bedrock and human activities, the latter being the major source of high levels of metals in soils (Jean et al. 2007). The world production of Cr is in the order of 10^7 tonnes per year, of which 60–70% is used in alloys, including stainless steel, and 15% is used in chemical industrial processes, mainly leather tanning, pigments, electroplating and wood preservation (McGrath 1995). Due to its widespread industrial use, Cr has become a serious pollutant.

Cr is an essential trace element in the metabolism of human beings and animals (Shrivastava et al. 2002). Low concentrations of Cr can enhance the growth of plants. However, excess Cr is highly toxic to animals and plants and may induce cancer and teratism (Shanker et al. 2005). Cr occurs mainly in two stable oxidation states in the environment, Cr(III) and Cr(VI). Cr(III) is sparingly soluble and less toxic, while Cr(VI) being more soluble in water, is highly toxic to biota (Adriano 1986). Exposure to Cr(VI) compounds causes skin ulcerations, irritative dermatitis, allergic skin reactions, perforation of the nasal septum and bronchial carcinomas (Langard and Norseth 1979).

Cr in soils is essentially located in either primary or secondary minerals through isomorphic substitution with $Fe^{3?}$ and $Al^{3?}$ (Andersson 1977; Huisman et al. 1997). Some Cr may also be contained in insoluble hydroxides and oxides (Esser et al. 1991). Further studies of soils at the horizon or profile scale have shown that Cr is mostly present in the residual fraction (e.g. Gasser et al. 1995; Kaupenjohann and Wilcke 1995; Wilcke and Amelung 1996). Thus, Cr is widely considered to have a very low geochemical mobility (Alloway 1990). Consequently, Cr may be regarded as a conservative tracer chiefly indicating mechanical redistribution, such as accumulation of soil material and erosion.

Cr in soil may accumulate rapidly since it is only slowly depleted through plant uptake or erosion. Cr has been widely used in alloys and industrial processes and it has become a serious pollutant (Müller et al. 2005; Suciu et al. 2008). It is therefore necessary to study soil Cr accumulation in rapidly industrialized regions of China such as the Yangtze Delta and the Pearl Delta.

In past decades, a large number of soil pollution surveys on trace elements have been carried out at different scales and numerous studies have been reported in the scientific literature (Cullbard et al. 1988; von Steiger et al. 1996; Li et al. 2004; Murray et al. 2004; Shi et al. 2007). However, few investigations have been conducted on the accumulation of soil trace elements. Zhangjiagang City and Changshu City are two typical rapidly industrialized cities with a large number of township enterprises and self-employed firms in rural areas of the Yangtze Delta. In this study, the two cities were selected for investigation and the status of soil Cr accumulation was studied. The objectives were to investigate the differences in Cr concentrations between the topsoil and the subsoil of agricultural fields in the two cities and to elucidate the status of soil Cr mechanical redistribution.

Materials

Description of study area

The study area covers two coterminous cities, Zhangjiagang City (ZJG) and Changshu City (CS) located in southeast Jiangsu Province, east China (Fig. 1). ZJG is bounded by latitude 31 43^0–32 02^0 N and longitude 120 21^0–120 52^0 E with a total area of 999.5 km^2 and CS is bounded by latitude 31 30^0–31 50^0 N and longitude 120 33^0–121 03^0 E with a total area of 1266 km^2. They belong to the northern subtropical humid climatic zone and enjoy a temperate and humid climate throughout the year with four distinct seasons. The average annual temperature at ZJG is 15.2C and at CS 15.6, respectively. The mean annual precipitation in ZJG is about 1025 mm and in CS 1056 mm. A large number of township enterprises and self-employed firms were unevenly distributed in the rural areas of the two cities.

Soil sampling and chemical analysis

A total of 782 topsoil samples (0–20 cm) and 371 subsoil samples (20–40 cm) covering the two cities (ZJG and CS) were collected from agricultural land using a mixed sampling method from October to November 2003 with consideration of land use uniformity and soil types to ensure all samples were located in agricultural fields and a soil sample was collected from each soil group. The soil groups

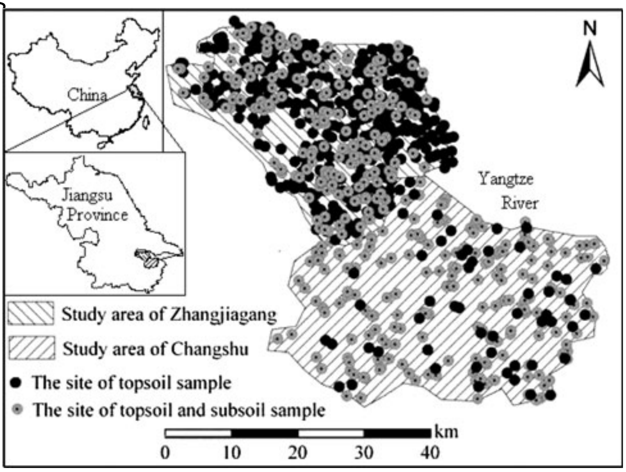

Fig. 1 General location of the study area, soil map, and sample distribution in the area

were classified according to the Chinese Soil Taxonomic Classification (see Soil Science Taxonomic Classification Group, Institute of Soil Science (CAS) 1995; Gong et al. 2003; Zhang and Gong 2004). The soil taxonomic classification is based on the diagnostic horizons and diagnostic characteristics, and the whole system consists of 14 soil orders, 39 suborders, 138 soil groups, and 588 soil subgroups (Gong and Zhang 2007). When sampling, soils in the top layer of 6–8 points in each site of an area of about 0.1–0.2 ha were collected and then thoroughly mixed, and finally divided into potions of 1–2 kg each. Only one of the portions was packed in a bag and brought back to the laboratory for analysis. All sampling sites were recorded using a hand-held global positioning system (GPS). The number of samples was as follows: ZJG topsoil samples, 543; CS subsoil samples, 185; ZJG topsoil samples, 239; and CS subsoil samples, 186.

The total concentration of Cr was determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES) after the samples were digested with aqua regia. Quality control of the methodology was checked based on certified reference samples of GSS-3.

Methods

Statistical methods

In multivariate statistics and linear geostatistics, a normal distribution for the variables under study is desirable (Gallego et al. 2002; Webster and Oliver 2001; Zhang and McGrath 2004). Even though normality may not be strictly required, serious violation of normality, such as too high skewness, can impair the reliability of statistical results (Zhang and McGrath 2004). To avoid the resulting distortions and low levels of significance, logarithmic transformation was performed on the Cr concentrations in CS.

One-way analysis of variance

To compare the difference in soil Cr concentrations between ZJG and CS, the natural logarithm transformed chromium data on concentrations in topsoil and subsoil were classified into two classes based on sampling site (ZJG or CS). The variances of the two classes were analyzed by using one-way analysis of variance (ANOVA) using SPSS for Windows ver.13. In ANOVA, Scheffe's test was chosen for post hoc multiple comparisons.

Paired-samples *t* test

Paired-samples *t* test was used to compare means on the same or related subject over time or in differing circumstances. The test statistic is *t* with $n - 1$ degrees of freedom. If the *P* value associated with *t* is low (<0.05), there is evidence that there is a difference in means across the paired observations. To compare the difference of Cr concentrations between in topsoil and in corresponding subsoil, the data of the natural logarithm transformed chromium concentrations in topsoil and subsoil were classified into two classes. The difference of two classes was analyzed by using Paired-samples *t* test carrying on SPSS for Windows ver.13.

Semivariance analysis and kriging interpolation

The semivariogram or cross-semivariogram is an effective tool that can evaluate spatial variability (Boyer et al. 1991; Cahn et al. 1994), which should be illustrated before conducting spatial prediction using kriging and cokriging (Kerry and Oliver 2004). Spherical model and exponential model are the two models that are frequently used in environmental studies. Although differences between spherical model and exponential model are distinct, the prediction accuracy would not be affected significantly by the choice of model unless it was unreasonable (Heuvelink and Webster 2001). Anisotropy of variograms was not found for the data set. All the semivariograms and cross-semivariograms in isotropic form were fitted to spherical model or exponential model or Gaussian model, and the best fitting semivariogram/cross-semivariogram model that had relatively higher coefficient of determination and lower residual sums of squares for geostatistical prediction was selected (Wang 1999).

Geostatistical prediction methods can be used in unbiased prediction with minimum variance for the concentration of a given pollutant (Stein and Corsten 1991). Kriging and cokriging interpolation are two such geostatistical prediction methods that can provide an estimate for a discrete area around an interpolation point. To know the spatial distribution of Cr in topsoil and subsoil, they were predicted by kriging and cokriging, respectively. Before interpolation, natural logarithmic transformation was applied to the Cr concentration data in CS. The local discretization grid was 3 × 3, with the minimum number of neighbors of eight used in interpolation. After interpolation, an exponential function was used to back transform the data that had been logarithmically transformed. All semivariance analyses and kriging/cokriging interpolations were performed using GS+ Geostatistics for the Environmental Sciences ver.7.

To compare the difference in spatial distribution between Cr concentrations in topsoil and in subsoil, the spatial distribution maps of soil Cr concentration differences in ZJG and CS were acquired by means of two prediction maps in imagery form by subtraction using version 9.0 of ERDAS IMAGINE by using operators function.

Results

Pollution status of soil Cr

The total Cr concentrations had a wide range for ZJG topsoils, 17.6–104.4 mg kg^{-1} with a mean of 57.9 mg kg^{-1}; ZJG subsoil, 18.0–88.3 mg kg^{-1} with a mean of 51.2 mg kg^{-1}; CS topsoil, 18.0–112.9 mg kg^{-1} with a mean of 52.3 mg kg^{-1}; and CS subsoil, 24.0–99.7 mg kg^{-1} with a mean of 48.1 mg kg^{-1}, respectively. The coefficients of variation (CV) were ZJG topsoil, 22.9%; ZJG subsoil, 25.6%; CS topsoil, 35.8%; and CS subsoil, 30.1%, respectively (Table 1). There were 8 topsoil samples and 0 subsoil samples in ZJG, and 15 topsoil samples and 2 subsoil samples in CS whose total Cr concentrations exceed the pedogeochemical background value (90 mg kg^{-1}) based on the Chinese Environmental Quality Standard for Soils (GB 15618-1995) (State Environmental Protection Administration of China 1995).

Comparison of Cr concentrations

Both mean and median concentrations of Cr in ZJG were greater than in CS, whether in topsoil or subsoil (Table 1).

Table 1 Statistics of the chromium concentrations (mg kg^{-1}) in different regions and soil depths

	ZJG		CS	
	Topsoil (0–20 cm)	Subsoil (20–40 cm)	Topsoil (0–20 cm)	Subsoil (20–40 cm)
n	543	185	239	186
Min.	17.6	18.0	18.0	24.0
Max.	104.4	88.3	112.9	99.7
Mean	57.9	51.2	52.3	48.1
Median	58.10	49.8	46.1	44.7
SD	13.28	13.09	18.70	14.50
CV (%)	22.9	25.6	35.8	30.1
Skew[a]	0.25	0.15	1.27 (0.57)	1.58 (0.18)
Kurt[a]	0.58	- 0.34	0.82 (0.12)	2.12 (0.35)

n sample number, *Min* minimum, *Max* maximum, *SD* standard deviation, *CV* coefficient of variation, *Skew* skewness, *Kurt* kurtosis

ZJG all soil samples collected from Zhangjiagang City, Jiangsu Province; *CS* all soil samples collected from Changshu City, Jiangsu Province

[a] Outside the parentheses is the Skewness or Kurtosis of the original variable, and inside the parentheses is the Skewness or Kurtosis of the natural logarithm-transformed variable

Table 2 Post hoc multiple comparisons using Scheffe's test of the natural logarithm-transformed chromium concentrations at the two soil depths from the two study areas

Object		Probability level
Study area	Soil layer	
Source		
ZJG	Topsoil	0.000
	Subsoil	0.000
CS	Topsoil	0.000
	Subsoil	0.000

Topsoil soil sample at 0–20 cm depth, *Subsoil* soil sample at 20–40 cm depth

ZJG soil samples collected from Zhangjiagang City, Jiangsu Province, *CS* soil samples collected from Changshu City, Jiangsu Province

Table 3 Paired-samples *t* test of the chromium concentrations in topsoils (0–20 cm) and corresponding subsoils (20–40 cm) from the two study areas (ZJG and CS)

Study area	N (pair)	Correlation coefficient	t	Sig. (2-tailed)[b]
ZJG	185	0.83**	5.13	0.000
CS	186	0.84**	1.86	0.065

Raw data of chromium concentrations in ZJG and the natural logarithm transformed data of chromium concentrations at CS

ZJG paired soil samples collected from Zhangjiagang City, Jiangsu Province; *CS* paired soil samples collected from Changshu City, Jiangsu Province, *t*, value of *t*, *Sig.*, significance level

** significant at $P < 0.01$

The result of post hoc multiple comparisons by Scheffe's test (Table 2) reveal that differences in mean concentrations were found between ZJG topsoil and CS topsoil and between ZJG subsoil and CS subsoil at the 0.05 level.

The mean and median concentrations of Cr in topsoil were also greater than in subsoil, whether in ZJG or CS (Table 1). The result of paired-samples *t* test (Table 3) reveals that differences in Cr concentrations were found between in ZJG topsoil and ZJG subsoil at the 0.000 level; however, the Cr concentrations in CS topsoil were not significantly different from those in CS subsoil at the 0.05 level. The difference in Cr concentrations between topsoil and subsoil had a wide range for ZJG, 23.7–59.6 mg kg^{-1} with a mean of 3.04 mg kg^{-1}; and CS, - 25.6–45.9 mg kg^{-1} with a mean of 1.42 mg kg^{-1} (Table 4). The Cr concentrations in topsoil were strongly correlated with those in subsoil, and the correlation coefficients were ZJG, 0.83 ($P < 0.05$); and CS, 0.84 ($P < 0.05$), respectively. This indicates that the Cr in topsoil has the potential to be a good auxiliary variable for Cr in subsoil prediction.

Table 4 Statistics of the differences in chromium concentrations (mg kg^{-1}) between topsoils (0–20 cm) and corresponding subsoils (20–40 cm) from the two cities (ZJG and CS)

	Min.	Max.	Mean	Median
ZJG (n = 185)[a]	- 23.7	59.6	3.04	0.17
CS (n = 186)[a]	- 25.6	45.9	1.42	0.66
	Min.	**Max.**	**Mean**	**Std dev.**
ZJG[b]	- 18.2	24.9	6.32	5.63
CS[b]	- 31.2	28.9	2.13	6.84

Min. minimum, Max. maximum, Std dev. standard deviation

ZJG Zhangjiagang City, Jiangsu Province, CS Changshu City, Jiangsu Province

[a] Comparison based on paired samples

[b] Comparison based on soil Cr prediction maps

Semivariograms/cross-semivariograms and spatial prediction comparison

Soil Cr in environment science is a regionalized variable as it is distributed in geographical space. It has spatial structures, including spatial autocorrelation. In this study, the geostatistics method was used for spatial structure analysis and visualization of interpretation results. The semivariogram/cross-semivariogram and the fitted models for Cr in different soil layers and different sub-areas are presented in Fig. 2. Semivariograms/cross-semivariograms showed that soil Cr in topsoil and subsoil of ZJG and CS were all fitted an exponential model. Figure 2 shows that the semivariograms/cross-semivariograms parameters (model type, nugget, sill and effective range) of Cr were different in different soil layers and different sub-areas under isotropic situations, although the best fitting model was the same.

Figure 3 presents the prediction maps of Cr concentrations. Figure 3 shows that the Cr in ZJG topsoil and CS subsoil had clear local spatial variability; however, the Cr in ZJG subsoil and CS topsoil had obvious whole spatial variability. The spatial map of Cr in ZJG subsoil showed a distribution trend with the high concentrations in the central area and low concentrations in the northwest area, and the map of Cr in CS topsoil showed a distribution trend with high concentrations in the southern area and low concentrations in northern area. The spatial maps of Cr concentration difference between in topsoil and in subsoil also showed some coarse distribution trends. Overall, the map of Cr concentration difference in ZJG had a distribution trend with positive difference in the northwest area and minus difference in the southeast area, and the map of Cr concentration difference in CS had a distribution trend with positive difference in central area and minus difference in other areas.

Discussion

The Cr concentrations in the majority soil samples in the study area were far lower than the pedogeochemical background value (90 mg kg^{-1}) of China. This indicates that the study area has a low Cr pedogeochemical background compared to other areas of China and the soil in the majority of the area was unaffected by Cr pollution. Although the study area had similar geographically and geologically conditions and all the samples collected from the same land use (agricultural land). The agricultural soils in both ZJG and CS had a wide range of Cr concentrations with a large spatial variability. It suggests that anthropic factor might be one of the major factors. To the areas with $>$90 mg/kg of Cr, further investigation is needed in order to define its possible source.

The results of the statistical analysis show that the mean concentration of Cr in ZJG was greater that in CS although the difference was small. This indicates that the Cr concentrations in the two regions (ZJG and CS) had significant differences and therefore it may be possible to predict their spatial distributions. The main industries in ZJG are the metallurgical industries, textiles, the chemical industry, machinery manufacturing and electronics, and in CS the garment industry, machinery manufacturing, the auto-parts industry, solar photovoltaics, and electronic information. Differences in the main industries may help to explain the differences in average soil Cr concentrations between ZJG and CS. The mean concentration of Cr in ZJG topsoil was greater than in ZJG subsoil but there was no significant difference between mean concentrations of Cr in CS topsoil and CS subsoil.

Semivariograms and their parameters show the spatial variability quantitatively. The nugget/sill ratio is assumed a criterion to classify the spatial dependence of soil properties. Ratio values lower than 25% and higher than 75% correspond to strong and weak spatial dependency, respectively, while ratio values between 25 and 75% correspond to moderate spatial dependence (Cambardella et al. 1994). The nugget/sill ratios of Cr in ZJG topsoil, Cr in ZJG subsoil and natural logarithm-transformed (lnCr) Cr in CS topsoil were in the 25–75% range and the nugget/sill ratio of lnCr in CS subsoil was lower than 25%. This indicates that the Cr in ZJG topsoil, ZJG subsoil and CS topsoil had moderate spatial dependence and the Cr in CS subsoil had strong spatial dependence. Range is also one of the parameters used to describe the spatial structure of data and its value is a measure of extension where autocorrelation exists (Li et al. 2007; Webster and Oliver 1990). The range values were: Cr in ZJG topsoil, 4560 m; Cr in ZJG subsoil, 10170 m; Cr in CS topsoil, 14520 m; and Cr in CS subsoil, 5640 m. Comparing the range of Cr at different soil depths, it was found that Cr in ZJG subsoil had longer

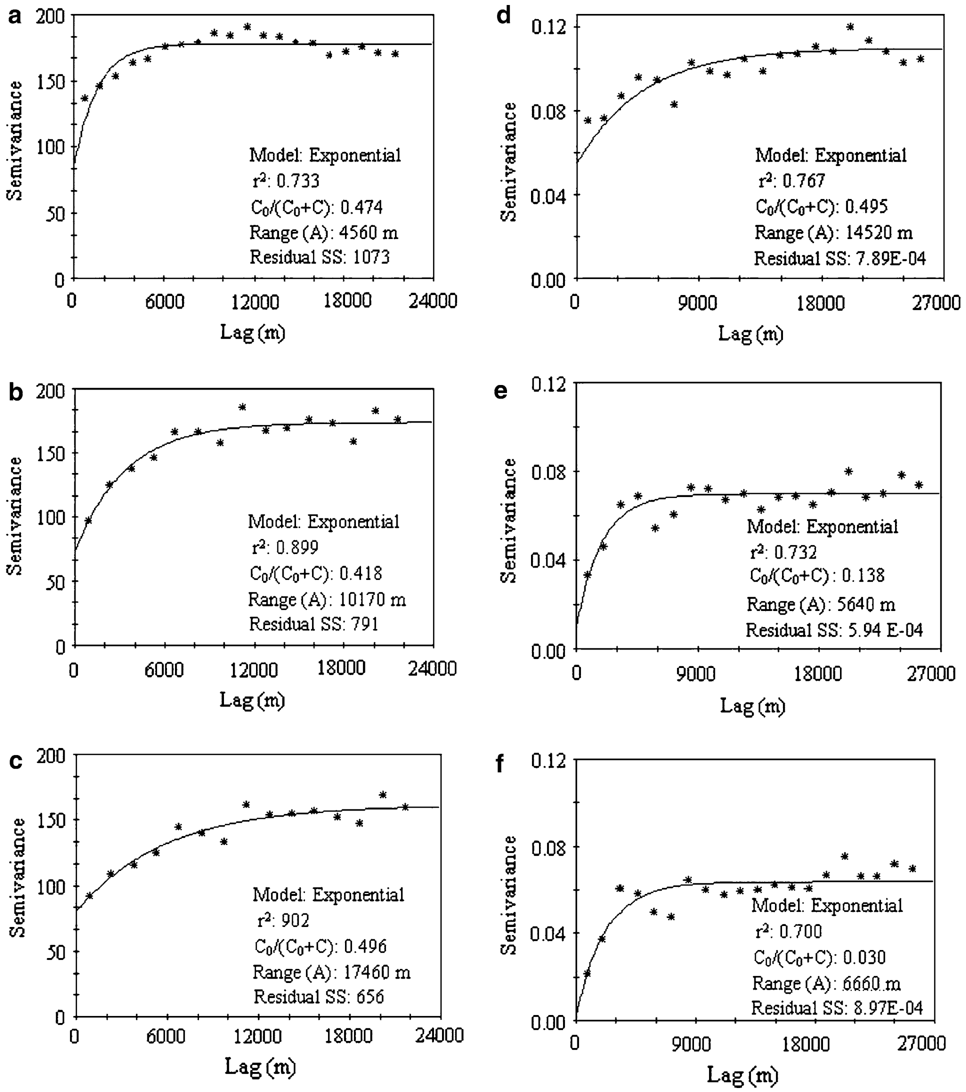

Fig. 2 Experimental semivariograms and cross-semivariograms of soil chromium (Cr) with fitted models (a and b are the semivariograms of Cr in topsoils and subsoils from Zhangjiagang City, respectively; c is the cross-semivariogram of Cr in subsoils from Zhangjiagang City; d and e are the semivariograms of natural logarithm-transformed Cr in topsoils and subsoils from Changshu City, respectively; f is the cross-semivariogram of natural logarithm-transformed Cr in subsoils from Changshu City)

effective range than Cr in ZJG topsoil, short-range spatial correlation of Cr in ZJG topsoil, suggesting that anthropogenic factors affect Cr distribution in ZJG topsoils. However, Cr in CS topsoil had longer effective range than Cr in CS subsoil. The soil formation of local areas may be the major explanation for the decrease in the variation of Cr in topsoils.

The Cr in ZJG subsoil had a distribution trend with high concentrations in the central area and low concentrations in the northwest area. The urban area was located near the centre of ZJG (Fig. 1), and high intensity of industrial activity and high-density factories in the urban area may be the main explanation for high concentrations Cr in ZJG topsoils. Generally the distribution of metals in soil is influenced by the nature of parent materials, climate and their relative mobility depending on soil parameters such as mineralogy, texture, and soil classification (Krishna and Govil 2007). The sand content of topsoils in CS had a clear spatial difference and the content decreased with increasing distance to the Yangtze River (detail location in Fig. 1). Williams et al. (1994) and Rae (1997) found that soil particle size plays a significant role in the accumulation of heavy metals and fine grained soils often show higher concentrations of heavy metals. This may be the main explanation for the Cr having a spatial distribution trend with high concentrations in the south and low concentrations in the north. To our knowledge, the subsoils in study area have been little influenced by anthropogenic activities and the Cr concentrations in subsoils were relatively low. Thus, natural factors might be the major determinants controlling subsoil element distribution in the study area. From Fig. 3, we can find that the distribution patterns of Cr in ZJG and CS topsoils were different from those in ZJG and CS subsoils, suggesting that Cr in topsoils and subsoils are controlled by different factors or combinations of factors and extrinsic factors play an important role controlling

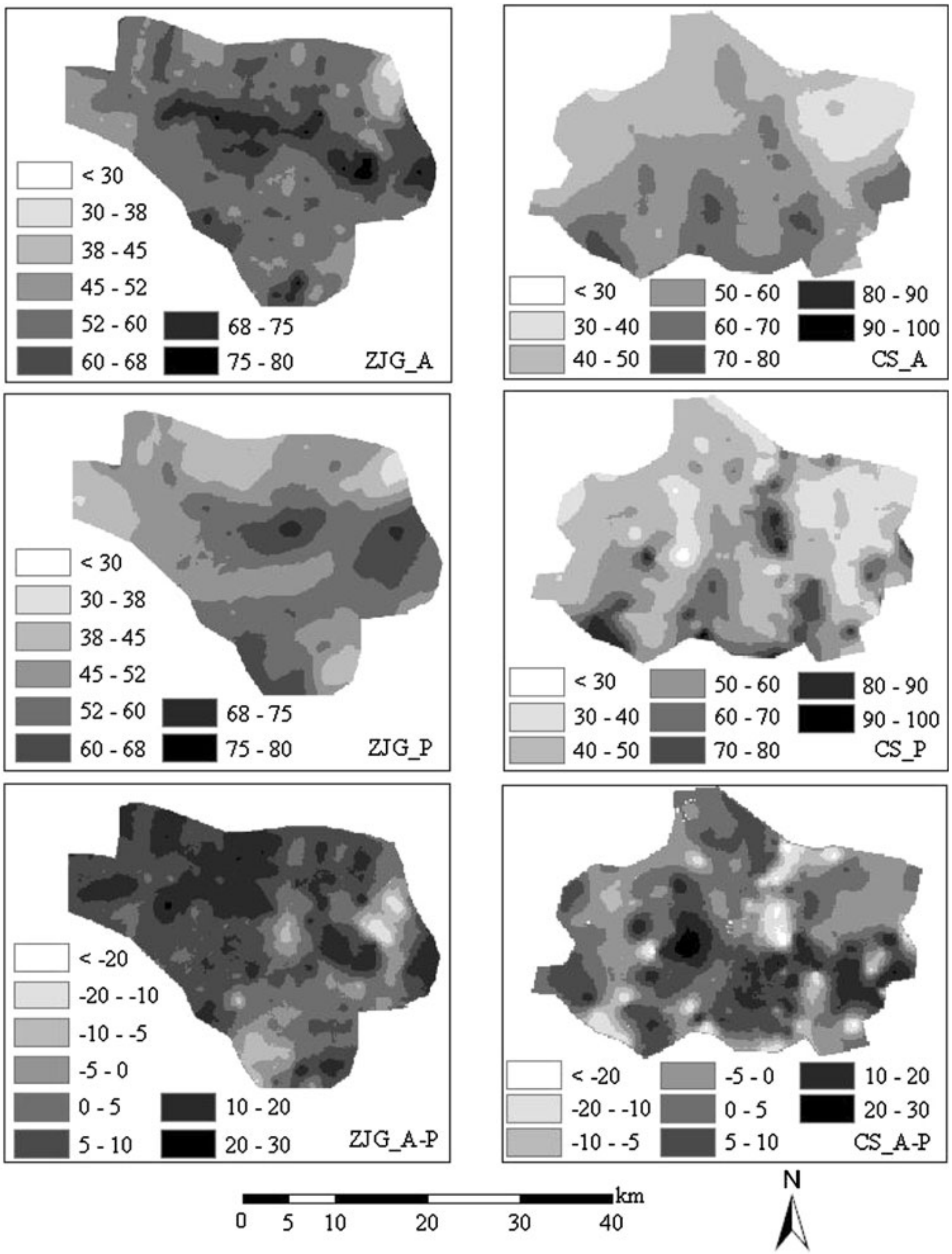

Fig. 3 Prediction maps of chromium (Cr) concentration (mg kg^{-1}) in topsoils, subsoils and the difference between them (ZJG_A and CS_A are the maps of Cr concentration in topsoils from Zhangjiagang City and Changshu City, respectively; ZJG_P and CS_P are the maps of Cr concentrations in subsoils from Zhangjiagang City and Changshu City, respectively; ZJG_A-P and CS_A-P are the maps of differences in Cr concentration between topsoils and subsoils from Zhangjiagang City and Changshu City, respectively)

the Cr in ZJG and CS topsoil distributions. A significant correlation was found between the Cr in topsoil and subsoil whether in ZJG or CS, indicating that natural factors also play an important role controlling the Cr in ZJG and CS topsoil distributions.

There was a wide range of differences in Cr concentration between ZJG topsoils and ZJG subsoils and between CS topsoils and CS subsoils, suggesting that both ZJG and CS topsoils were affected by extrinsic factors and the degree of influence was uneven. From the maps of difference in Cr concentration, it can be seen that the Cr concentrations in topsoil were higher than in subsoil over the majority of the area of ZJG and part of CS, indicating that Cr accumulation had occurred over most ZJG topsoils and some CS topsoils. Although the soil Cr concentrations over most of the study area were still low and within safety limits, they will increase rapidly and present a potential risk of Cr if action is not taken to reduce or stop Cr accumulation. Considerable attention must be paid to Cr accumulation in the study area and similar rapidly industrialized regions. To increase effective area of cultivated land and enhance land us efficiency, soil formation has been carried out in part of the study area in recent years. The maps of difference in Cr concentration show that Cr concentrations in topsoil were lower than that in subsoil in part of ZJG and about half of CS and this may be due to turnover processes in soil formation. The Cr concentrations in CS topsoils and CS subsoils were not significantly different.

Conclusions

The study area had a low Cr pedogeochemical background value. Over most of the area, the Cr concentrations in topsoils and subsoils were lower than the national mean pedogeochemical background value (90 mg kg^{-1}) in China. The soil was within safety limits for Cr. The Cr concentrations in the two regions (ZJG and CS) showed some significant differences. The mean concentrations of Cr in ZJG were significantly higher than in CS whether topsoil or subsoil although they were similar. The Cr concentrations in topsoils were higher than in subsoils in the northwest part of ZJG and in the central area of CS. Influence of extrinsic factors on Cr accumulation in topsoils existed over most of the area of ZJG and part of CS, leading to higher concentrations of Cr in topsoils than in subsoils over most of ZJG and part of CS.

Cr in ZJG topsoils, ZJG subsoils, and CS topsoils showed moderate spatial dependence and the Cr in CS subsoils had a strong spatial dependence range. Cr in ZJG subsoils had longer effective range than Cr in ZJG topsoils, suggesting that anthropogenic factors had affected Cr distribution in ZJG topsoils. Cr in CS topsoils had longer effective range than Cr in CS subsoils, indicating that soil formation might be the main explanation for the decreasing variation in Cr in topsoils. Cr in topsoils was strong correlated with Cr in subsoils in both topsoils and subsoils, indicating that natural factors also played an important role as extrinsic factors controlling the distribution of Cr in topsoils.

Most of the areas of ZJG and part of CS showed Cr accumulation in topsoil, and the spatial distribution of Cr in topsoil is obviously in relation to local industrial activities. It implied that Cr accumulation in topsoils would be a potential environmental problem in rapidly industrialized regions such as the Yangtze Delta, the Pearl Delta in China and other areas in developing countries. Input of anthropogenic Cr into the soil should be controlled and mitigated urgently in order to decrease the risk of soil Cr contamination.

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