Nat. Hazards Earth Syst. Sci. Discuss., 2, 1977–2010, 2014
www.nat-hazards-earth-syst-sci-discuss.net/2/1977/2014/
doi:10.5194/nhessd-2-1977-2014

Natural Hazards and Earth System Sciences
Open Access
Discussions

Potential ecological risk assessment and prediction of soil heavy metal pollution around coal gangue dump

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Received: 14 June 2013 – Accepted: 13 February 2014 – Published: 7 March 2014

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Aim of the present study is to evaluate the potential ecological risk and predict the trend of soil heavy metal pollution around a coal gangue dump in Jilin Province (Northeast China). The concentrations of Cd, Pb, Cu, Cr and Zn were monitored by inductively coupled plasma mass spectrometry (ICP-MS). The potential ecological risk index method developed by Hakanson (1980) was employed to assess the potential risk of heavy metal pollution. The potential ecological risk in an order of E(Cd) > E(Pb) > E(Cu) > E(Cr) > E(Zn) have been obtained, which showed that Cd was the most important factor led to risk. Based on the Cd pollution history, the cumulative acceleration and cumulative rate of Cd were estimated, and the fixed number of years exceeding standard prediction model was established, which was used to predict the pollution trend of Cd under the accelerated accumulation mode and the uniform mode. Pearson correlation analysis and correspondence analysis are employed to identify the sources of heavy metal, and the relationship between sampling points and variables. These findings provide some useful insights for making appropriate management strategies to prevent and decrease heavy metal pollution around coal gangue dump in Yangcaogou coal mine and other similar areas elsewhere.

1 Introduction

Coal is the main fossil fuel in China and it provides more than 70 % of total energy. The considerable growth of the mining industry in China over the last decades has resulted in a large number of mining wastes accumulated in wastelands. These wastes in addition to their land use impact cause water and soil pollution as well as soil erosion and other environmental problems (Querol et al., 2008). Mining industry generated 265 Mt tailings, 130 Mt gangue, and 107 Mt smelting slag in 2002. Among which, the most concern is coal gangue, as the direct outputs of coal mining, has substantially generated and accumulated around coal mine area. Currently, the cumulative amount of coal

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gangue in China was estimated to be more than 5.0 billion tons given the large coal production. Despite of its economic and social benefit, the generation and accumulation of coal gangue has brought a variety of environmental and social problems. Among which, heavy metal pollution is of the most concerned interests (Gong et al., 2008). Exposed to the weathering, leaching and decomposing, water containing heavy metal element will be emitted into the soil. Heavy metal pollution in soils around coal gangue dump is a important threat to human health because it can be easily transferred into human bodies through ways of ingestion via the hand-mouth pathway, inhalation and dermal contact. Moreover, the long-term input of heavy metal elements could result in decreased buffering capacity of soil, threatening the ecological environment and even having potential risk to human health. In order to understand the potential ecological risk degree of heavy metals in soil, the synergy and toxic level of the heavy metals, concentration of pollution and ecological sensitivity, it is particularly significant to evaluate the potential ecological risk and predict the trend of soil heavy metal pollution around coal gangue dump for carrying out a targeted treatment and prevention for soil heavy metal pollution around the coal gangue dump.

In recent years, the heavy metal pollution of the coal gangue dump caused by the considerable growth of the mining industry and the effects of human activity has called for extensive concern worldwide. Many researchers have performed related researches on mineral composition in coal gangue (Zhou et al., 2012; Fernandes, 1997), chemical speciation of heavy metals in coal gangue (Dang et al., 2002), pollution characteristics of heavy metal in soil (Dragović et al., 2008; Luan et al., 2008), potential ecological risk assessment of soil heavy metals contamination around coal gangue piles (Abdu et al., 2011; Krishna, and Govil, 2008; Zhao et al., 2012). However, the development trend of soil heavy metal pollution around coal gangue dump has received less formal attention than it should be.

Yangcaogou coal mine is a large state-owned coal mine in Changchun, North East China. Since the coal mine put into production, the average annual emission of coal gangue is about 20 300 m^3, and the coal gangue mainly are plied within the indus-

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trial square. Without taking any protection and treatment measures, soil around coal gangue dump have been polluted in a certain degree, but few studies could be found in the literature about this area. This work presents a study on the potential ecological risk of soil heavy metal pollution using Potential Ecological Risk Index with Yangcaogou coal mine as the study objective. The sampling points are designed based on the site-specific characteristics (wind direction and slope direction), and fixed number of years exceeding standard prediction model was applied to predict the development trend of Cd in soil (the most serious pollution heavy metal element) pollution around coal gangue dump in the future. These results provide useful insights for seeking appropriate management strategies to prevent and decrease soil heavy metals contamination around coal gangue dump in Yangcaogou coal mine and other similar areas.

2 Materials and methods

2.1 Study area

The Yangcaogou coal mine is located at east part of Changchun city, Jilin Province, China. It lies between latitudes 43°35′59 N and 43°56′44 N, and longitudes 125°34′58 E and 125°38′20 E, with an area of over 16 km^2 (Fig. 1). It is a major coal production base in Jilin Province with over 20 yr of mining history. The climate in the study area is characterized with continental monsoon climate. The annual average temperature of the region is 4.6 °C and the average annual rainfall is 569.6 mm. The geology of this region consists of quaternary deposits and the natural soil types, black soil, dark brown soil and meadow soil are the main geological formations according to the classification and codes for Chinese soil. The land contains cultivated land, planting corn and soybean. The prevailing wind direction is from the southwest to northeast throughout the year, and there is an annual average wind speed of 5 ~ 6 $m s^{-1}$, and the maximum wind speed is 30 $m s^{-1}$.

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2.2 Sampling design and analysis procedure

2.2.1 Sampling

There are several coal mine dumps in this area. In order to evaluate heavy metal pollution in soil from coal gangue dump, one 15 yr of history coal gangue dump in Yangcaogou Coal Mine was selected for this study. This coal gangue dump is a cone with a diameter of 40 m and a height of 15 m.

Generally, the heavy metals in coal gangue are emitted into the soil environment through two ways: (1) coal gangue dust by wind erosion suspended in the atmosphere land in soil around coal gangue dump; (2), the contaminants formed by the atmospheric precipitation eluviations move into the soil with surface runoff and groundwater. The present study does not consider the role of groundwater because the coal gangue dump is located in the groundwater drainage area.

The prevailing wind direction is from the southwest to northeast throughout the year, and the slope direction is towards north-east. Considering the wind direction and the local slope direction, along the center of the coal gangue dump, three sampling lines (L1, L2 and L3) were designed, and each sample with a mass of approximately 1.0 kg. The different depths (0.15 m, 0.4 m, 0.8 m, 1.2 m, 1.6 m, 2.0 m, 2.5 m, 3.0 m) of coal gangue and soil samples were collected through drilling holes. The drilling (sampling points) location distribution was shown in Fig. 1. One group (0.15 m, 0.4 m, 0.8 m, 1.2 m, 1.6 m, 2.0 m, 2.5 m, 3.0 m) of back ground samples (BK01) were taken at soil surface about 600 m from the coal gangue dump. The detailed location of each sampling point can be seen in Table 1.

Coal gangue is a heterogeneous material, and it is a complex and difficult task to take the samples. In order to make the gangue samples to be representative, five samples from four different depths of the coal gangue dump were collected, making the average of heavy metal elements concentrations at different depths as the heavy metal elements concentrations of coal gangue. The coordinates of soil and coal gangue sampling locations were recorded with a GPS receiver, and environmental observations were described during the fieldwork.

2.2.2 Detection and analysis methods

The soil and coal gangue samples were dried and grinded in laboratory, and then were sieved using a 20 meshes sieve for pH measurement experiment and 200 meshes for heavy metals measurement experiment, respectively.

All analysis were completed in Experimental Center Testing Science of Jilin University, which is a research institute passed the national metrology authentication of China. The prepared soil and coal gangue samples were passed through a 200-mesh sieve. For each sample, approximately 0.1 g of soil was digested using 3 mL HNO_3 (68 %, GR), 1 mL HF (40 %, GR), and 1 mL H_2O_2 (30 %, GR), for measuring the heavy metals concentrations. The concentrations of Cd, Pb, Cu, Cr and Zn were determined by inductively coupled plasma mass spectrometry (ICP-MS, Aglient7500a, USA).

For each sample, approximately 10 g of 20-mesh sieved soil sample was weighted using electronic balance for measuring the pH of the soil or coal gangue. Values of pH were measured in 25 mL of deionized water (DI water) with a soil/solution ratio of 1 : 2.5 ($m\,v^{-1}$), using the Glass Electrode method (GL, pHS-3C, REX, Shanghai, China) according to the agricultural sector standard (NY/T 1377-2007) of People's Republic China (Eidukeviciene et al., 2010; Ščančar et al., 2000; Kim et al., 2012; Yang et al., 2008).

Descriptive statistics, including the range, mean, standard deviation (SD) and coefficient of variation (CV) were performed. The SD and CV were incorporated to represent the degree of dispersion distribution of different heavy metals and to indirectly indicate the activity of the selected elements in the examined environment (Han et al., 2006).

Pearson correlation coefficients were calculated to determine the relationships among the different heavy metals and the pH values in the soil samplings around coal gangue dump.

2.3 Methods of heavy metal pollution assessment

To assess the degree of metal contamination, a revised pollution index (PI) for each metal and a Nemerow integrated pollution index (NIPI) of the five metals were attributed to each sampling site. The PI was defined as follows:

$$\mathrm{PI} = C_i / S_i \quad (1)$$

where C_i is the measured concentration of each metal (Cd, Pb, Cu, Cr, Zn) in this study, S_i is the background value in this paper, The PI of each metal was classified as: non-pollution (PI < 1), low level of pollution (1 ≤ PI < 2); moderate level of pollution (2 ≤ PI < 3); strong level of pollution (3 ≤ PI < 5) and very strong level of pollution (PI ≥ 5). The NIPI of the five metals for each sampling site was defined as follows:

$$\mathrm{NIPI} = \sqrt{\frac{\mathrm{PI}^2_{i\mathrm{ave}} + \mathrm{PI}^2_{i\max}}{2}} \quad (2)$$

Where $\mathrm{PI}^2_{i\max}$ is the maximum PI value of each heavy metal and $\mathrm{PI}^2_{i\mathrm{ave}}$ is the average PI value of each heavy metal. The NIPI was classified as: non-pollution (NIPI ≤ 0.7); warning line of pollution (0.7 < NIPI ≤ 1); low level of pollution (1 < NIPI ≤ 2); moderate level of pollution (2 < NIPI ≤ 3) and high level of pollution (NIPI > 3) (Yang et al., 2010). The calculated results are summarized in results section at the end of this paper.

2.4 Methods of potential ecological risk assessment

This research employed the Potential Ecological Risk Index (PERI) proposed by Hakanson (1980). PERI is a diagnostic tool for contamination control of lakes and coastal system; it was originally developed for Scandinavian environments. PERI is formed by three basic modules: degree of contamination (CD); toxic-response factor (TR); and potential ecological risk factor (ER).

This method comprehensively considers the synergy, toxic level, concentration of the heavy metals and ecological sensitivity to heavy metals (Nabholz, 1991; Singh et al., 2010; Douay et al., 2012). According to this method, the potential risk coefficient (E_R^i) of single element and the potential risk index RI of multi-element can be computed via the following equations:

$$C_f^i = C_D^i / C_R^i \tag{3}$$

$$E_R^i = T_R^i \times C_f^i \tag{4}$$

$$\mathrm{RI} = \sum_{i-1}^{m} E_R^i \tag{5}$$

Where C_D^i is the measured concentration of heavy metal in each sampling point; C_R^i is reference value, baseline level or national criteria, here the background value of each heavy metal in soil is used; $C_f^i = C_D^i / C_R^i$ is the accumulating coefficient for the element of "i" and is single heavy metal pollution index; E_R^i is the potential risk index of single element; RI is integrated potential risk index of multi-element; T_R^i is the toxicity coefficient for the heavy metal elements of "i", represents its toxicity level and the sensitivity to harm the human and the ecosystem (Hakanson (1980). The toxicity coefficients for common heavy metals are Zn = 1 < Cr = 2 < Cu = Pb = 5 < Cd = 30 according to Hakanson (1980).

Classic Hakanson potential ecological risk index method considers eight pollutants, including PCBs, Hg, Cd, As, Pb, Cu, Cr and Zn, lacking of the parameter of toxic response of PCBs, Hg, As in this paper. Due to the difference between the pollutant types and the number, the present study adjust the grading standard of coefficients and indices of heavy metals' ecological risks based on the types and quantity of pollutants (Li et al., 2012). The adjusted grading standard of potential risk of heavy metals in soil was summarized in Table 2.

2.5 Prediction methods of heavy metals pollution

It is a well-recognized opinion that when heavy metals emitted into the soil, it will experience a series of complicated physical, chemical and biological reactions. In the view of the dynamic equilibrium, the concentrations of heavy metals in soil can be predicted when the flux of input and output heavy metals within a certain period of time is clearly known. Also, at the same time, the flux of input and output heavy metals in the future is also unknown, so it is very difficult to quantitatively forecast using the dynamic equilibrium model. In addition, due to a lack of series historical data of heavy metal concentration in soil in this area, regression prediction model to forecast the future situation can not be established. In view of these, Fan et al. (2005), and Yan et al. (2007) developed early warning methods to forecast the development trend of heavy metal pollution in the future through reasonable default of future scenarios, and it has achieved good effects in practical application. In this paper, the method mentioned above was employed to predict the trend of heavy metal (Cd) pollution in the surface soil around coal gangue dump.

Some soil scientists think that the higher industrialization degree, the greater "contribution" to the soil pollution (Yan et al., 2007; Fan et al., 2005), and the pollutants accumulation in the soil is not uniform, but with an accelerated speed. Of course, the impact of industrial development on soil quality does not change if people have strong sense of environmental protection, on the contrary, it will develop with a increasing speed (Yang et al., 2010).

The history of soil pollution caused by coal gangue in coal mine areas can be traced back to 1989, when it was put into operation. Since the year of 2012, the thorough investigation and research on heavy metal pollution in soil were conducted. Based on the above analysis, the present study intended to take 2012 as a benchmark year, the 23 yr (from the year of 1989 to 2012) served as a accelerating stage, using the measured values of heavy metals in 2012 to calculate cumulative acceleration rate

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and cumulative rate (the year of 2012). The present study set the year of 2013 as a starting forecast year.

The formula of heavy metal element accumulation per unit in soil (Q) can be written as

$$Q = a - b \tag{6}$$

$$A = 2Q/T^2 \tag{7}$$

$$V_0 = A \times T \tag{8}$$

Where a is heavy metal concentration in each surface soil sample (measured concentration of heavy metal in soil in 2012), $\mathrm{mg\,kg^{-1}}$; b is the background value of heavy metal in soil, $\mathrm{mg\,kg^{-1}}$; A is cumulative acceleration rate; V_0 is cumulative rate (the year of 2012); $T = 23$ (the heavy metals accumulation years since coal mine put into production).

(1) If social environment protection consciousness is poor in the future, and a lack of environmental protection measures, the accumulation of heavy metals in the soil will be speed up, computation formula is as follows:

$$t = \sqrt{V_0^2 - 2A(c-a)}/A \tag{9}$$

(2) If social environment protection consciousness is strengthened in the future, according to the existing environmental protection strategy, the pollution velocity of soil heavy metals will keep constant, and computation formula is as follows:

$$t = Q/V_0 \tag{10}$$

Where c is the value of soil environment quality standard (national primary and secondary standard GB 15618-1995), $\mathrm{mg\,kg^{-1}}$; a is the concentration of heavy metal element in surface soil, $\mathrm{mg\ kg^{-1}}$, t is fixed number of years exceeding standard, the unit is year; and the notations of A, V_0 and Q have the same meaning as in Eqs. (6)–(8).

3 Results and discussion

3.1 Characteristics of heavy metal concentration

The soil within the scope of the industrial square is covered with different thickness backfill, most of the backfill is coal gangue. The topsoil (0 ~ 0.2 m) is highly complex, so the soil at the depth of 0.4 m was chosen to evaluate the potential ecological risk and predict the trend of heavy metal pollution.

Using the concentrations of heavy metal elements in soil (0.4 m) where the distance from coal gangue dump is more than 600 m as background values, and this area is not affected by coal gangue. The analysis results of heavy metals in coal gangue and the background values of soil (0.4 m) in Yangcaogou coal mine were listed in Table 3.

The value of pH in coal gangue sample is 8.63, showing an alkaline property. Compared with the Environmental Quality Standard for Soils in China published by National Environmental Protection Agency of China (NEPA, 1995), the mean concentrations of Cu, Cd and Pb in coal gangue were lower than the national primary standard of agricultural soil, and the mean concentrations of Cr and Zn were lower than the national secondary standard, but the mean concentrations of the five kinds of heavy metals in coal gangue were all higher than background values of soil in the study area with an order of Cd > Zn > Cr > Cu > Pb.

3.2 Characteristic analysis of pH value and concentration of heavy metal in soil

The pH value of soil was usually divided into five grade, that is strongly acid (pH < 5.0), acid (5.0 ~ 6.5), neutral (6.5 ~ 7.5), alkaline (7.5 ~ 8.5) and the strong alkaline (pH > 8.5). Figure 2 showed the pH value of soil around coal gangue dump. The pH values of soil (0.4 m) around the coal gangue dump ranged from 5.48 to 7.91, which imply a gradual change from acid to alkaline for all soil samples. The change trend of pH value in each sampling line is similar, inversely proportional to the distance from the coal gangue dump.

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Compared with Environmental Quality Standard for Soils in China (GB 15618-1995), the mean concentrations of Cd, Zn, Pb, Cu and Cr in soil (0.4 m) were lower than the national primary standard of agricultural soil. But the mean concentrations of Zn, Cd and Pb were higher than background values of soil (0.4 m), and the mean concentrations of Cu and Cr in most of soil samples were higher than background values of soil (0.4 m). The concentrations of heavy metals in the soil (0.4 m) around coal gangue dump and their descriptive statistical results are listed in Table 4. The concentration distribution contour map of Cd, Pb, Cu, Cr, Zn in the soil (0.4 m) around the coal gangue dump are showed in Fig. 3.

3.3 Heavy metal pollution assessment

Based on the monitoring data of soil quality in study area, a quantitative analysis of soil heavy metal pollution around coal gangue dump was conducted using the method of single-factor pollution index (PI) and Nemerow integrated pollution index (NIPI) (Yang et al., 2010). The PI values varied greatly among heavy metals. The PI values for Cr and Cu in soil around coal gangue dump ranged from 0.80 to 1.13 and from 0.92 to 1.13, respectively, indicating that the soil was uncontaminated to slightly contaminate. The PI values for Zn and Pb in soil around coal gangue dump ranged from 1.16 to 1.42 and from 1.00 to 1.22, respectively, indicating that the soil was slightly contaminated. The PI values for Cd in soil around coal gangue dump ranged from 2.05 to 2.84, indicating that the soil was moderately contaminated. The analysis results showed that the average single pollution index descended in the order of Cd(2.43) > Zn(1.27) > Pb(1.08) > Cu(1.05) > Cr(0.98).

The NIPI values in soil around coal gangue dump ranged from 1.71 to 2.24. Eight units of soil samples were lightly polluted (NIPI ranged from 1.0 to 2.0), and other units of soil samples were moderately polluted (NIPI ranged from 2.0 to 3.0). 61.54 % and 38.46 % of samples were affected by light and moderate pollution, respectively. Statistical results of PI and NIPI of heavy metals in soils around coal gangue dump are listed in Table 5.

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3.4 Concentration distribution of Cd on the soil profile

Compared with the background value of soil in the study area, the soil around coal gangue dump was seriously enriched with heavy metal Cd, and the number of sampling points in L2 got the maximum. In addition, L2 is located in the downwind direction, so the present study chose L2 sample profile to analyze the vertical change of heavy metal Cd (Fig. 4).

From Fig. 4, one can see that the concentration of Cd clearly decreases with increasing distance from the coal gangue dump and with increasing vertical distance from the soil surface.

3.5 Pearson correlation analysis

Usually the content of heavy metal elements originated from the same or similar source tend to have a significant correlation (Hirschfeld, 1935; Rodríguez, 2008), so the correlation between the heavy metal content in soil can be considered as an indicator of whether the source of heavy metals was same or not. The current study used the pearson correlation analysis method embedded in SPSS to analyze the correlation among the different heavy metals and the pH values in the soil samplings around coal gangue dump. The analysis results are listed in Table 6, from where we can observe that the correlation between the heavy metal elements clearly decreases with increasing the absolute of correlation coefficient.

$$r = \frac{N\sum x_i y_i - \sum x_i \sum y_i}{\sqrt{N\sum x_i^2 - (\sum x_i)^2}\sqrt{N\sum y_i^2 - (\sum y_i)^2} \qquad (11)$$

From Table 6, pH and Cd are significantly correlated at the 0.05 level (2-tailed), which implies that they may have the same origin. The same could be found for Cr with Zn and Cd, Cu and Pb, Cd and Pb. And Zn with Cd and Pb are significantly correlated at the 0.01 level (2-tailed). We can infer that the above mentioned five pairs of heavy metal elements may originate from the same source.

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3.6 Correspondence analysis

Corresponding analysis (R-Q type factor analysis), proposed by Hirschfeld (1935) and later developed by Benzécri (1976), has been applied to indicate the main pollution factor in each soil partition of the study area considering the variable load size and the relationship between variables and sampling points partition. Correspondence analysis is a robust multivariate statistical method of studying the relationships between sampling points and variables, underlining R type factor analysis (study of relationships between variables) and Q type factor analysis (study of relationship between samples), and it can reveal variables and samples in the same plane. Using this method, many complicated variables could be compressed to two principal component variables. Then, the relationships between sampling points and variables can be easily analyzed and explained, also, the main variable(s) could be found at the same time (Benzécri, 1973; Dillon and Goldstein, 1984; Rodriguez et al., 2008; Armand et al., 2013; Carmona et al., 2013). The Fig. 5 shows the correspondence map of the relationship among different heavy metals.

It can be seen from Fig. 5 that the sample points were polluted by heavy metals in different degree. From the distribution trend of sample points, 13 points is divided into three zones. I zone: ZK10, ZK11, ZK12, ZK13, ZK14; II zone: ZK01, ZK04, ZK07, ZK08, ZK09; III area: ZK02, ZK03, ZK06. Cr was found to have a close relationship with I zone; Pb and Cd was closely related to II zone; The relationship between sample points and heavy metals Cu and Zn was close in III zone.

3.7 Single and integrated potential risk assessment of heavy metal

Refer to the Hakanson assessment method, the potential risk index of single element in soil (E_R^i) and integrated potential risk index (RI) of multi-element were calculated and the results were shown in Table 7. From Table 7, we can observe that the scopes of the potential risk indices of five kinds of heavy metals are: E_R^i(Cr)1.60 ~ 2.26, E_R^i(Cu)4.62 ~ 5.64, E_R^i(Zn)1.16 ~ 1.42, E_R^i(Cd)61.46 ~ 85.12,

E^i_R(Pb)4.98 ~ 6.12. From the means of the potential risk indices of five kinds of heavy metals, the potential risk arrayed in the order of E(Cd) > E(Pb) > E(Cu) > E(Cr) > E(Zn). Cd was the most dangerous element with average of E^i_R, 72.92. All of the sampling points have strong potential risk of Cd element, and other heavy metals only showed slight potential risk to the environment. Therefore, Cd element was the key element to be further studied.

The scope of RI of multi-element is 75.07 ~ 98.61 (Table 7). Soil potential ecological risk levels around coal gangue dump were level B and C (Fig. 6), and the corresponding ecological damage degrees are moderate and strong, respectively. Strong ecological risk have been estimated to exist in the region near the coal gangue, while at the region a bit far from the coal gangue dump, there exists moderate ecological risk. The researches showed that the soil around coal gangue dump had been polluted in different degrees. Appropriate engineering measures and ecological measures should be taken to control the concentrations of soil heavy metals, and also it is necessary to conduct ecological restoration for pollution area.

3.8 Predicted results and discussion

Cd was the main influence factor to cause the risk based on the above results. Therefore, Cd element was chosen as the key element to predict the risk.

Based on the heavy metal pollution history of Cd in soil (0.4 m) around coal gangue dump, the heavy metal (Cd) cumulative acceleration and cumulative rate were estimated. The fixed number of years exceeding standard prediction model was also established to predict the trend of surface soil heavy metal pollution of Cd under the accelerated accumulation plan mode and the uniform mode. The results were shown in Table 8.

The predicted results indicated that in uniform accumulation mode, the concentration of Cd in each sampling point around the coal gangue dump was higher than primary standard and secondary standard in 12 yr and 30 yr, respectively. In accelerating accumulation mode, the concentration of Cd in each sampling point around the coal gangue dump was higher than primary standard and secondary standard in 10 yr and 20 yr, respectively.

4 Conclusions

This paper presents an investigation of heavy metal pollution in a coal gangue area in Changchun (Jilin Province, North-East China). The present work is performed via field sampling, laboratory experiment and mathematical analysis. Pearson correlation analysis and correspondence analysis are employed to address the sources of heavy metal, relationship between sampling points and variables. Single and integrated ecological risk index have been calculated to reveal the potential pollution of heavy metal. The conclusive findings can be summarized as follows:

(1) The single pollution index of Cd in all soil samples around coal gangue dump was more than 2, and the single pollution indices of Zn, Pb in all samples were more than 1; the single pollution index of Cu in 12 samples was more than 1; the single pollution index of Cr in 7 samples was more than 1, while the single pollution indices of Cu and Cr in other samples were less than 1, and the average single pollution index descended in the order of Cd(2.43) > Zn(1.27) > Pb(1.08) > Cu(1.05) > Cr(0.98). In terms of NIPI, eight soil samples were light pollution (NIPI ranged from 1.0 to2.0), and other soil samples were medium pollution (NIPI ranged from 2.0 to 3.0). 61.54 % samples affected by light pollution and 38.46 % samples affected by medium pollution.

(2) From the averages of the potential risk indices of five heavy metals, the potential ecological risk is arranged in the order of E(Cd) > E(Pb) > E(Cu) > E(Cr) > E(Zn). Cd was the key influence factor to cause the risk, and its mean value of E_R^i was up to 72.92. All of the sampling points at the depth of 0.4 m demonstrated a strong potential risk of Cd element, and other heavy metals in soil around coal gangue dump only existed slight potential risk to the environment.

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(3) The range of RI of multi-element is 75.07 ~ 98.61. Soil potential ecological risk levels around coal gangue dump were level B and C, and the corresponding ecological damage degrees are moderate and strong, respectively. The potential ecological harms decrease with increasing distance from the coal gangue dump. The region near the coal gangue dump was found to exist strong ecological risk, region a bit far from the coal gangue have moderate ecological risk. The researches showed that coal gangue dump had polluted the soil around coal gangue dump. Appropriate engineering measures and ecological measures should be taken to control the concentration of soil heavy metals, and it is urgent to conduct ecological restoration for pollution area.

(4) The predicted results imply that heavy metal pollution of Cd in the study area have a tendency of deterioration within the next few decades. In uniform accumulation mode, the concentration of Cd in each sampling point around the coal gangue dump was higher than primary standard and secondary standard in 12 yr and 30 yr, respectively. In accelerating accumulation mode, the concentration of Cd in each sampling point around the coal gangue dump will be higher than primary standard and secondary standard in 10 yr and 20 yr, respectively.

The findings from the present study can provide useful insights for making precautionary strategy in the study area, and also will be of the most importance for taking ecological restoration of coal mine due to the improper treatment of coal gangue during the coal production.

Supplementary material related to this article is available online at http://www.nat-hazards-earth-syst-sci-discuss.net/2/1977/2014/nhessd-2-1977-2014-supplement.pdf.

Acknowledgements. This research was supported by "China Geological Survey Project" (1212011140027) and Laboratory of scientific test experiment center, Jilin University, Changchun, China. And the financial foundation from base construction of Jilin University (450060481244), Scientific Research Foundation for Returned Scholars of Jilin Univer-

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sity (419080500024) and Seed Science Foundation from Ministry of Education of China (450060488107) are also greatly appreciated.

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Table 1. Brief description of sampling sites around coal gangue dump.

Sampling sites	Longitude	Latitude
ZK01	125°36′07 E	43°57′36 N
ZK02	125°36′06 E	43°57′36 N
ZK03	125°36′05 E	43°57′37 N
ZK04	125°36′08 E	43°57′36 N
ZK06	125°36′11 E	43°57′37 N
ZK07	125°36′11 E	43°57′35 N
ZK08	125°36′09 E	43°57′37 N
ZK09	125°36′15 E	43°57′36 N
ZK10	125°36′18 E	43°57′36 N
ZK11	125°36′12 E	43°57′39 N
ZK12	125°36′12 E	43°57′39 N
ZK13	125°36′07 E	43°57′40 N
ZK14	125°36′05 E	43°57′39 N

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Table 2. The adjusted grading standard of potential risk of heavy metals in soil.

E_R^i	Pollution degree	RI	Risk level	Risk degree
$E_R^i < 30$	Slight	$RI < 40$	A	Slight
$30 \leq E_R^i < 60$	Medium	$40 \leq RI < 80$	B	Medium
$60 \leq E_R^i < 120$	Strong	$80 \leq RI < 160$	C	Strong
$120 \leq E_R^i < 240$	Very strong	$160 \leq RI < 320$	D	Very strong
$E_R^i \geq 240$	Extremely strong	$RI \geq 320$	–	

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Table 3. The concentrations of heavy metals in coal gangue and the background values of soil ($mg\,kg^{-1}$).

	Cr	Cu	Zn	Cd	Pb
Coal gangue	92.29	24.81	107.89	0.18	21.30
Background values	71.22	20.39	51.07	0.06	19.35

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Table 4. The concentrations of heavy metals in the soil (0.4 m) around coal gangue dump.

element	sampling line	Minimum ($mg\,kg^{-1}$)	Maximum ($mg\,kg^{-1}$)	Mean ($mg\,kg^{-1}$)	Standard deviation	Variation coefficient	Background values ($mg\,kg^{-1}$)
Cr	L1	59.75	77.43	66.48	8.20	0.12	71.22
	L2	57.11	80.54	69.69	9.01	0.13	
	L3	65.02	80.54	72.88	7.76	0.11	
Cu	L1	18.82	21.46	20.52	1.25	0.06	20.39
	L2	20.63	22.58	21.57	0.83	0.04	
	L3	21.83	23.02	22.60	0.67	0.03	
Zn	L1	59.36	68.81	64.45	3.94	0.06	51.07
	L2	61.27	68.73	64.51	2.97	0.05	
	L3	61.11	72.27	67.55	5.77	0.09	
Cd	L1	0.13	0.18	0.16	0.02	0.14	0.06
	L2	0.12	0.17	0.15	0.02	0.11	
	L3	0.13	0.17	0.16	0.02	0.15	
Pb	L1	19.54	20.16	19.92	0.29	0.01	19.35
	L2	19.42	23.70	20.85	1.80	0.09	
	L3	19.29	23.64	22.15	2.48	0.11	

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Table 5. Statistical results of pollution index (PI) of heavy metals in soils around coal gangue dump.

	PI					NIPI
	Cr	Cu	Zn	Cd	Pb	
Max	0.8	0.92	1.16	2.05	1	1.71
Min	1.13	1.13	1.42	2.84	1.22	2.24
Mean	0.98	1.05	1.27	2.43	1.08	1.97

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Table 6. The results of the correlation analysis between the different measured index.

Correlation	pH	Cr	Cu	Zn	Cd	Pb
pH	1	–0.395	–0.292	0.542	0.670[a]	0.368
Cr	–0.395	1	0.258	–0.591[a]	–0.598[a]	–0.088
Cu	–0.292	0.258	1	0.418	0.155	0.583[a]
Zn	0.542	–0.591[a]	0.418	1	0.846[b]	0.768[b]
Cd	0.670[a]	–0.598[a]	0.155	0.846[b]	1	0.654[a]
Pb	0.368	–0.088	0.583[a]	0.768[b]	0.654[a]	1

[a] Correlation is significant at the 0.05 level (2-tailed).
[b] Correlation is significant at the 0.01 level (2-tailed).

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Table 7. Statistics of potential risk index (E_R^i) and potential risk index (RI).

	Potential risk index of single element E_R^i					RI
	Cr	Cu	Zn	Cd	Pb	
Maximum	2.26	5.64	1.42	85.12	6.12	98.61
Minimum	1.60	4.62	1.16	61.46	4.98	75.07
Mean	1.96	5.24	1.27	72.92	5.39	86.79

Table 8. The fixed number of years exceeding standard of Cd in soil around coal gangue dump (yr).

Sampling line	Sample number	Accelerating accumulation mode		Uniform accumulation mode	
		Fixed number of years exceeding standard			
		Primary standard	Secondary standard	Primary standard	Secondary standard
L1	1-05	6	15	6	19
	2-07	3	11	3	13
	3-07	2	10	2	12
	14-2	10	20	11	28
L2	4-07	6	15	7	20
	8-08	3	11	3	14
	6-06	7	16	8	22
	12-2	10	20	12	29
	11-1	10	20	12	30
L3	7-05	3	11	3	13
	9-02	2	10	3	13
	10-2	9	20	11	28

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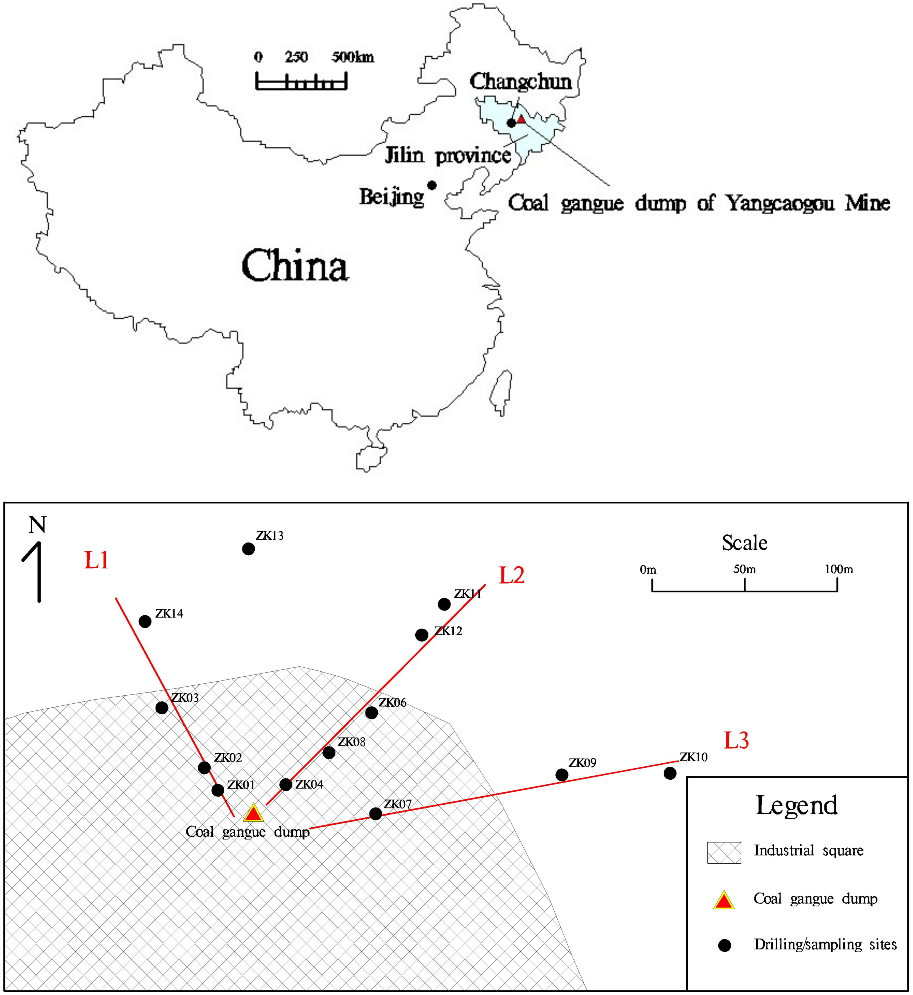

Fig. 1. Location of Yangcaogou Mine and soil sampling points around the coal gangue dump.

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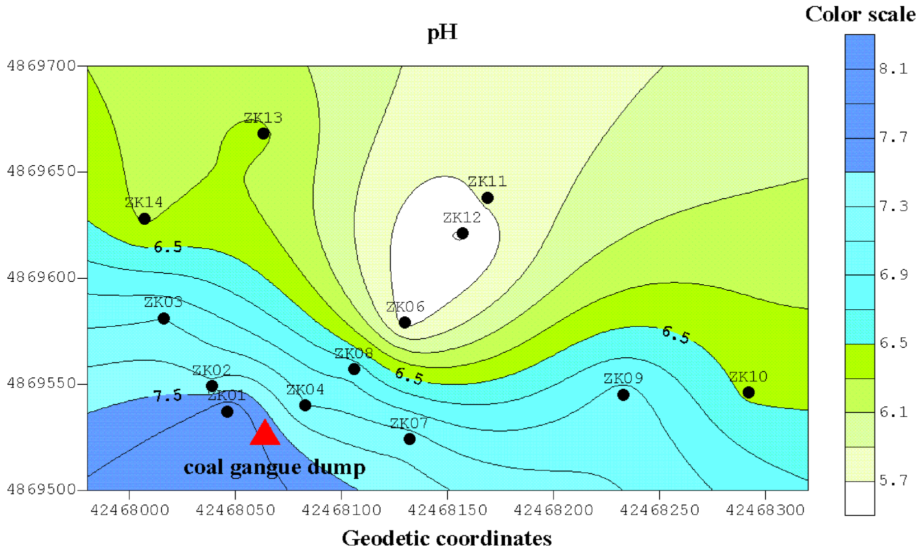

Fig. 2. Contour map of pH in soil around the coal gangue.

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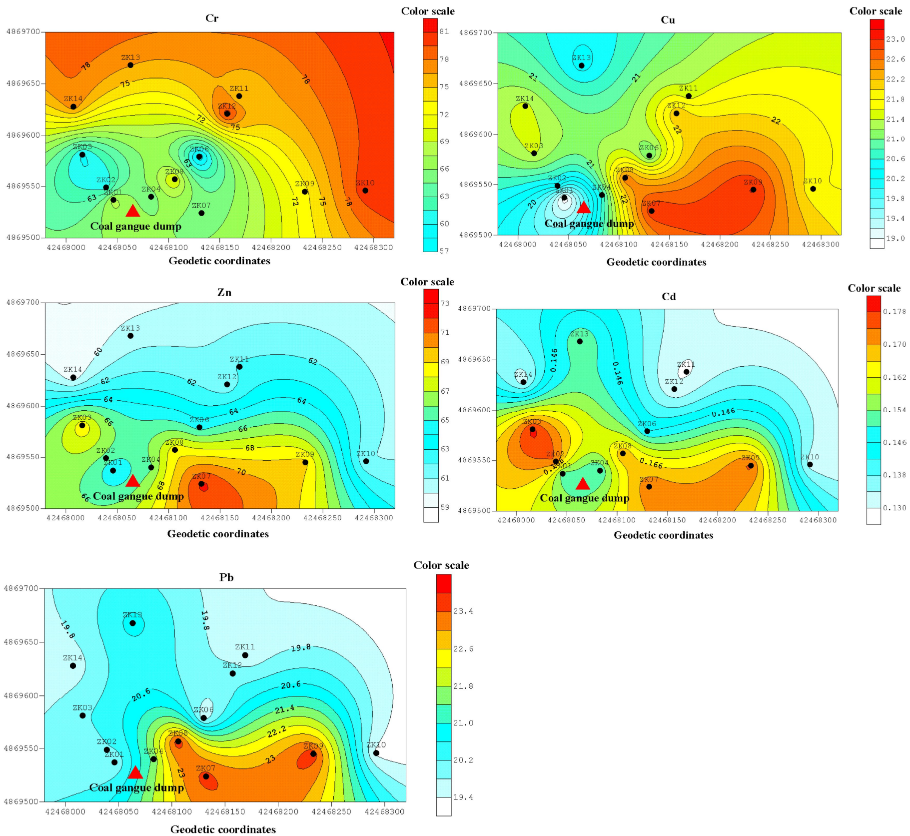

Fig. 3. The concentration distribution contour map of Cr, Cu, Zn, Cd, Pb in the soil (0.4 m) around the coal gangue dump (The black circular points represent sampling points, and the red triangle represent the coal gangue dump).

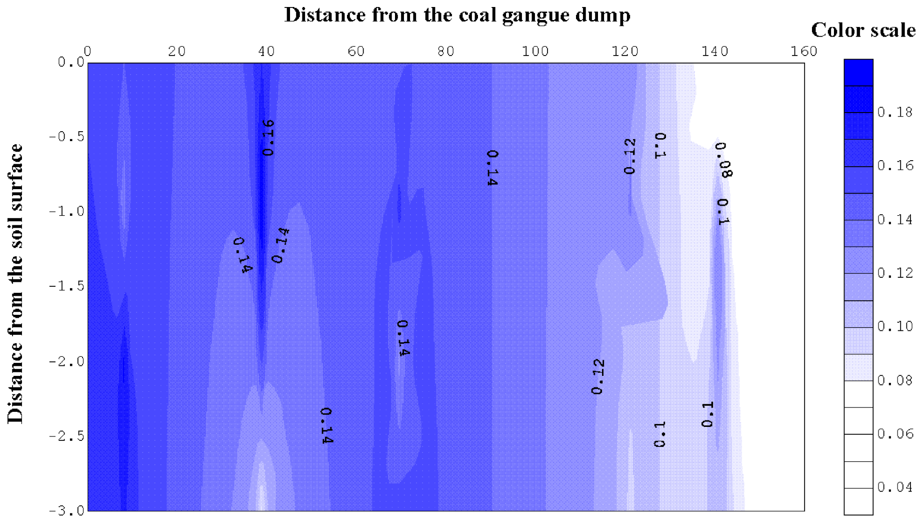

Fig. 4. The concentration distribution map of Cd on the soil profile.

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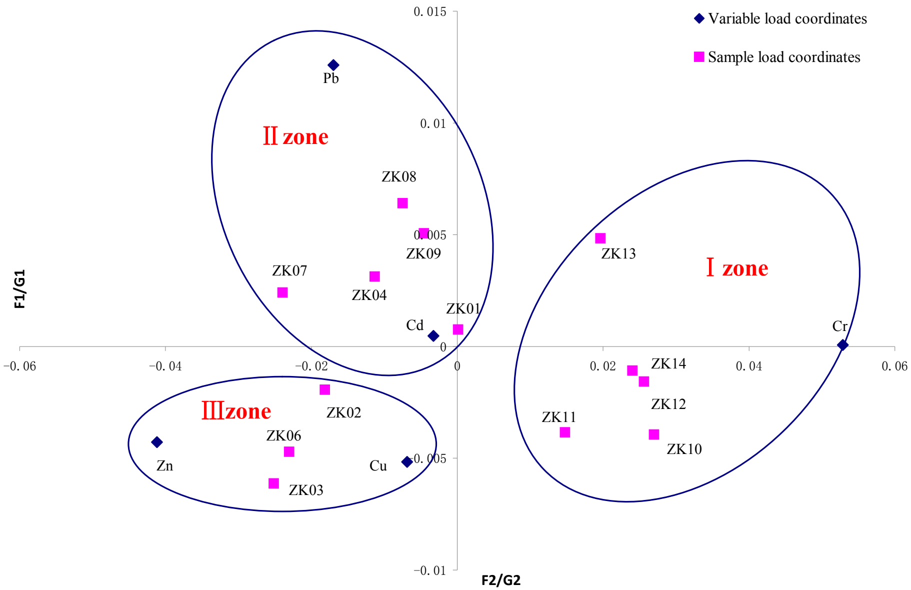

Fig. 5. The correspondence map of the relationship.

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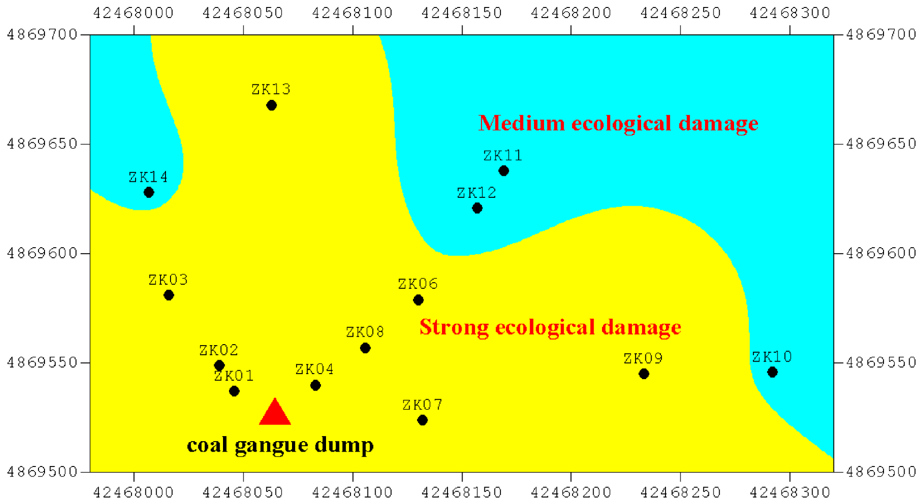

Fig. 6. The map of potential ecological harm distribution (The black circular points represent sampling points, and the red triangle represents the coal gangue dump).

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