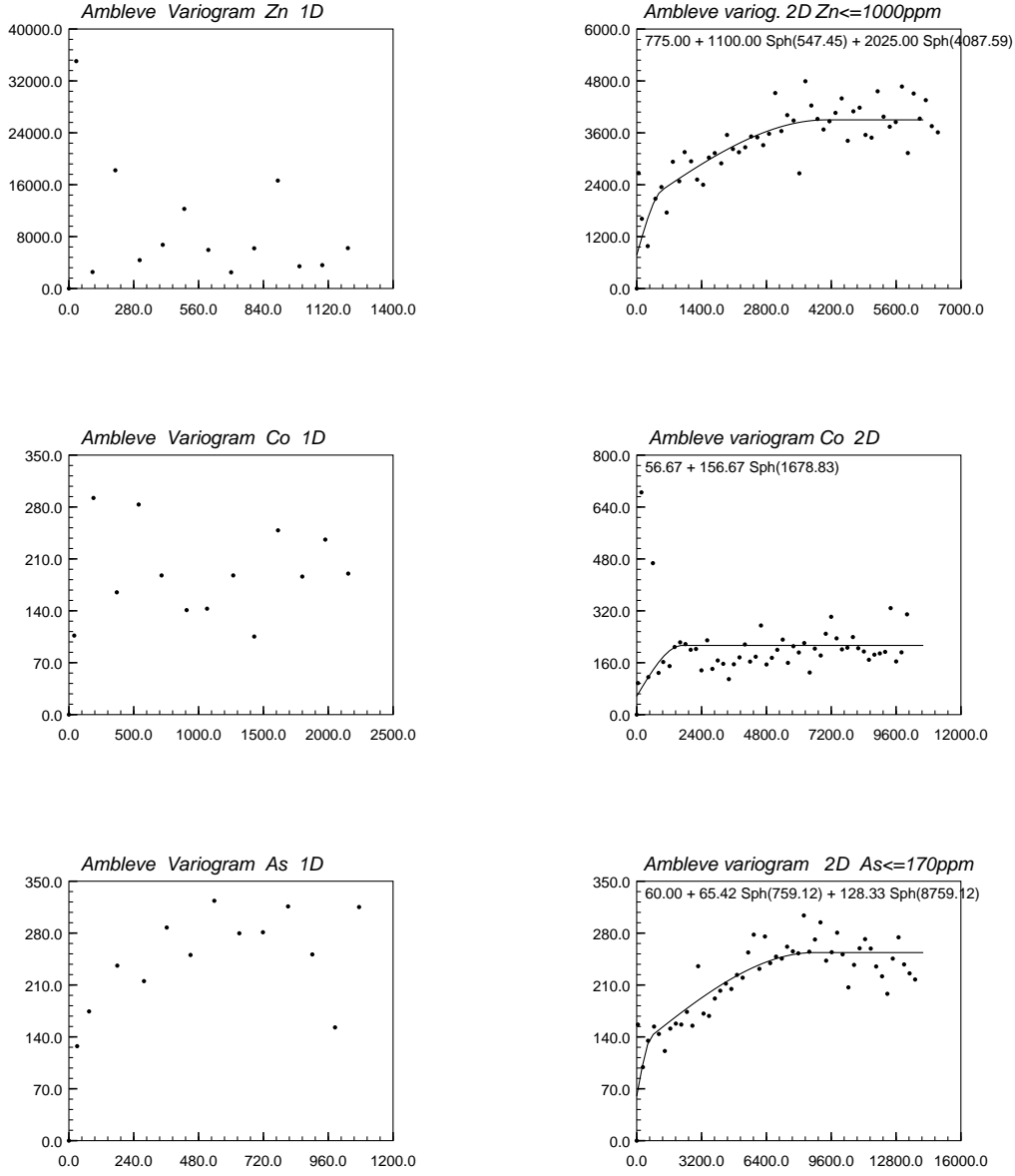


Figure 2: left column: 1D semi-variograms, right column: 2D omnidirectional semi-variograms; experimental (dots) and fitted model (solid line).

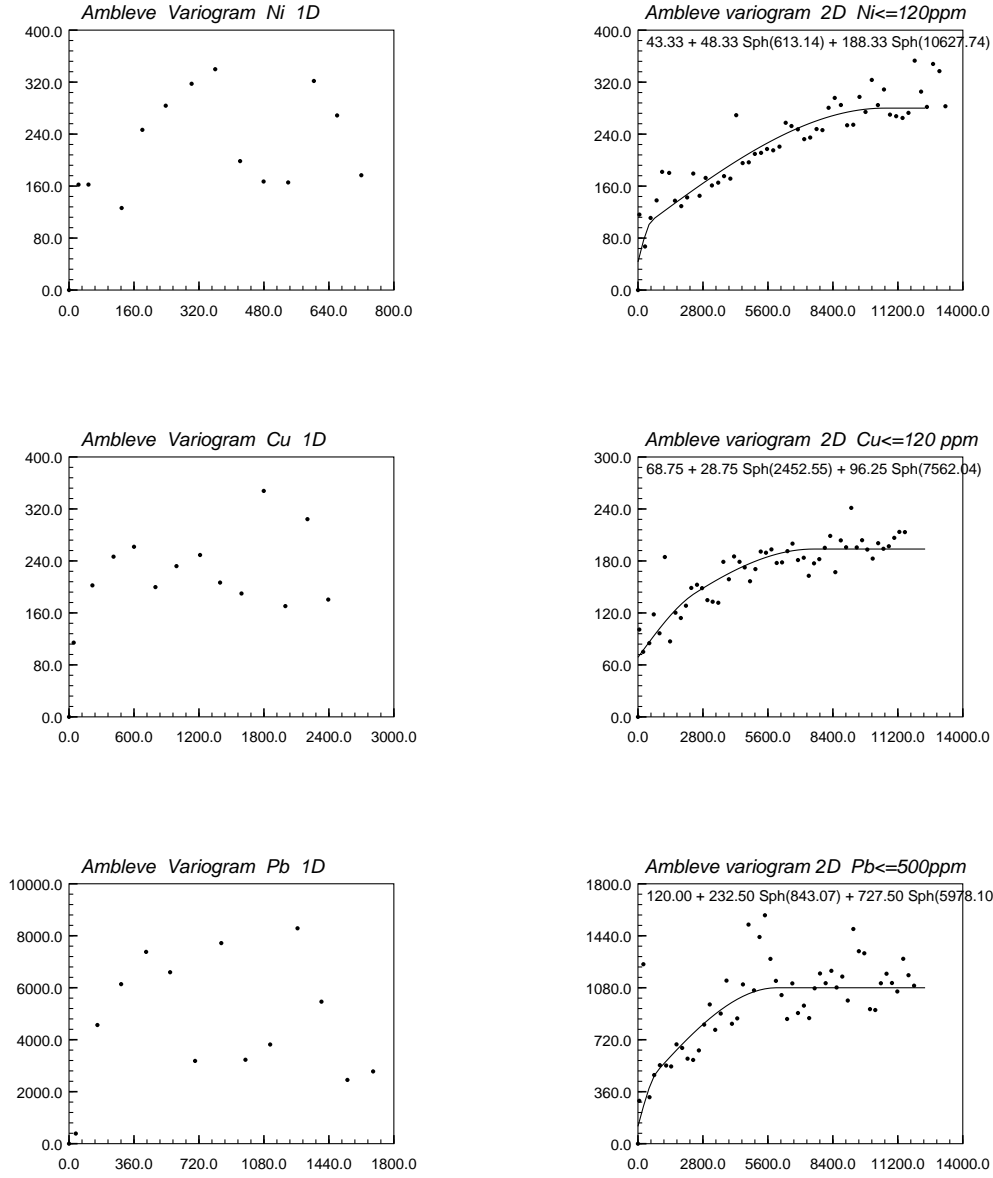


Figure 3: left column: 1D semi-variograms, right column: 2D omnidirectional semi-variograms; experimental (dots) and fitted model (solid line).

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