

Spatial analysis and hazard assessment of mercury in soil around the coal-fired power plant: a case study from the city of Baoji, China

Xiaopeng Yang · Lingqing Wang

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Abstract Based on systematic sampling of soil around the coal-fired power plant (CFPP), the content of Hg was determined, using atomic fluorescence spectrometry. The result shows that the content of Hg in soil is different horizontally and vertically, ranges from 0.137 to 2.105 mg/kg (the average value is 0.606 mg/kg) and is more than the average content of Hg in Shaanxi, Chinese and world soil. In this study, spatial distribution and hazard assessment of mercury in soils around a CFPP were investigated using statistics, geostatistics and geographic information system (GIS) techniques. Ordinary kriging was carried out to map the spatial patterns of mercury and disjunctive kriging was used to quantify the probability of the Hg concentration higher than the threshold. The maps show that the spatial variability of the Hg concentration in soils was apparent. These results of this study could provide valuable information for risk assessment of environmental Hg pollution and decision support.

Keywords Coal-fired plant · Soil · Mercury · Spatial analysis · Hazard assessment · Geostatistics

Introduction

Mercury (Hg) is a highly bio-accumulated toxic metal and even small releases of Hg can lead to long-term and severe

environmental and health consequences (Sell et al. 1975; Simpson et al. 1997). Recently, many researches have provided consistent evidence of noticeable increases in environmental Hg levels compared to that of the natural background (Schmolke et al. 1999; Liu et al. 2002, 2003; David et al. 2005). Mercury can be released and mobilized through both natural processes and anthropogenic activities. It has been estimated that anthropogenic emission of Hg is about 60–80% of the global Hg emission (Dvonch et al. 1999). Anthropogenic sources of incidental Hg include stationary combustion of fossil fuels, non-ferrous metal production, pig iron and steel production, cement production and waste disposal primarily from incineration (Pacyna et al. 2003). Among them, coal combustion, because of the huge amount of coal consumed, accounts for a significant portion of the anthropogenic releases of Hg into the environment.

Coal is the main natural resource and fossil fuel available in abundance in China and it is used widely as fuel for thermal power plants producing electricity. The average Hg content in Chinese coals was 0.22 mg/kg (Wang et al. 1999). According to statistical data from NBS (2005), coal-fired power plants (CFPP) consumed 1,637 million tons of raw coal in China in 2003, a 55.2% increase from 1990. David et al. (2005) estimated that China's emissions from anthropogenic sources were 536 (± 236) t of total Hg in 1999 and approximately 38% of the Hg comes from coal combustion. Therefore, exposure of the population around the CFPP to Hg may occur due to inhalation of Hg present in air and consumption of Hg contaminated food and water. Hg pollution around the CFPP is an active area of current research in Europe and North America (Schmolke et al. 1999; Laura et al. 2004; Tan et al. 2004; Carlos et al. 2006). Comparatively, there is little research regarding this in China.

Influenced by the CFPP, the distribution properties of the Hg in soils, even in short distances differ from point to point.

X. Yang
Department of Mathematics, Physics and Engineering,
Shaanxi Institute of Education, Xi'an 710061, China

L. Wang (✉)
College of Tourism and Environment,
Shaanxi Normal University, Xi'an 710062, China
e-mail: lingqing054@stu.snnu.edu.cn

Understanding the spatial distribution of Hg in soil is critical for environmental management and decision-making. The spatial variability of the Hg in soils around the CFPP may be described by geostatistics. Geostatistics has been proved useful in characterizing spatial variability and mapping a variety of soil properties (White et al. 1997; Romić and Romić 2003; McGraph et al. 2004; Goovaerts 1999, 2001).

In this study, soil samples were collected around the Baoji CFPP and the Hg concentration in samples has been estimated using AFS-810 atomic fluorescence spectrometry. The aim of this study was to evaluate the situation of mercury pollution, analyze the spatial distribution of mercury in soil and quantify their hazard assessment around the Baoji CFPP.

Materials and methods

Study area

Baoji is the second largest city in Shaanxi province in central China, which has the latitude of 33°34′ to 35°6′N and the longitude of 106°18′ to 108°3′E. It is located at the western end of the Guanzhong (Weihe) valley about 150 km west from the provincial capital city Xi'an. Baoji is surrounded by mountains and plateau in the north, the west, and the south. Only the east is open toward the lower reach of the Weihe River—a major branch of the Yellow River in Shaanxi province. The north of the city is bounded by the south edge of the loess land plateau over 800 m above sea level. The Weihe River runs through the city from west to east. The research area has a typical continental monsoon climate: hot and rainy summers, and cold and dry winters, the annual temperature is about 10°C and the annual precipitation is about 600–700 mm. The main soil type is cinnamon soil and the main crops are wheat and vegetables in the study area. There are about 550,000 people living in Baoji city.

The CFPP with a 60 m stack, as studied in this work, is located in southwest 4 km away from Baoji urban district (Fig. 1), and has been operational since 1960s. The Baoji power plant with 1.5×10^6 kWh annual production capacity consumes low quality bituminous coal reserves from Tongchuan of Shaanxi and Huating of Gansu, and produces approximately 4,500 tons (t) of fly and bottom ash per day from more than 14,000 t of coal. The filter system (filtering bag) was installed in 2001 to reduce the particulate emission through the 60 m stack.

Sample collection and analytical methods

Soil samples were collected around the CFPP from 20 locations as marked on the map in Fig. 1. Thirty-two soil

samples were collected around the CFPP, at a distance of 1 and 3 km, in the principal compass directions. Eight soil samples were collected within the 1 km distance of the CFPP. Topsoil (depth from 0 to 25 cm) and subsoil (depth from 25 to 50 cm) were both collected from every sampling site, named horizon A and horizon B, respectively. All the samples were collected with a stainless steel spatula and kept in PVC packages.

The soil samples were dried in a shady and aerated place at room temperature until constant weight. The impurities such as pebbles, roots and tree leaves were taken out from these samples, and then all samples were crushed and purified until all of them passed through a 0.075 mm nylon sieve. Parameters such as pH and organic matter were determined for general characterization of the soil samples. The pH was determined by potential method and organic matter was determined by High TOC II analyzer (produced by Elementar Analysensysteme GmbH).

Mercury in the soil samples was determined in dry weight by AFS-810 double channel atomic fluorescence spectrometry with hollow cathode lamp of mercury (both of these instruments are produced by Beijing Titan Instrument Corporation) and high purity argon gas (Cai 2005). The operating parameters of AFS-810 double channel atomic fluorescence spectrometry were as follows: electrical current is 15 mA, negative high pressure is 270 V, and airflow is about 600 ml per minute. The analysis process is shown as follows: weigh a sample of 0.2000–0.3000 g and put it into a 50 ml volumetric flask with cover, add 10 ml freshly prepared aqua regia (1 ml high concentration HNO₃:3 ml high concentration HCl), put the cover in the bottle and mix them up, then place the volumetric flask removing away its cover in a boiler with boiling water for 2 h, shaking every 30 min. After cooling, add 5 ml of 5% thiocarbamide-5% ascorbic acid solution and dilute it to 50 ml with redistilled water. Mercury was determined in the 5% HCl with 20 g/L sodium borohydride (NaBH₄) as the reducing agent, at the same time the blank and the standard substances were determined too. The standard substances (geochemical standard reference sample soil in China, GSS-1) were used to examine the precision and accuracy of determination and the results were satisfying.

Geostatistical methods

The theoretical basis of geostatistics has been described by several authors (Matheron 1963; Davis 1987; Isaaks and Srivastava 1989; Olea 1999). The main tool in geostatistics is the variogram, which expresses the spatial dependence between neighboring observations. The variogram can be defined as half the average squared difference between pairs of data values separated by the direction vector

Fig. 1 Location of the coal-fired plant and the sampling site



(where anisotropy is considered) and lag h (the distance separating two experimental points):

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} \{Z(x_i) - Z(x_i + h)\}^2 \quad (1)$$

where $\gamma(h)$ is the semi-variogram, $Z(x_i)$ is the value of the measured variable at location of x_i , $N(h)$ is the number of pairs of sample points separated by the distance h , and x is the position of samples.

The experimental semi-variogram is commonly calculated for a number of directions and for a number of lag values along each direction. Then a suitable theoretical model is fitted, the most commonly used models are spherical, exponential, Gaussian, and pure nugget effect (Isaaks and Srivastava 1989). Weights for sample values are calculated based on the parameters of the model. The nugget effect is the tendency, presents the experimental errors that occur in measurement and/or micro scale variation (variation at spatial scales too fine to detect). The sill depicts the maximum variance. The range is which the lag distance increasing at to approach the sill. The direction describes in certain directions closer things may be more alike than in other directions because of the wind, runoff, a geological structure, or a wide variety of other processes.

Ordinary kriging was used in this study to create the final prediction map of Hg concentration. Cross validation was used to compare the prediction performances of these geostatistical interpolation algorithms (Isaaks and Srivastava 1989). Cross validation indicators and additional model parameters helped to choose the most appropriate model. Kriging cross-validation was used to choose the best

semivariogram models, which could give the most accurate predictions of the unknown values of the field. For a model that provides accurate predictions, the mean error (ME) should be close to 0; the root mean square error (RMSE) and average standard error (ASE) should be as close as possible. The root mean square standardized error (RMSSE) signified that the prediction values were closer to measured values (Wackernagel 1995). If the predictions are close to the measured values the root mean square standardized error should be close to 1 and the root-mean-square prediction error should be small. In this study, the fitted Spherical model and exponential model were selected.

The spherical function is:

$$\gamma(h) = \begin{cases} c_0 + c \left(\frac{3h}{2a} - \frac{h^3}{2a^3} \right), & 0 \leq h < a \\ c_0 + c, & h \geq a \end{cases} \quad (2)$$

The exponential function is:

$$\gamma(h) = c_0 + c \left(1 - \exp^{-\left(\frac{h}{a}\right)} \right), \quad h \geq 0 \quad (3)$$

Where $\gamma(h)$ is the semivariance for the interval distance class h , C_0 is the nugget variance ($h = 0$), C is the structural variance, a is the spatial range across which the data exhibit spatial correlation and $C_0 + C$ is the sill variance.

Disjunctive kriging is the principal technique to estimate the probability that the true values of Hg at an unsampled location exceed a specified threshold. It is based on the assumption that our data are a realization of a process with a second-order stationary bivariate distribution. The assumption of second-order stationarity means that the

covariance function exists and that the variogram is therefore bounded. It is assumed that the Hg concentration is a realization of a random variable $Z(x)$, where x denotes the spatial coordinates in two dimensions. If a threshold concentration z_c is defined, marking the limit of what is acceptable, then the scale is dissected into two classes which is less and more than z_c , respectively. The soil must belong to either class at any one place. The value 0 and 1, respectively, can be assigned to two classes, thereby creating a new binary variable, or indicator, which is denoted by $Q[Z(x) \geq z_c]$ (Steiger et al. 1996; Lark and Ferguson 2004).

Software resources

The data analysis was carried out with different software packages. SPSS (version 13.0) was used to transform the data and calculate the descriptive statistical parameters. The geostatistical analysis was carried out with the extension Geostatistical Analyst of the GIS software ArcGIS (version 9.0). A global positioning system (GPS) recorded the samples' location and all the maps were produced with Arcinfo and Arcmap (version 9.0).

Results and discussion

Descriptive statistics

The pH of the soil samples around the Baoji CFPP is between 8.1 and 8.4, and the organic matter ranges from 1.06 to 2.52% with an average is about 1.85%. Table 1 gives the summary statistics of the data sets for Hg concentration in the soil samples around the Baoji CFPP. The Hg concen-

tration ranges from 0.137 to 2.105 mg/kg with the mean value is 0.606 mg/kg and mercury concentrations in majority of these samples are higher compared to worldwide uncontaminated soils which ranged from 0.01 to 0.5 mg/kg in total Hg (Senesi et al. 1999). It is shown that the average values of mercury concentration in horizon A and B both exceed that in Shaanxi (0.101 mg/kg), Chinese (0.042 mg/kg) and world soil (0.030 mg/kg) (State Environmental Protection Administration of China 1993). The average value of Hg concentration in soils is lower than the Canadian Soil Quality Guideline for agricultural soils (6.6 mg/kg) (Appleton et al. 2006), the proposed Dutch Intervention value (SRC-serious risk concentration) for inorganic Hg (36 mg/kg; Appleton et al. 2006) and the USEPA soil ingestion soil screening level (SSL) for inorganic Hg (23 mg/kg) (USEPA 1996). The maximum permissible concentration of Hg in agricultural soil in the UK (Appleton et al. 2006) and China (State Environmental Protection Administration of China 1995) was 1.0 mg/kg and it would contribute to the environmental pollution and ultimately threaten the health of plant and human when Hg concentration in soils exceeds the value of 1 mg/kg. In this study, Hg concentrations in some samples collected <1 km from the CFPP were high than this value.

Mercury concentrations in the soil reported in the literature are also listed in Table 1. In comparison with other cities in China, mercury concentration in the soil samples around the Baoji CFPP is much higher than that in Changchun (0.139–0.479 mg/kg) (Fang et al. 2004), Xuzhou (0.02–1.3 mg/kg) (Wang and Qin 2005) and Beijing (0.01–0.966 mg/kg) (Zhang et al. 2006). Contrasting with other cities in the world, mercury concentration in the soil samples around the Baoji CFPP is higher than that of Pittsburg, USA (0.51 mg/kg) (Carey et al. 1980), Canada

Table 1 Summary statistics of Hg concentration in the study areas and mercury concentration reported in literatures (mg/kg)

Soil type/land use	Location	Mean	Median	Min	Max	SD	Kurtosis	Skewness	K-S p
Agricultural soil	Horizon A	0.692	0.658	0.197	2.105	0.429	5.603	2.001	0.303
Agricultural soil	Horizon B	0.533	0.539	0.137	1.066	0.272	-0.270	0.525	0.544
	Ln (Hg in Horizon A)						0.068	0.292	0.932
Urban topsoil	Beijing, China	0.278	0.185	0.01	0.966	(Zhang et al. 2006)			
Urban soil	Changchun, China			0.139	0.479	(Fang et al. 2004)			
Urban topsoil	Xuzhou, China		0.18	0.02	1.3	(Wang and Qin 2005)			
Agricultural soil	Canada		0.04	0.005	0.13	(Reimann and Caritat 1998)			
Urban soil	Pittsburg, USA	0.51				(Carey et al. 1980)			
Typical normal range in soil	UK			0.008	0.19	(Thornton 1991)			
Urban soil, town gardens	Wexford, Ireland	0.68		0.09	2.97	(McGrath 1995)			
Urban soil	Palermo, Italy		0.68	0.04	6.96	(Manta et al. 2002)			
Urban soil	Trondheim, Norway		0.13	<0.2	4.49	(Reimann and Caritat 1998)			
Urban soils, industrial centre	Cornwall, Canada	0.698		0.04	5.1	(Sherbin 1979)			

(0.005–0.13 mg/kg) (Reimann and Caritat 1998), and UK (0.008–0.19 mg/kg)(Thornton 1991), at the same time, mercury concentration in the soil samples around the Baoji CFPP is lower than that of Wexford, Ireland (0.09–2.97 mg/kg) (McGrath 1995), and Palermo, Italy (0.04–6.96 mg/kg) (Manta et al. 2002), and Trondheim, Norway (<0.2–4.49 mg/kg) (Reimann and Caritat 1998), and Cornwall, Canada (0.04–5.1 mg/kg) (Sherbin 1979).

A box plot graphic representation was applied to evaluate the distribution of Hg in soils around the coal-fired power plant (Fig. 2). This methodology allows a visualization of the Hg concentration among different sampling locations, the range of data variation, average and median concentrations. The result presented in Fig. 2 showed that the concentrations of Hg in horizon A were significantly greater than those in horizon B, and the highest concentration was found in location <1 km from the CFPP. It can also be seen in Fig. 2 that the average concentrations of Hg did not decrease with the distance from the discharge point for samples collected at 1 and 3 km, contrarily higher average values were observed in the samples collected at 3 km. This may be caused by intrinsic factors (soil formation factors, such as soil parent materials) and extrinsic factors (climate factors such as prevailing wind and human activity such as cultivation). It is reported that the stack height of the CFPP also affected the diffusion of Hg (Beck and Miller 1980).

As in conventional statistics, a normal distribution for the variable under study is desirable in linear geostatistics (Clark and Harper 2000). The parameters of skewness, kurtosis, and the significance level of Kolmogorov–Smirnov test for normality (K–S p) are shown in Table 1. It is

shown in Table 1 that the skewness and kurtosis values for Hg concentrations in horizon A were high and these raw data were not normally distributed. After these raw data sets in horizon A were logarithmically transformed the kurtosis and skewness decreased and the K–S p value was higher. Then they all pass the normality test.

The histograms of Hg concentration with a normal distribution curve of both the raw and the logarithmically transformed data are shown in Fig. 3. It is clear that the statistical distribution of the raw data of Hg concentration in horizon B and the log transformed data of Hg concentration in horizon A are near normal.

Geostatistical analysis

Table 2 shows the semivariogram model and best-fit model parameters. Spherical and Exponential models were chosen as appropriate for all direct-semivariograms after visual inspection of sample semivariograms and cross-validation check. It shows that Hg concentration in horizon A is fitted for an Exponential model; Hg in horizon B is best fitted for Spherical model. The Nugget/Sill ratio of Hg concentration in horizon A is less than 25%, suggesting the variable has strong spatial dependence. The ratio of Hg concentration in horizon B are between 25 and 75%, suggesting the variable has moderate spatial dependence, which showed that the extrinsic factors such as fertilization, ploughing and other human activities weakened their spatial correlation after a long history of cultivation.

Spatial distribution and hazard assessment

Figure 4 is the prediction map of the Hg concentration. It shows the spatial variation of Hg concentration in soils around the CFPP generated from their semivariograms. From the Fig. 4, it can be seen that the Hg concentration has distinct geographical distribution. The spatial variation map for Hg concentration in horizon A shows that Hg concentration is higher in the vicinity of the CFPP and lowers with the distance from the CFPP. It also shows a geographical trend with high concentrations in NW–SE direction, which might be caused by the prevailing wind. The map for Hg concentration in horizon B clearly presents a strong gradient concentration in the northwestern area.

The estimated probability of exceeding threshold was kriged by disjunctive kriging and given in Fig. 5. The maps show the probability of its value exceeding the defined threshold. The guide value of Hg (1 mg/kg) in soil was decided to serve as the threshold in this study. Like observed in Fig. 5, the highest probability of exceeding this threshold occurs in the vicinity of the CFPP. This character of spatial distribution confirms the directional features revealed by the prediction maps.

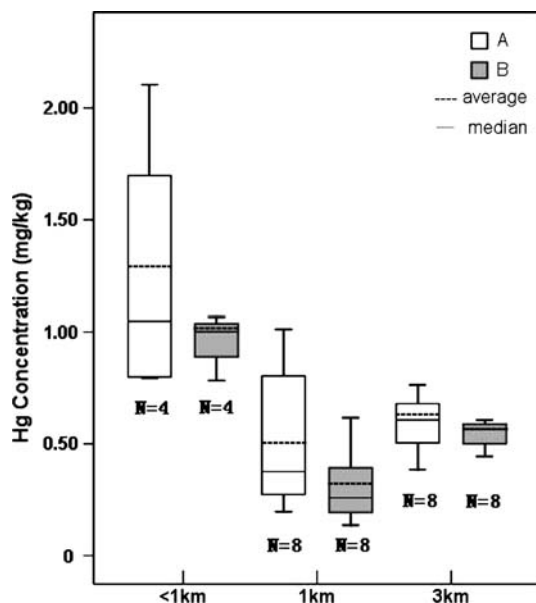


Fig. 2 “Box plot” graphic representation of Hg concentration in soil samples around the CFPP

Fig. 3 The histograms of Hg concentration with a normal distribution curve

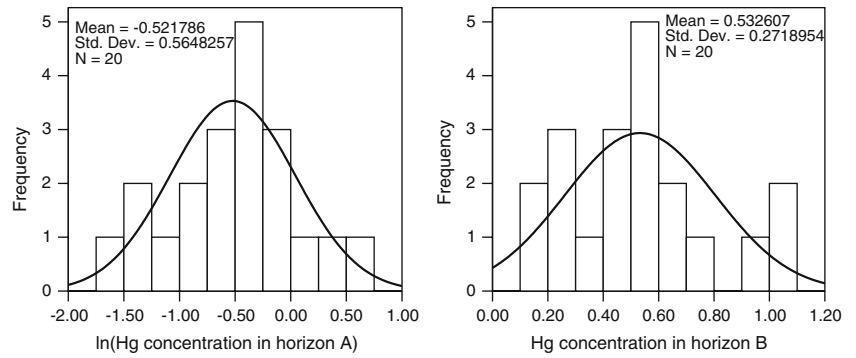


Table 2 Best-fitted semivariogram models of Hg concentration and their parameters

Profile	Model	Nugget	Sill	Range	Direction	Nugget/Sill	ME	RMSE	ASE	RMSSE
A	Exponential	0.002	0.321	321.4	423.43	0.005	-0.029	0.275	0.274	1.013
B	Spherical	0.062	0.234	283.8	508.57	0.265	0.031	0.470	0.490	1.026

Fig. 4 The prediction maps of Hg concentration in soils

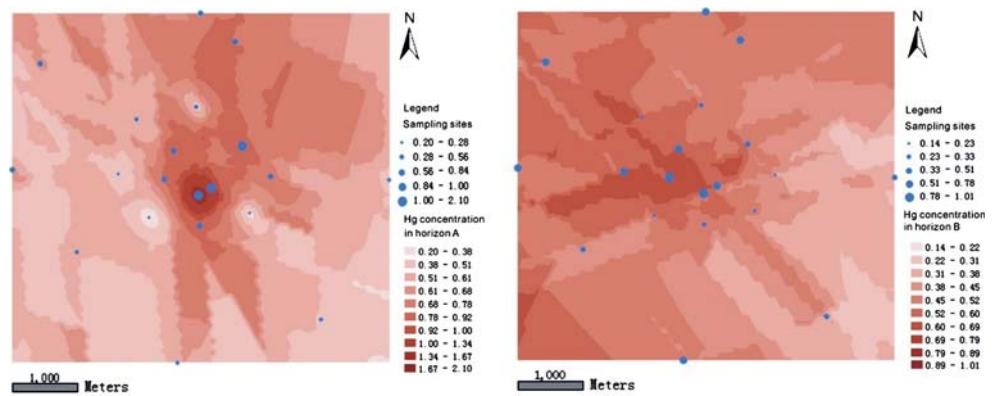
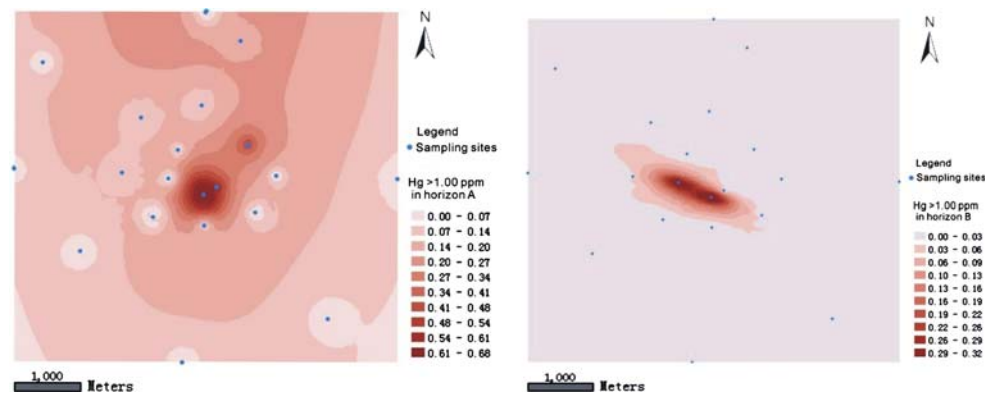


Fig. 5 The estimated probability maps of Hg



Conclusions

About forty-five years of operating the CFPP, as studied in this work, caused an increase in the Hg concentrations in

soils around it. The Hg concentration ranges from 0.137 to 2.105 mg/kg with the mean value is 0.606 mg/kg. The average value of Hg concentration in this study area exceeds the maximum permissible concentration of Hg in

agricultural soil in the UK and China, although it is less than the Canadian Soil Quality Guideline for agricultural soils, the proposed Dutch Intervention value and the USEPA soil ingestion soil screening level. It is shown that the average values of mercury concentration in horizon A and B both exceed that in Shaanxi, Chinese and world soil. Contrasting with other mercury concentration studies mercury pollution in soil around the Baoji CFPP is serious. It is shown that the spatial variability of the Hg concentration in soils were apparent in this study. That may be affected by several factors, such as soil formation factors, prevail wind and human activity. All the obtained data can be used as a valuable database for future estimations of the impact of Hg pollution around the CFPP and for the developmental prospects of the region.

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