

## **SPATIAL VARIABILITY OF SOME HEAVY METALS IN SOILS AND CROPS DUE TO SLUDGE-AMENDMENT; Some preliminary results**

by Ronny Berndtsson\*, Akissa Bahri\*\*, and Kenji Jinno\*\*\*

The paper gives a preliminary analysis of intensively sampled soil and wheat plant tissue in the field in order to determine spatial properties of some selected heavy metals in a sludge-applied agricultural soil in northern Tunisia. A preliminary geostatistical analysis shows that both soil and plant metal contents have a spatial structure with a range of less than about 10 m. The sludge application has affected mainly Cu contents of both soil and plant tissue.

Keywords: Spatial variability, heavy metals, sludge-application, agricultural soils.

### **1. Introduction**

Spatial variability of agricultural fields is a multi-faceted and interdisciplinary problem that in its final stage results in spatially varying crop yields. There are many reasons why spatial variability of farming lands is important. The spatial variation of soil texture governs the transport of soil moisture as well as solutes. This is important to consider both for nutrient application for crops as well as in evaluation of potential pollutant transport to the groundwater. The variability in both the lateral and the vertical of soil-water and chemical transport properties seriously limits the applicability of the traditional column-scale approaches to analyze water and solute transport processes (e.g., Jury et al., 1987). The chemical properties of the soil interact with the infiltrating solutes and change the chemical properties of soil-water along its direction of movement. Thus, the spatial variation of soil chemical properties and the resulting variation of soil-water chemical properties (and vice versa) are among other things important to consider for crop yield, management of farmlands, and for the prediction of solute interaction and groundwater quality. This is especially important in arid and semi-arid areas where water and fertilizers are limiting factors for the crop growth.

Field variability of crop yield is a consequence of variation in the genetic properties of the plants and environmental factors (e.g., Bresler et al., 1981). The environmental factors in a single field that result in a certain spatial crop yield may be viewed as stemming from soil properties (e.g., hydraulic conductivity and dispersion) and soil variables (e.g., soil moisture, electrical conductivity, and sodium adsorption ratio; Bresler and Laufer, 1988). Besides this, applied water, fertilizers, and potentially toxic substances from sludge have to be recognized as a third important source of variability. Each of these components has a specific inherent scale of variability that may enhance or cancel the other superposed influencing effects.

Spatial variability of soil properties such as hydraulic conductivity has been studied rather extensively by, e.g., Nielsen et al. (1973) and Biggar and Nielsen (1976). The spatial variability of soil chemical properties has been studied to a rather small extent (e.g., Beckett and Webster, 1971; McBratney et al., 1982; Webster and Nortcliff, 1984; Wopereis et al., 1988). The spatial variability of crop yield components and its relation to field soil variability of soil properties and soil variables have been investigated by only a few researchers, e.g., Bresler et al. (1981, 1983), Warrick and Gardner (1983), Morcoc et al.,

\* Department of Water Resources Engineering, University of Lund, Lund, Sweden

\*\* Rural Engineering Research Center, Ministry of Agriculture, Tunis, Tunisia

\*\*\* Department of Civil Engineering (SUIKO), Kyushu University, Fukuoka, Japan

(1985), Bresler and Laufer(1988), and Bresler (1989). The field variability of applied sludge and resulting variability of uptake of toxic substances of crops, in turn, have been even less investigated. Boekhold et al. (1990) showed that spatial variability of Cd contents in an agricultural soil could be explained by spatial variability of pH and organic matter content. Boekhold et al. (1991) studied long-term behavior of Cd uptake by barley and effects of soil heterogeneity by a deterministic model approach. They found that groundwater and crop quality standards are exceeded earlier in heterogeneous fields as compared to a homogeneous soil profile. Yet, however, actual field studies on this topic are very rare.

The present study gives some preliminary findings from an extensive field sampling program at the experimental field research station Cherfech in northern Tunisia. In total 314 soil (20 cm long columns with a diameter of 7 cm from the upper soil horizon) and 78 plant samples (root, straw, and grain from a square area of  $0.3 \cdot 0.3 \text{ m}^2$ ) were collected in a  $3200 \text{ m}^2$  field plot. Half of this area was amended with domestic sewage sludge. Spatial properties of some heavy metals in soils and plant tissue are reported in this study.

## 2. Experimental layout and analytical methods

The sampling was done in two bordering squared plots each measuring  $40 \cdot 40 \text{ m}^2$  (see Figure 1). Soil samples were taken by a steel auger (a diameter of 7 cm) down to a depth of 20 cm and with a spatially similar pattern at three different scales as shown in Figure 1. Plant samples consisted of the wheat grown in a square area of  $0.3 \cdot 0.3 \text{ m}^2$ .

Until 1988 the two fields were treated equally concerning mineral fertilizers. In April 1988, one of the fields received about 47 tonnes anaerobically digested dry matter sludge per ha. The soil sampling was done directly after harvesting in June, 1990 (af Klercker and Svensson, 1991). Durum wheat was cultivated during two years previous the sampling (1989-1990).

The upper 20 cm of the soil profile appear spatially rather homogeneous with a clay content ( $< 2 \mu\text{m}$ ) of 55-75 % (Berndtsson and Bahri, 1990). The main clay mineral is montmorillonite (30-70 %). The organic matter content (estimated by loss of ignition) varies between 3-5 % of dry matter for the upper 20 cm. The crops, agricultural management, and hydrology of the area is further described by Bahri (1987a).

The sifted soil samples were dried and then dissolved by HF (40 %) and  $\text{HClO}_4$  (70 %) in closed teflon beakers (sludge samples were extracted in the same way). Plant samples were divided into ears, straw, and roots. The dried plant samples were dissolved by  $\text{HNO}_3$  (65 %) and  $\text{HClO}_4$  (70 %). The samples were analyzed by use of inductively coupled plasma mass spectrometry at the Department of Plant Ecology, University of Lund. The extraction technique for both soil and plants is meant to give total contents of metals. Further analytical details are given by Bahri et al. (1992).

## 3. Spatial patterns of heavy metals

Analyses of sludge samples revealed that especially some metals are likely to influence total contents in the soil. Ratios were calculated between the median metal concentrations found in the sludge and in the soil, respectively (based on 5 sludge and 314 soil samples). The highest ratios found are; Cu (7.61), Zn (5.28), Pb (4.12). Consequently, these metals are the ones most likely to influence the total concentration in the soil after sludge application. Other metals like Cd (1.00), Co (0.29), Cr (0.83), and Ni (0.66) display much lower ratios and sludge application therefore does not seem to constitute an immediate threat to the soil quality.

Figure 2 shows an example of the spatial variability of Cu (the metal with the highest ratio of sludge to soil concentration) for soil and wheat plant tissue. The figure is based on 26 samples for soil and 78 samples for crop components (26 samples each for root, straw, and grain) and interpolated and smoothed by use of kriging. A few outliers have been removed in order to fit the data to a theoretical normal probability density function (Bahri et al., 1992; Berndtsson et al., 1992). The figure shows in a rather striking way an apparent effect of the sludge-amendment. The left half of the field was sludge-amended and this is clearly seen in the figure as a rather apparent increase in average contents for all components starting at a distance of about 40 m. The variability appears to be modest for both soil and crop components except for roots. The roots display elevated concentrations in the middle of the sludge-applied field. At present it is not possible to explain these elevated concentrations. The overall increase for the sludge-amended field was found to be 18 % for soil, 15 % for roots, 38 % for straw, and 11 % for grain (based on 314 soil and 26 plant component samples). Similar results were obtained by Bahri (1987b) in another field study.

Figures 3-5 similarly show the spatial variation of Zn, Pb, and Cd, respectively, in soil and plant tissue. It is seen that for Zn there is no obvious increase in neither soil or plant tissue. For Pb there appears to be a gradient towards the sludge-applied field. However, no similar increase can be seen for the plant tissue. For Cd also a significant gradient appears to be present. However, this gradient may not stem from the sludge

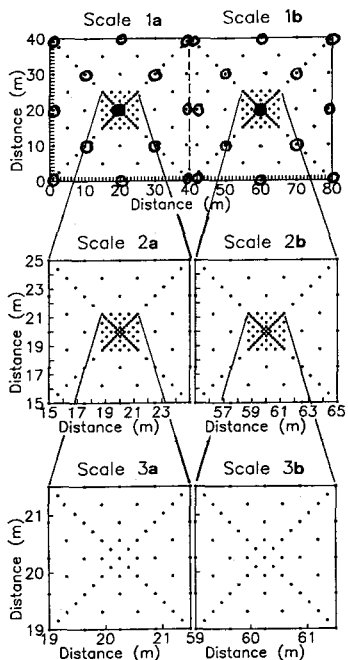


Figure 1 Soil and wheat plant sampling in the 40 × 80 m<sup>2</sup> experimental field. Plant samples are indicated by encircled symbols (a; not sludge-applied, b; sludge-applied).

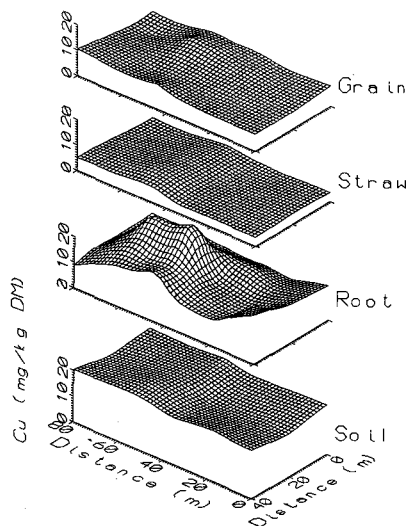


Figure 2 Spatial variation of Cu in soil and wheat plant tissue (mg/kg DM; the left half of the field was sludge-amended).

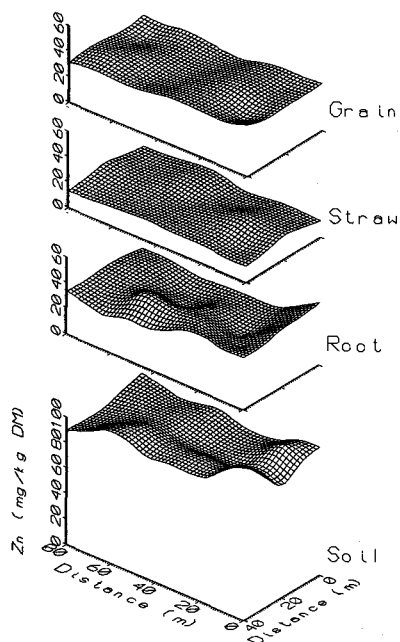


Figure 3 Spatial variation of Zn in soil and wheat plant tissue (mg/kg DM; the left half of the field was sludge-amended).

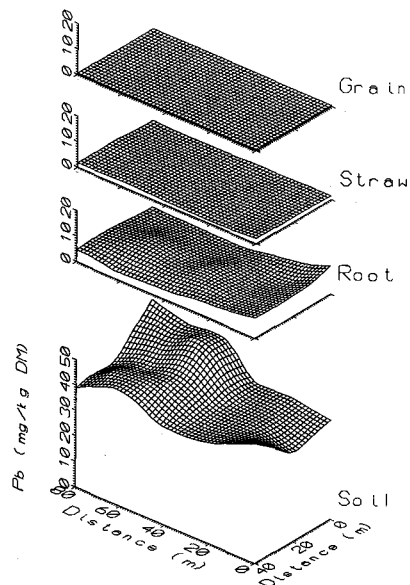


Figure 4 Spatial variation of Pb in soil and wheat plant tissue (mg/kg DM; the left half of field was sludge-amended).

application since the Cd contents in the sludge is similar to the natural soil content.

Table 1 shows the linear correlation between soil and plant tissue contents for selected heavy metals. It is seen that in general the correlation between soil and plant tissue contents is small. The correlation between plant tissue components is occasionally higher but coefficients are usually insignificant and no clear trend is observable.

Figure 6 shows normalized metal contents depending on soil and plant tissue for 7 heavy metals. The figure is based on 26 samples for each of the soil and plant components. The contents were normalized with the average soil content. It is seen how the content reduces from soil to plant tissue in a decreasing order. For some of the metals an accumulation appears to occur in the grain as compared to the straw tissue. This is the case for Cu, Zn, Co, and Cd. Especially for Cu, Zn, and Cd the concentration in the grain represents a significant part of the concentration in the soil.

Table 1 Linear correlation coefficients between concentrations in soil and plant tissue (based on 26 samples; Co concentrations were below the detection limit for straw and grain).

	Cd	Co	Cr	Cu	Ni	Pb	Zn
soil-root	0.16	0.49	0.12	0.27	0.51	-0.34	0.29
soil-straw	-0.40	-	0.07	-0.00	-0.01	-0.44	-0.30
soil-grain	0.00	-	0.32	-0.17	0.08	-0.15	-0.02
root-straw	0.16	-	-0.13	0.07	0.14	0.34	-0.13
root-grain	0.42	-	0.45	-0.04	0.08	0.25	0.08
straw-grain	0.46	-	0.22	0.15	-0.32	0.08	0.37

#### 4. Variogram analyses

Figures 2-5 are based on kriging-interpolated and smoothed data. However, analyses of the variograms and the correlation scale may yield a quantitative estimate over which scales individual samples are correlated. This is thus important for knowing the number of samples required to estimate mean values of the entire field within a desirable error margin as well as for investigating the connection between spatial soil and plant variability properties.

Figure 7 shows an example of variograms for Zn concentrations. It is seen that a clear spatial structure for soil, root, and grain is present. The range appears to be less than about 10 m. Since the spatial resolution of the observations was mainly greater than 10 it is not possible to elaborate on differences in the range between soil and plant components for scales less than 10 m. The variogram for straw appears almost completely random with no range.

Figure 8, similarly, shows variograms for Pb concentrations. Also here it is possible to notice a clear spatial structure. Soil, root, and straw display a clear tendency for increasing variance with separation distance. The variograms indicate a drift since no sill is observable. This is also noted in Figure 4 as a rather clear drift from the not sludge-applied field towards the sludge-applied field for soil and in opposite direction for roots. This drift was not separated before the variogram calculations. The variogram for grain in Figure 8 appears more erratic and it is difficult to distinguish meaningful spatial properties.

#### 5. Summary and discussion

The present paper has given a preliminary analysis of an intensive sampling campaign of total heavy metal concentrations in soil and wheat plant tissue for an agricultural field soil in northern Tunisia. This analysis may be concluded according to the following:

- 1 Spatial variability of soil chemical properties (such as heavy metals) are among other things important to consider for crop yield and management of farmlands and for the prediction of solute interaction and groundwater quality.
- 2 The sludge-applied soil showed a clear increase in Cu contents for soil and all plant components. The sludge-applied soil also displayed elevated Pb concentrations. No effect was discernible for other metals. Cu was the only metal with a clear increase in the plant concentrations for the sludge-applied field.
- 3 Spatial variability of heavy metals in soil and plant tissue displays a clear spatial structure and in most cases with a correlation scale less than about 10 m. Thus,

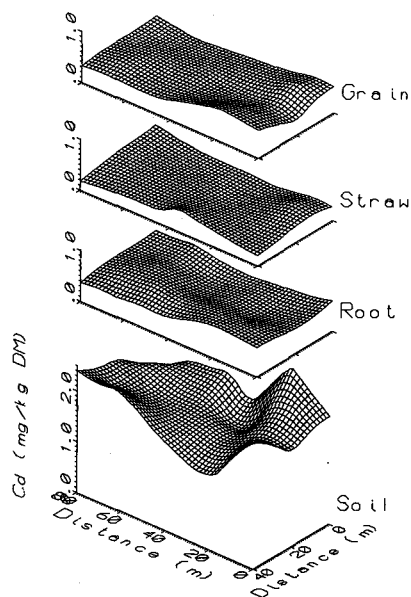


Figure 5 Spatial variation of Cd in soil and wheat plant tissue (mg/kg DM; the left half of the field was sludge-amended).

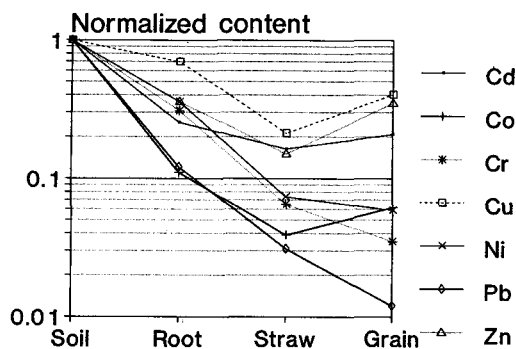


Figure 6 Normalized metal contents depending on component. The samples (26 samples) were normalized regarding average soil content.

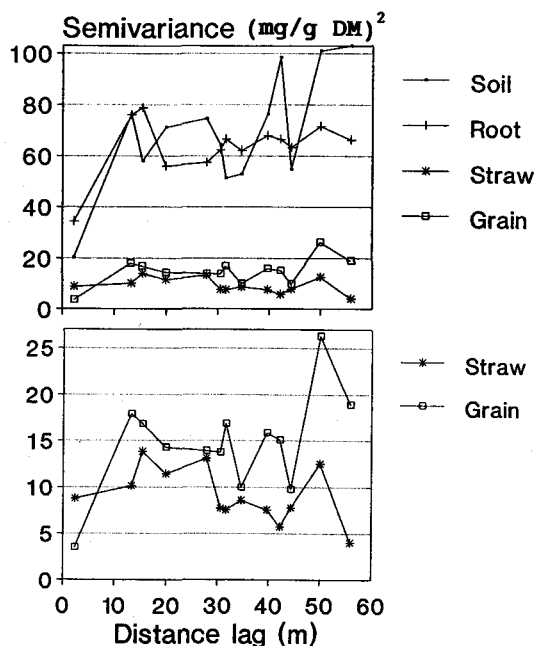


Figure 7 Variograms for Zn concentrations in soil and plant components.

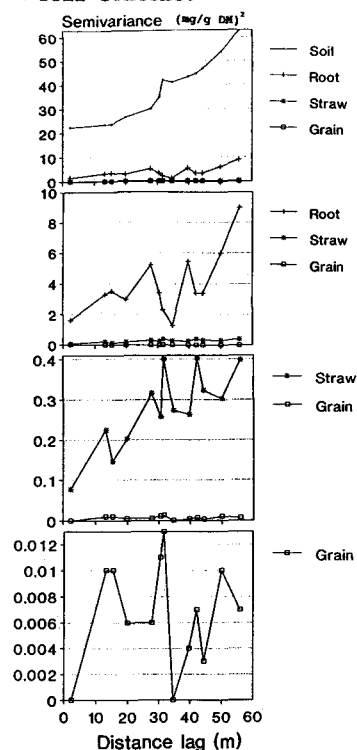


Figure 8 Variograms for Pb concentrations in soil and plant components.

optimal sampling should be done at an inter-sampling distance of more than 10 m to be spatially uncorrelated.

- 4 Some of the heavy metals appears to accumulate in the grain as compared to the straw tissue. This is the case for Cu, Zn, Co, and Cd. Especially for Cu, Zn, and Cd the concentration in the grain represents a significant part of the concentration in the soil (grain contents larger than 10 % of the soil content).
- 5 The joint variability of heavy metal contents in soil, roots, straw, and grain, respectively, appears to be poorly correlated. Thus, the spatial variability of heavy metals in the soil may to a less extent be revealed by the variability pattern of the soil (at least for total heavy metal contents).

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#### References

- af Klercker, P., and C. Svensson. 1991. Variation of trace elements in wheat and soil due to sludge application, a study at the experimental field station Cherfech in Tunisia. M. Sc. Thesis, Dep. of Water Resour. Eng., Univ. of Lund, Lund, 1-120.
- Bahri, A. 1987a. Water and salt movement in a silty clay loam. Research Center for Rural Engineering, Tunis, 1-13.
- Bahri, A. 1987b. Utilization of treated wastewaters and sewage sludge in agriculture in Tunisia. Desalination 67:233-244.
- Bahri, A., R. Berndtsson, and K. Jinno. 1992. Spatial dependence of major and trace elements in a semi-arid agricultural field soil, 1. Basic statistics and scale properties, Subm. paper to J. Soil Sci.
- Beckett, P. H. T., and R. Webster. 1971. Soil variability: A review. Soils and fertilizers 34:1-15.
- Berndtsson, R., and A. Bahri. 1990. Investigation of soil properties and solute transport at the Cherfech experimental field station. Rep. 3141, Dep. of Water Resour. Eng., Univ. of Lund, Lund, 1-60.
- Berndtsson, R., A. Bahri, and K. Jinno. 1992. Spatial dependence of major and trace elements in a semi-arid agricultural field soil. 2. Geostatistical properties. Subm. paper to J. Soil Sci.
- Biggar, J. W., and D. R. Nielsen. 1976. The spatial variability of the leaching characteristics of a field soil. Water Resour. Res. 12:78-84.
- Boekhold, A. E., and S. E. A. T. M. Van der Zee. 1991. Long-term effects of soil heterogeneity on cadmium behaviour in soil. J. Contam. Hydrol. 7:371-390.
- Boekhold, A. E., S. E. A. T. M. Van der Zee, and F. A. M. De Haan. 1990. Spatial patterns of cadmium contents related to soil heterogeneity. Water, Air, Soil Pollut. In press.
- Bresler, E. 1989. Estimation of statistical moments of spatial field averages for soil properties and crop yields. Soil Sci. Soc. Am. J. 53:1645-1653.
- Bresler, E., and A. Laufer. 1988. Statistical inferences of soil properties and crop yields as spatial random functions, Soil Sci. Soc. Am. J. 52:1234-1244.
- Bresler, E., G. Dagan, R. J. Wagenet, and A. Laufer. 1983. Statistical analysis of salinity and texture effects on spatial variability of soil hydraulic conductivity. Soil Sci. Soc. Am. J. 48:16-25.
- Bresler, E., S. Dasberg, D. Russo, and G. Dagan. 1981. Spatial variability of crop yield as a stochastic soil process. Soil Sci. Soc. Am. J. 45:600-605.
- Jury, W. A., D. Russo, G. Sposito, and H. Elabd. 1987. The spatial variability of water and solute transport properties in unsaturated soil, 1. Analysis of property variation and spatial structure with statistical models. Hilgardia 55:1-32.
- McBratney, A. B., R. Webster, R. G. McLaren, and R. B. Spiers. 1982. Regional variation of extractable copper and cobalt in the topsoil of south-east Scotland. Agronomie 2:969-982.
- Morcoc, F., J. W. Biggar, R. J. Millar, and D. R. Nielsen. 1985. Statistical analysis of Sorghum yield: A stochastic approach. Soil Sci. Soc. Am. J. 49:1342-1348.
- Nielsen, D. R., J. W. Biggar, and K. T. Erh. 1973. Spatial variability of field measured soil-water properties. Hilgardia 42:215-259.
- Warrick, A. W., and W. R. Gardner. 1983. Crop yield as affected by spatial variations of soil and irrigation. Water Resour. Res. 19:181-186.
- Webster, R., and S. Nortcliff. 1984. Improved estimation of micro nutrients in hectare plots of the Sonning Series. J. Soil Sci. 35:667-672.
- Wopereis, M. C., C. Gascuel-Oudoux, G. Bourrie, and G. Soignet. 1988. Spatial variability of heavy metals in soil on a one-hectare scale. Soil Sci. 146:113-118.