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REPORT

Geostatistical Assessment of the Impact of World War I on the Spatial Occurrence of Soil Heavy Metals

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Abstract Previous research showed a regional Cu enrichment of 6 mg kg^{-1} in the top soil of the Ypres war zone (Belgium), caused by corrosion of WWI shell fragments. Further research was required since in addition to Cu, also As, Pb, and Zn were used during the manufacturing of ammunition. Therefore, an additional data collection was conducted in which the initial Cu data set was tripled to 731 data points and extended to eight heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) which permitted (1) to evaluate the environmental impact of the heavy metals at a regional scale and (2) to assess their regional spatial occurrence by performing an optimized geostatistical modeling. The results showed no pollution at a regional scale, but sometimes locally concentrations exceeded the soil sanitation threshold, especially for Cu, Pb, and Zn. The spatial patterns of Ni and Cr were related to variations in soil texture whereas the occurrences of Cu and Pb were clearly linked to WWI activities. This difference in spatial behavior was confirmed by an analysis of coregionalization.

Keywords Soil heavy metals · World War I · Geostatistics · Spatial variability

INTRODUCTION

Only recently it was realized that soil contamination was also a form of collateral damage of the First World War (WWI). This was due to: (i) the corrosion of left-behind unexploded ordnance and metallic fragments of exploded ammunition (Pirc and Budkovič 1996; Souvent and Pirc 2001) and (ii) organic compounds originating from nitroaromatic explosives and the leaking of shells containing war gasses (Bausinger and Preuß 2005; Bausinger et al. 2008). Traces of this contamination can still be found today.

Metallic fragments found in the Soča front area (Slovenia) consisted of steel and cast iron fragments of shells, Cu–Zn alloy (brass) cartridges and Pb bullets and shrapnel balls. Those fragments caused an enrichment of Cu, Pb, Zn, Hg, and Sb with maximum concentrations of 225, 1005, 278, 9.3, and 7 mg kg^{-1}, respectively, obtained after sampling seven locations (Souvent and Pirc 2001). Around a former ammunition burning site in Belgium, 20 soil samples were taken revealing large heavy metal concentrations up to 4,331 for Cu and 369 mg kg^{-1} for Pb (Bausinger and Preuß 2005). Especially As (further admitted as heavy metal although it is a metalloid), which was used as a toxic agent since 1917, was defined as one of the major soil contaminants with concentrations between 1,120 and 2,595 mg kg^{-1} (Bausinger and Preuß 2005). Two years later, an even more severe soil contamination by As, Cu, Pb, and Zn was detected on a former ammunition burning site in Verdun (France) (Bausinger et al. 2008). On the other hand, an analysis of 30 soil samples in the Gallipoli front (Turkey) revealed that the soils were, in general, not contaminated. The low average concentrations of 15.1 for Cu, 13.9 for Pb and 36.7 mg kg^{-1} for Zn were explained by the good leaching characteristics of the soil and by surface runoff. A few high concentrations were measured and their locations were linked with intensive WWI activities (Baba and Deniz 2004).

The above mentioned studies were limited in spatial extent and based on a small number of soil samples. Van Meirvenne et al. (2008) were the first to assess the impact of WWI on soil heavy metal concentrations at a landscape scale based on a large amount of data points. Geostatistics was used to map topsoil Cu concentrations (0–50 cm) for the entire province of West-Flanders, Belgium (3,144 km^2)

revealing an average Cu enrichment of 6 mg kg^{-1} in the region around Ypres (covering 640 km^2). It was concluded that the enrichment was a heritage of WWI and was caused by corrosion of shell fragments. During WWI an estimated 1.45 billion (1.45 × 10^9) shells were fired by the combined German, French, and British armies on all fronts (Prentiss 1937). Around 95% of these were conventional explosive shells; the others released toxic gasses. The exact number of shells fired in the war zone around Ypres during WWI is unrecorded, but it must have been several tens of millions. For example, at the start of the "Third Battle of Ypres" the British forces already fired more than four million shells during the 15 days preceding the first infantry attack on 31 July 1917 (Keegan 2000, p. 361).

Although this study used a large database (2786) only a limited proportion of soil samples (199) were located within the war zone around Ypres. Furthermore, the chemical composition of the WWI artillery ammunition consisted not only of Cu, but also Zn was used in the top fuse of a shell, shrapnel balls, and bullets were made out of Pb and As was used in smoke generators and chemical warfare agents.

Even though no risk for human health was found, the Belgian government became conscious of the feasible effects of WWI on the geochemical soil composition. In order to assess the overall impact of WWI on soil heavy metal concentrations, a more detailed inventory and an extension towards other heavy metals were required. Therefore, the Belgian government provided the means to perform an additional data collection in which the number of available topsoil heavy metal data in the region around Ypres was raised to 731 data points. Each soil sample was analyzed for the heavy metals: As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn.

The primary purpose of this article is to present the results of the analysis of the enlarged data set. The objective is two-fold: (1) evaluate the environmental impact of the soil heavy metals as a result of WWI at a regional scale and (2) characterize their regional spatial distribution using geostatistics.

MATERIALS AND METHODS

Study Area

The study area of 640 km^2 is located in the region around Ypres in the south of West-Flanders, Belgium. It corresponds to the former WWI war zone along the Western front, which extended from the Belgian coast to the Swiss border (Fig. 1). The front line curved along the east of Ypres, which was the centre of intense and sustained warfare between the German and the Allied forces, such as the First (1914), the Second (1915), and the Third (1917) Battle of Ypres and the German Spring Offensive (1918). The city itself was never captured by the German army, but it was completely destroyed by artillery fire.

The soil in the war zone consists of Pleistocene (Weichsilian) wind-blown sediments deposited over Tertiary marine clayey sediments (Ypresian). The topsoil texture is mainly sandy silt in the north and centre of the area and changes gradually to silt in the south. In the utmost south and north-east, clayey soils occur. At present, most of the area is used for agriculture. The topography is weakly undulating with elevations mostly between 10 and 30 m. A chain of hills, from which the Kemmelberg is the highest (151 m), is present to the Southwest of Ypres.

Soil Sampling and Chemical Analysis

A first part (392) of our data originated from a database maintained by the Public Waste Agency of Flanders (OVAM), where data from soil pollution investigation studies are systematically recorded. Since 2000 it is required that data submitted are located precisely by their geographical coordinates. A former sampling campaign conducted by the Ghent University provided 38 additional data points (Tack et al. 2005). In 2008, we selected 199 locations based on geostatistical criteria and sampled them within the framework of this research. More details about the sampling campaign can be found in Meerschman et al. (in press). Finally, 102 data points were sampled according to a grid and considered as independent validation data. Figure 1 gives an overview of the 731 sampled locations.

The heavy metal concentrations of the soil samples were determined by a microwave digestion of the air dry fine-earth fraction (<2 mm) using HCl, HNO_3, and HF. In the digest, the metals were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES) or electrothermal atomic absorption spectrometry (ET-AAS), in conformity with the Flemish soil legislation.

Legal Soil Thresholds

Since 2008 a new legislation came into force in Flanders (Belgium), in which three legal soil thresholds were determined for each heavy metal: a background value, a target value and a soil sanitation threshold (Table 2). Target values can be interpreted as soil sanitation objectives (OVAM 2008). The 731 heavy metal concentrations were compared with the legal soil thresholds for a standard agricultural soil (defined as containing 10% clay and 2% organic matter, which are reasonable approximations of the average conditions in the area) to evaluate their environmental impact.

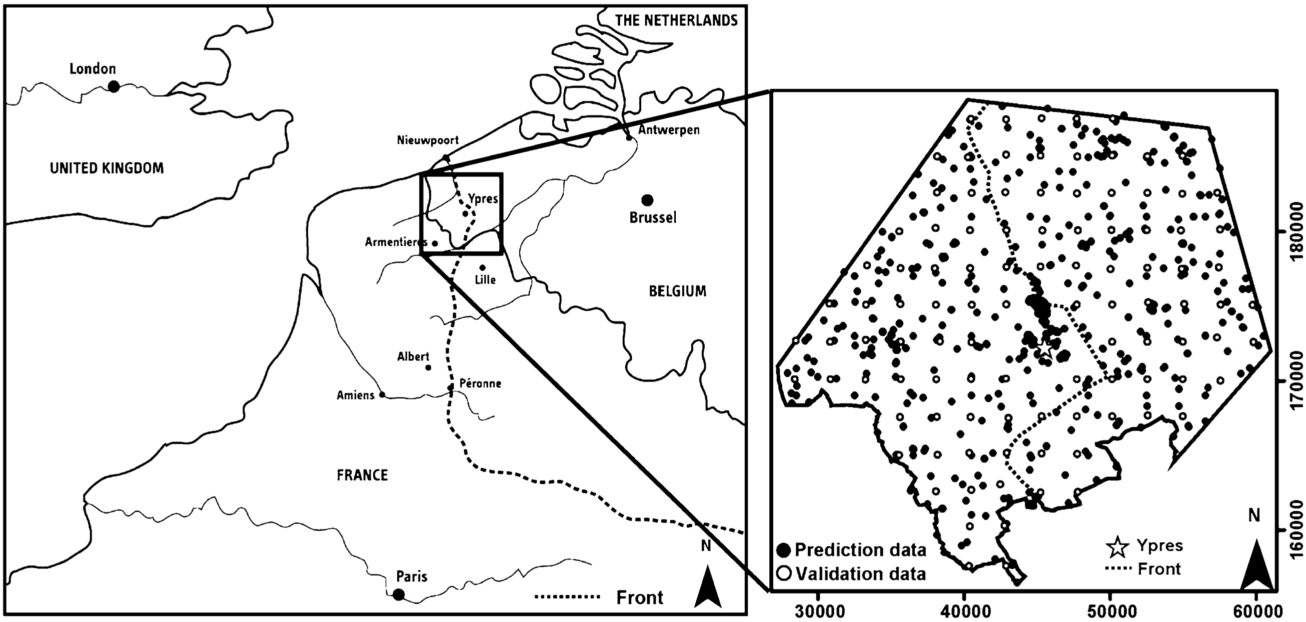

Fig. 1 Map of the Western front (Flanders Fields Documentation Centre Ypres) with indication of the study area and the available dataset with 629 prediction data and 102 validation data. The coordinates are in meter according to the Belgium Lambert-72 projection

Geostatistical Modeling

A geostatistical analysis was performed to set up prediction maps for each heavy metal. An accurate prediction for As, Cd, and Hg was complicated by the large amount of censored data. No less than 77% of the As data, 48% of the Cd data, and 68% of the Hg data were lower than or equal to the detection limit. A normal score transformation is not well suited for censored data since it requires a necessarily subjective ordering of all the equally valued observations (Saito and Goovaerts 2000). Histograms showed a positive skewness (Table 1; Fig. 2b). To overcome these limitations, Indicator Kriging (IK) was applied, using AUTO-IK software which executes the subsequent steps of IK including the modeling of the indicator variograms (Goovaerts 2009). We chose the detection limit as the first threshold z_1. The major drawback of IK is that high (tail) concentrations can be underestimated strongly, but the algorithm yields better results for the other classes (Saito and Goovaerts 2000).

The data sets with no censored data were interpolated using sequential Gaussian simulation (sGs) (Goovaerts 1997). TerraSeer STIS (TerraSeer Inc.) was used to transform the data values into normal scores, hereby transforming tied data values conditional to the local mean (radius 1,000 m) (Saito and Goovaerts 2000). In a second step, variograms of the normal scores were modeled using Variowin. Finally, SGeMS (Stanford University) was used to perform sGs at a 200 m by 200 m prediction grid (Remy

Table 1 Summary statistics of the heavy metal data (mg kg^{-1})

	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Minimum	3.7	0.01	5.9	0.85	0.04	1	1	6.7
Median	10	0.43	38	18	0.1	12.8	31	60
Maximum	70	2.5	210	250	1	70.2	950	620
Mean	10.5	0.52	40.7	23.3	0.14	14.2	47.6	81.9
Variance	21.4	0.09	272	423	0.01	59.5	3,826	5,795
Coefficient of variation	0.44	0.57	0.40	0.88	0.76	0.54	1.30	0.93
Skewness	5.3	2.4	2.4	4.1	3.7	2.3	7.2	3.6

et al. 2009). We chose Ordinary Kriging (OK) as kriging type and defined the search window as a circle with a radius of 5,000 m, which is larger than the variogram range. The maximum number of conditioning data was 12 and the number of realizations 500. The histogram of the non-transformed data was selected as the target histogram for back-transformation and the extrapolation model was optimized based on the independent validation data set.

Module *postsim* in SGeMS was used to derive the E-type (average of the ccdf) and M-type (median of the ccdf) from the realizations (Remy et al. 2009). An independent validation revealed that while the E-type was the best estimation for Cr and Ni, the M-type was best for Cu, Pb, and Zn, which can be explained by the strong positive skewness of those data distributions (Table 1). Finally, the optimized algorithms were repeated for all the available data, including validation data, to yield prediction maps.

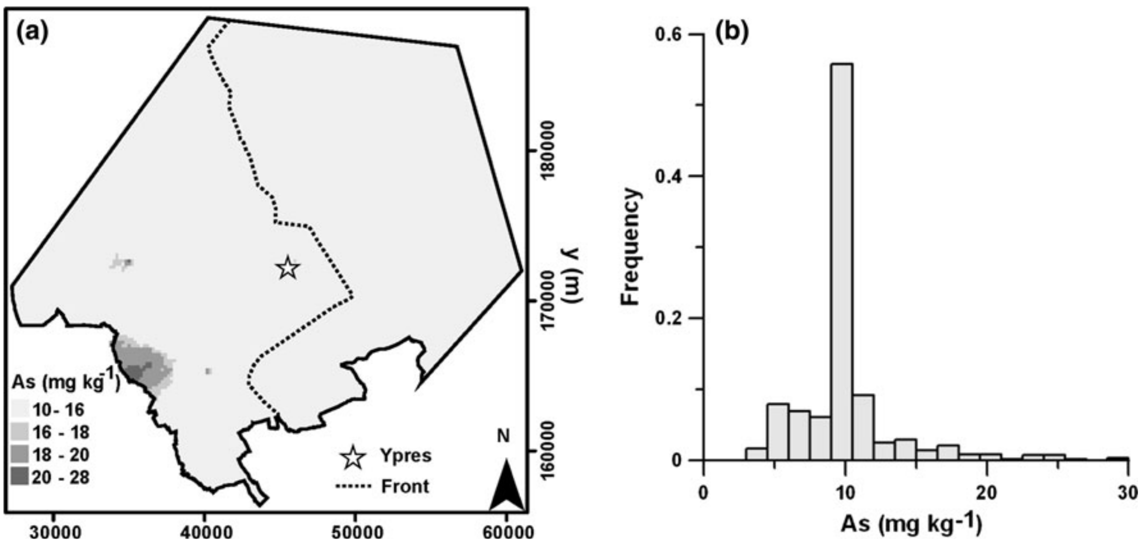

Fig. 2 **a** Prediction map for the topsoil As content obtained by indicator kriging (E-type) with indication of the centre of Ypres and the frontline which remained more or less stable between 1915 and 1917 (Chielens et al. 2006) and **b** the histogram of the 722 As data

Multivariate Data-Analysis

The non-spatial relationship between the eight variables was investigated by the Pearson's linear correlation coefficient r (Webster and Oliver 2007). Since r is sensitive to outlying pairs and strongly skewed distributions, data were first ln-transformed (Meklit 2009). We calculated also the Spearman's rank correlation coefficient r_R, which is a more robust measure of the bivariate relationship than r. A large deviation between r and r_R indicates a non-linear relationship between the variables or the presence of strongly disturbing outlying pairs (Isaaks and Srivastava 1989).

In order to investigate the spatial correlation, an analysis of coregionalization was performed. Therefore, two direct variograms $\gamma_1(h)$ and $\gamma_2(h)$ and the cross-variogram $\gamma_{12}(h)$ were modeled for four pairs of variables, satisfying the Cauchy–Schwartz triangular inequality (Ahmed and De Marsily 1987):

$$|\gamma_{12}(\mathrm{h})| \leq \sqrt{\gamma_1(\mathrm{h}) \cdot \gamma_2(\mathrm{h})} \quad \forall \mathrm{h}.$$

We chose ln Cu as the primary variable Z_1 and ln Pb, ln Zn, ln Ni, and ln Cr as secondary variables Z_2.

RESULTS AND DISCUSSION

Environmental Impact of the Heavy Metals

Table 1 shows the summary statistics of the heavy metal data, including the validation data. For each heavy metal, the median (as a robust measure of central tendency) was lower than or equal to the background value (Table 2). Therefore, we conclude that on average the soil is not contaminated at a regional scale. For Cu, Hg, Pb, and Zn the mean exceeded the background values, indicating a regional enrichment for Cu, Pb, and Zn. The result for Hg can be misleading because its background value is very low (equal to the detection limit).

In general, the majority of the data was lower than the background value (Table 2). However, 58% of the Pb data were higher than the background value, followed by Cu (46 %) and Zn (40%). For Ni, Cd, Cr, and As less than 30% of the data points were higher than the background value. Regarding the exceedance of the target value, Pb, Cu, and Zn scored the highest, followed by Cd and Cr. At 18 locations concentrations of Zn were higher than the sanitation threshold, while for Pb this was found at 16 locations, at 5 for Cu, and at 3 for Cd. For As and Cr one measurement was higher than the sanitation threshold.

We conclude that the environmental impact of the heavy metals is negligible at a regional scale. Recall that this conclusion is limited to the fine-earth fraction of the topsoil: fragments of ammunition larger than 2 mm have not

Table 2 Flemish legal thresholds for soil heavy metals (mg kg^{-1}) and number of data points higher than those thresholds

	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
ST	58	2	130	120	2.9	93	200	333
TV	35	1.2	91	72	1.7	56	120	200
BV	16	0.7	62	20	0.1	16	31	77
No of data > ST	1	3	1	5	0	0	16	18
No of data > TV	2	17	9	22	0	2	51	38
No of data > BV	46	127	59	306	234	189	351	229

ST sanitation threshold, *TV* target value, *BV* background value for standard agricultural soils (OVAM 2008) been investigated. At a local scale concentrations above the sanitation threshold were detected. Because studies of military contaminated sites typically reveal an inhomogeneous distribution of contaminants in soil, especially regarding metal fragments, no general conclusion about the impact of WWI at a local scale can be drawn. Whether there is an environmental risk at those locations, can only be verified by a large number of detailed local studies including historical investigations of land use, organic pollutants, etc., which is beyond the scope of this paper. It can be concluded that Cu, Pb, and Zn are the dominant heavy metals in the soils of the region around Ypres, whereas there was no indication of a regional enrichment in As.

Spatial Occurrence of the Heavy Metals

Figures 2, 3, and 4 show the prediction maps of As, Ni, Cu, Pb, and Zn with indication of the centre of Ypres and the WWI front, which remained more or less stable during 1915 and 1917 (Chielens et al. 2006). On each map the lowest category of the legend represents estimations below or equal to the background value and the highest category contains the estimations above the target value. Coordinates are according to the Belgian Lambert-72 metric projection.

Almost all As estimations were below or equal to the background value of 16 mg kg^{-1} (Fig. 2a), which confirmed our conclusion that there is no regional enrichment for As. The hotspot in the southwest of the study area was caused by two observations of 70 and 55 mg kg^{-1}. A link with industrial activities could not be found. However, whether this hotspot is caused by WWI activities could not be confirmed. Targeting As hotspots is only possible by increasing the sampling density or by opting for deductive research starting from historical information on, e.g., burning ammunition grounds. For Cd and Hg the largest part of the estimations were below or equal to the background value. Hotspots could be related to industrial activities, so no link between Cd or Hg concentrations and WWI was found.

Figure 3 shows the E-type prediction map for Ni and the topsoil texture map based on the Belgian soil texture triangle. It can be clearly observed that there is a link: increased concentrations were found in areas with clay soils. A few regions with elevated Ni concentrations were linked with industrial activities, e.g., the raised Ni concentrations to the north of Ypres are situated in a large industrial area. The spatial patterns of Cr were similar to those of Ni. Both maps showed no similarity with the localization of the WWI frontline.

The M-type prediction maps of Cu and Pb show spatial patterns of elevated values, whereas for Zn 90% of the estimations were below or equal to the background value of 77 mg kg^{-1} (Fig. 4). This might be due to the higher content of Cu compared to Zn in brass and the higher mobility of Zn in soil (Sipos et al. 2008). Still, areas with elevated Zn predictions were found along the WWI frontline.

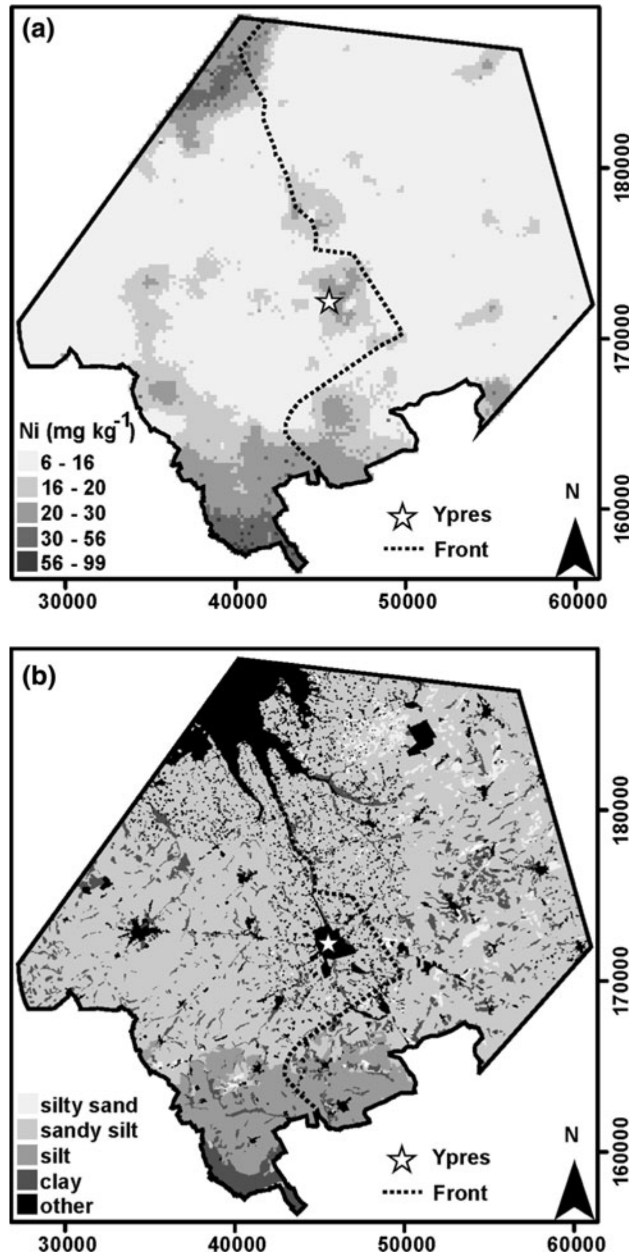

Fig. 3 **a** Prediction map for the topsoil Ni content obtained by sequential Gaussian simulation (E-type) with indication of the centre of Ypres and the frontline which remained more or less stable between 1915 and 1917 (Chielens et al. 2006) and **b** texture map of the study area based on the Belgian soil texture triangle with indication of silty sand (S), sandy silt (P + L), silt (L), clay (E + U), and "other soils" (mainly polder clay soils and anthropological soils); modified from the digital soil map of Flanders

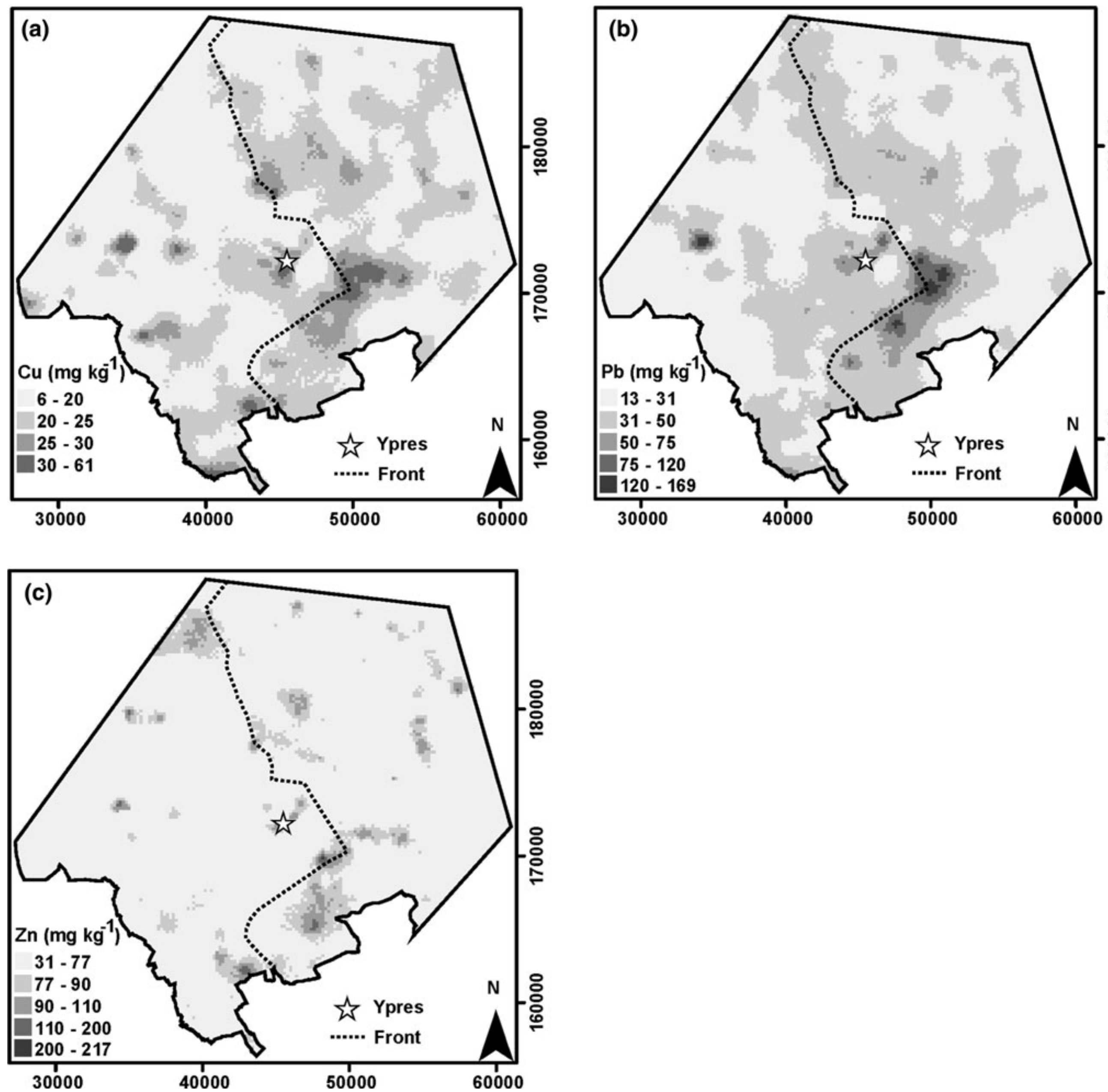

Fig. 4 Prediction maps for the topsoil **a** Cu, **b** Pb, and **c** Zn content obtained by sequential Gaussian simulation (M-type) with indication of the centre of Ypres and the frontline which remained more or less stable between 1915 and 1917 (Chielens et al. 2006)

Unlike with Ni and Cr, patterns of increased Cu and Pb estimations (Fig. 4a, b) were not related with soil texture (Fig. 3b). Besides to some isolated patches with elevated concentrations, a continuous band appeared around the WWI front line. The largest zone with increased Cu and Pb concentrations was situated in a prominent part of the Ypres Salient (between Zillebeke and Geluveld), where intensive battles took place (e.g., "Hill 60" which was heavily shelled and mined by both sides is located in this area). Furthermore, there were no industrial activities in or near to this area, arable land and forestry are the main land uses. No other heavy metal map showed increased concentrations in this area.

Finally, all the heavy metals show a local enrichment in the west of the study area which is situated around an industrial area in the town of Poperinge.

Multivariate Analysis

The correlation matrix of the ln-transformed data is given in Table 3. Both correlation coefficients were higher than 0.60 for Cr–Ni, Cu–Pb, Cu–Zn, and Pb–Zn. The strong

Table 3 Correlation matrix of the final dataset, including validation data, with the Pearson correlation coefficient *r* (left) and the Spearman rank correlation coefficient r_R (right) of the ln-transformed data; values larger than 0.60 are in bold

	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
As	**1.00/1.00**							
Cd	0.24/0.33	**1.00/1.00**						
Cr	0.29/0.30	0.13/0.22	**1.00/1.00**					
Cu	0.22/0.29	0.25/0.27	0.28/0.20	**1.00/1.00**				
Hg	0.18/0.18	0.06/0.06	0.13/0.11	0.35/0.38	**1.00/1.00**			
Ni	0.28/0.29	0.25/0.30	**0.66/0.63**	0.37/0.32	0.17/0.15	**1.00/1.00**		
Pb	0.18/0.24	0.29/0.28	0.16/0.17	**0.61/0.68**	0.37/0.40	0.30/0.33	**1.00/1.00**	
Zn	0.17/0.23	0.27/0.26	0.37/0.32	**0.67/0.70**	0.29/0.36	0.50/0.49	**0.63/0.68**	**1.00/1.00**

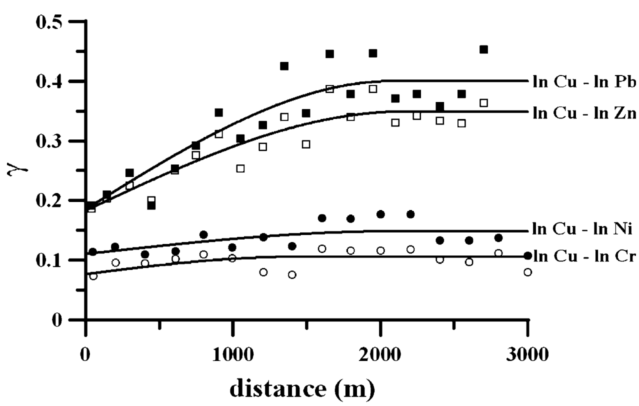

Fig. 5 Experimental cross-variograms of ln Cu – ln Pb (*filled square*), ln Cu − ln Zn (*open square*), ln Cu − ln Ni (*filled circle*), and ln Cu – ln Cr (*open circle*) and the corresponding cross-variogram models

correlation between Cr and Ni on one hand, and the war metals on the other, confirmed their joint occurrence as could be concluded from the prediction maps.

The coregionalization analysis showed that the cross-variograms of ln Cu − ln Pb and ln Cu – ln Zn displayed a strong spatially structured variability with a nugget to sill ratio (NSR) of 46% for Pb and 52% Zn (Fig. 5). The cross-variograms of ln Cu – ln Cr and ln Cu – ln Ni showed a weaker spatial structure with a NSR of 72% for Cr and 74% for Ni. Therefore, the war metals were not only strongly correlated but also characterized by a similar spatial structure.

Assessment of the Impact of WWI

We considered the differences in spatial structure between the war metals and Cr and Ni as an indication of the uniqueness of the source of Cu, Pb and, to a lesser degree, of Zn. Differences in parent material, metallurgical industry, amendments by slurry, and sewage sludge as possible heavy metal sources were considered and excluded by Van Meirvenne et al. (2008). Consequently, we identified WWI activities as being responsible for the occurrence of elevated concentrations of Cu, Pb, and Zn.

To our knowledge, this was the first time the impact of WWI on multiple heavy metal concentrations was investigated at a landscape scale based on a large number of soil samples. Even though the soil around Ypres is in general not contaminated, it was clearly shown that WWI affected the concentrations and spatial occurrence of Cu and especially Pb. Hence, this research contributed to our overall understanding of how war activities can affect the environment and more in particular the geochemical soil composition. It should incite researchers to acknowledge war activities as a possible source of soil heavy metals.

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