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Health risk assessment for typical and abandoned chromium-contaminated sites

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Abstract Chromium pollution were investigated by sampling the abandoned chromium contaminated sites in Chongqing. Its health risk was assessed following the procedures made by USEPA. The results showed that the most seriously polluted land was found at the former production area (G4) with average Cr(VI) concentration of 3369.2 $mg \cdot kg^{-1}$ on the surface soil. The assessment indicated that there was high risk of non-carcinogens for children. The assessed risk of the first layer of office area (G1) for children, chromium slag transition (G2), drainage pipeline chromium slag transition (G3) and production area (G4) for both children and adults were not acceptable ($>1.00\times10^{-6}$). It was strongly suggested that the accessible measures of remediation should be taken for a portion of contaminated sites before the reuse of abandoned lands.

Key words chromium(VI) contamination; health risk assessment; exposure; carcinogen; non-carcinogen

1 Introduction

With fast economic development and adjustment to the industrial structures in Chongqing City of Southwest China, many factories and enterprises dealing with metallurgy, petro-chemistry, pesticides and textile etc. were forced to be closed or relocated. And the original lands are planned to be commercial or residential areas. However, the utilization of abandoned sites arouses lots of worries with the potential impacts on people's health, particularly for the harmful residues in the soils of abandoned sites. Thus the environmental risk assessments, especially the health risk assessment of abandoned lands became a key step for the reconstruction projects.

In 1980s, US Environmental Protection Agency (USEPA) made the risk assessment guidelines and technical regulations as the authoritative standards for the performance of health risk assessment and the reclamation of typically contaminated sites (US Congress, 1980; USEPA, 1985, 1988, 1989). Meantime, USEPA also jointly established the frameworks of human health risk and ecological risk in cooperation with World Health Organization (WHO) and Organization for Economic Co-operation and Development (OECD) (Suter et al., 2005). The early researches reported a number of successful samples for the health risk assessment of contaminated sites. For example, the health risk of the smelting plant was assessed in Poland (Wcislo et al., 2002); GIS technique was used to complete a detailed analysis process of the human health risk assessment in a European factory (Morra et al., 2006). In China, the health risk assessment of organic pollution at a factory in Changzhou City (Chen et al., 2006) and the ecological and health risks of the polycyclic aromatic hydrocarbons in sediments of Taihu Lake (Qiao et al., 2007) were fulfilled and evaluated by local EPA.

Chemical-industrial enterprises is considered as the most threatening source of pollution, but the health risk assessment focused on Cr(VI) at the site of chemistry-industrial plant has seldom be found in publications. There are many large factories distributing widely in China, including one in Chongqing City. Since this enterprise was closed a few years ago, the abandoned site will be reused for construction of commercial or residential communities, so the evaluation of health risk at the abandoned site became a very necessary and critical procedure. This paper uses the framework of USEPA standards in correspondence

with local natural conditions and soil properties for health risk assessment at the site of abandoned chem-industrial plant and provides the essential scientific bases for people's health protection in addition to suggestions of remedial technologies.

2 Samples and methods

2.1 Study areas

The studied chem-industrial plant was built in 1959 and located in northwestern Chongqing where all the industrial factories and enterprises had to be relocated. In a total area of 281000 m^2, the plant mainly produced sodium dichromate, chromic anhydride, chrome green and chromium powder before being closed in 2008. In the long-term production process, the chromium slag was buried under ground at a landfill or piled on the land surface. Although the remained slag was moved away in 2010, chromium compounds, especially Cr(VI) residues, appeared in high concentrations in deep soils due to the vertical leaching by rainwater flushing. As this area was seriously contaminated, the health risk assessment must be conducted before the reuse of abandoned land.

2.2 Soil sampling and analysis

Based on the distribution of former plant constructions and local natural conditions, the range of whole plant was divided into 7 sampling areas marked as: office area (G1), chromium slag transition area (G2), drainage pipeline of chromium slag transition (G3), production area (G4), west side of mountain area (G5), bottom of interception dam (G6) and south area under the railway bridge (G7). A total of 192 soil samples taken from different depth (0–0.5, 2.5–3.0, 5.5–6.0 m) were collected within the former chemical plant circle. The soil samples were obtained by drilling, then by using the combined means of judgment sampling method and grid sampling method. After the removal of gravels, plant roots and other debris, the soil samples were air-dried, ground, sieved, and finally saved to be analyzed. The concentration of total Cr was determined on a flame atomic absorption spectrophotometry method while Cr(VI) was determined by two benzene carbonyl two hydrazine spectrophotometry method.

2.3 Health risk assessment

2.3.1 Exposure route analysis

Since the main pollutant at the site was chromium that has no gaseous phase, the exposure routes were generally through oral intake, dermal contact and respiring.

2.3.2 Exposure dose calculation

Chromium is carcinogenic material through respiring route, but there was no literature to evidence that it had carcinogenic effect by oral intake. The carcinogenic risk of chromium caused by the respiring route was only mentioned in a few papers (Ferreira-Baptista et al., 2005).

The average daily exposure through oral intake was calculated by equation (1), and the relevant definitions for all items are listed in Table 1.

$$\mathrm{CDI}_{\mathrm{oral}} = \frac{C \times IR_{\mathrm{oral}} \times EF \times ED \times CF}{BW \times AT} \quad (1)$$

The average daily exposure through dermal contact was obtained by equation (2):

$$\mathrm{CDI}_{\mathrm{dermal}} = \frac{C \times SA \times CF \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (2)$$

The average daily exposure through respiring intake was computed by equation (3):

$$\mathrm{CDI}_{\mathrm{res}} = \frac{C \times IR_{\mathrm{res}} \times EF \times ED}{PEF \times BW \times AT} \quad (3)$$

where, the meanings and values of all physical quantities in above equations are described in Table 1.

2.3.3 Characterization of non-carcinogenic risk

The non-carcinogenic risk index is defined by the ratio of daily intake as exposure and reference doses, and could be obtained by equations (4) and (5), respectively (USEPA, 1989).

When the potential risk caused only by one substance, the calculation should be calculated by equation (4):

$$\mathrm{HQ}=\mathrm{CDI}/\mathrm{RfD} \quad (4)$$

For the existence of more substances, the potential risk could be calculated by Equation (5):

$$\mathrm{HI}=\sum \mathrm{HQ}_i \quad (5)$$

where, HQ is defined as the non-carcinogenic risk index; CDI is long-term daily intake; RfD is the reference dose, and HI is a pollutant exposure pathways of non-carcinogenic risk. If HQ or HI<1, it indicates that risk is limited or negligible, but if HQ or HI>1, it presents that there exists non-carcinogenic risk.

2.3.4 Characterization of carcinogenic risk

The carcinogenic risk of chromium is caused by daily intake in a long time and the slope factor, which indicates that the incidence of cancer is caused by one kind of chemical substances exceeding the normal level. The carcinogenic risk was obtained by equations (6) and (7), respectively (USEPA, 1989):

$$\text{Risk}=\text{CDI}\times\text{SF} \quad (6)$$

$$(\text{Risk})_T=\sum(\text{Risk})_T \quad (7)$$

where, risk is the pollutant carcinogenic risk index, intake refers to the daily intake for a long-term, and SF is the slope factor.

The acceptable risk value for carcinogenic substance defined by USEPA is that the risk of cancer in life exceeds normal level of 1.00×10^{-6}–1.00×10^{-4}. The boundary was taken with 1.00×10^{-6} in this study.

The values of reference dose of RfD and the slope factor of SF are listed in Table 2.

Table 1 Adoptive exposure parameters and relevant reference values

Parameter	Definition	Unit	Value
CDI_{oral}	Dose of oral intake	mg/(kg·d)	Formula (1) calculate
CDI_{dermal}	Dose of dermal intake	mg/(kg·d)	Formula (2) calculate
CDI_{res}	Dose of respiring intake	mg/(kg·d)	Formula (3) calculate
C	Concentration of Cr	mg/(kg·d)	Actually measured
IR_{oral}	Oral intake rate of soil	mg / d	200 (Child) 100 (Adult)
IR_{res}	Respiratory rate	m^3/d	7.63 (Child) 20 (Adult)
AF	Skin adhesion	mg/(cm^2·d)	0.2 (Child) 0.07 (Adult)
ABS	Dermal absorption factor	dimensionless	0.001
SA	Skin contact area	$cm^2\cdot d^{-1}$	2800 (Child) 5700 (Adult)
EF	Exposure rate	d/a	350 (Child) 180 (Adult)
ED	Exposure time	a	6 (Child) 30 (Adult)
PEF	Dust emission factor	m^3/kg	1.36×10^9
BW	Average weight	kg	15 (Child) 60 (Adult)
AT	Average exposure time	d	ED×365 (Non-carcinogen) 70×365 (Carcinogen)
CF	Conversion factor	kg/mg	10^{-6}

Note: The parameters in Table 1 were essentially referred from USEPA database (Jiang and Wang et al., 2004) while a part of values were revised based on natural conditions at Chongqing City.

Table 2 Reference doses and gradient factors of Cr for different exposure routes

Pollutant	RfD_{oral} [mg/(kg·d)]	RfD_{dermal} [mg/(kg·d)]	RfD_{res} [mg/(kg·d)]	SF_{res} $[mg/(kg\cdot d)]^{-1}$
Cr	3.00×10^{-3}	6.00×10^{-5}	2.86×10^{-5}	42.00

3 Results and discussion

3.1 Concentrations of Cr in contaminated soils

From the concentrations distribution profiles of Cr at the former chemical plant listed in Table 3, it can be seen that the entire plant areas are seriously polluted by chromium in different degree. The most serious pollution is found at production area (G4) with average chromium concentration of 3369.2 $mg\cdot kg^{-1}$ on topsoil. The less serious spot is the drainage pipeline of chromium slag transition (G3) with average chromium concentration of 1015.6 $mg\cdot kg^{-1}$ on topsoil, while the average chromium concentration of 128.0 $mg\cdot kg^{-1}$ on topsoil at the west side of the mountain (G5) caused slight pollution. Furthermore, chromium contents in soils tended to be gradually decreased with the increase of soil depths in different soil profiles. However, the phenomenon of high accumulation of chromium in the spatial distributions was found in the former slag landfill and piled area, which was related to flushing and leaching of chromium slag by rainfall infiltration.

3.2 Non-carcinogenic risk

As shown in Figs. 1 and 2, the related non-carcinogenic risk values for children at sampling points appeared to be greater than the acceptable value of 1.0 except for the third layer of G1, the second and third layers of G7 and all layers of G5. Meanwhile, the non-carcinogenic risk value for adults on the first layer of G4 exceeded the acceptable value of 1.0. In general, the non-carcinogenic risk values of the first layer at all the sampling points were higher than the limitation, which indicated that soils within the former plant range could cause high non-carcinogenic risk to children.

3.3 Carcinogenic risk

The results of carcinogenic risk assessment at the chemical plant are shown in Figs. 3 and 4, respectively. It can be seen that the carcinogenic risk values for children are in a range between 1.26×10^{-7} and 4.49×10^{-6}. The values of carcinogenic risk for children of sampling points exceeded the acceptable value of 1.00×10^{-6}, were found at the first layer for both G1 and G2, the second and third layers of G3 and all lay-

ers of G4. Meanwhile, the carcinogenic risk values for adults were in a range of 2.13×10^{-7}–7.35×10^{-6}, in which all layers of G2, G3 and G4 were beyond the acceptable value of 1.00×10^{-6}. The soil quality of G2, G3 and G4 could result in the high carcinogenic risk to both adults and children; furthermore, the first layer of soil at G1 would also cause carcinogenic risk to children.

4 Conclusions

Based on the analysis, it could be summarized that the entire former plant areas were seriously polluted by chromium in different degree. The most serious pollution happens at production area (G4) with the average chromium concentration of 3369.2 mg·kg^{-1} on topsoil. The non-carcinogenic risk values of the first

Table 3 Total Cr concentrations measured in soil samples

Sampling point	Layer	Minimum (mg·kg^{-1})	Maximum (mg·kg^{-1})	Mean value (mg·kg^{-1})	Standard deviation (mg·kg^{-1})	Coefficient variation (%)	Standard value[d] (mg·kg^{-1})
G1	1[a]	98.5	490.2	217.7	105.2	48.3	250
	2[b]	41.7	486.3	210.7	126.9	60.2	
	3[c]	42.5	287.2	127.4	63.0	49.5	
G2	1	735.2	1030	882.6	208.5	23.6	
	2	614.2	827.2	720.7	150.6	20.9	
	3	309.0	602.1	455.6	207.3	45.5	
G3	1	566.0	3330.0	1524.3	1015.6	66.6	
	2	369.0	1210.0	817.5	312.9	38.3	
	3	93.2	876.0	584.5	273.9	46.9	
G4	1	450.0	13700.0	3369.2	3145.2	93.4	
	2	124.9	9220.0	2169.8	2135.2	98.4	
	3	19.6	4380.0	1190.0	1213.5	102.0	
G5	1	68.6	270.1	128.0	62.4	48.8	
	2	49.4	405.4	132.2	107.5	81.3	
	3	48.2	268.5	97.7	71.6	73.3	
G6	1	63.4	805.5	317.8	221.4	69.7	
	2	54.3	653.2	255.5	194.8	76.2	
	3	48.6	816.1	242.1	242.4	100.1	
G7	1	212.7	636.4	424.6	299.6	70.6	
	2	116.5	275.2	195.9	112.2	57.3	
	3	116.4	118.7	117.6	1.6	1.4	

Note: a. First layer depth of 0−0.5 m; b. second layer depth of 2.5−3.0 m; c. third layer depth of 5.5−6.0 m, and d. secondary standard of soil environmental quality standards.

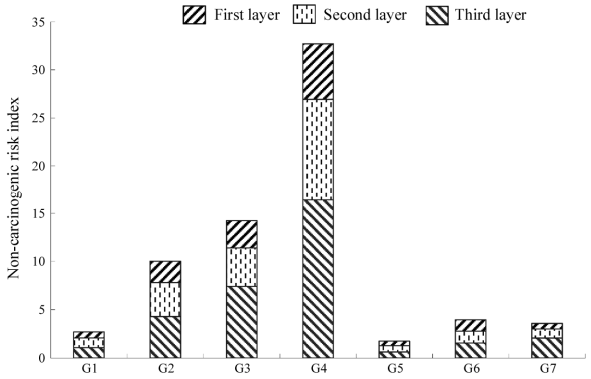

Fig. 1. Integrated risk index of non-carcinogens for children at different sampling points.

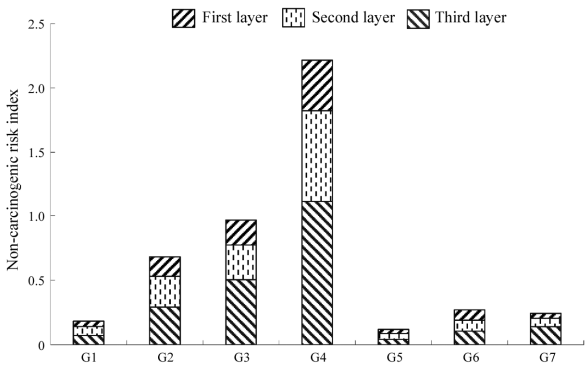

Fig. 2. Integrated risk index of non-carcinogens for adults at different sampling points.

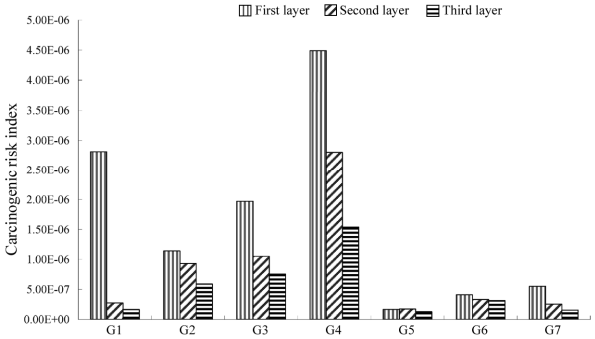

Fig. 3. Integrated risk of carcinogens for children at different sampling points.

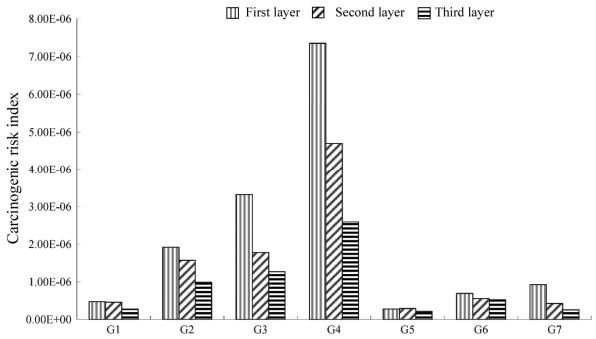

Fig. 4. Integrated risk of carcinogens for adults at different sampling points.

layer at all the sampling points were higher than limitation and could cause high non-carcinogenic risk to children. The carcinogenic risk values of the first layer of soil at G1 to children and the soils at G2, G3 and G4 to both adults and children obviously exceeded limitation of 1.00×10^{-6}. Considering a negative impact on people's living conditions in the future, it is strongly suggested remediation measures, such as phy-chemical remediation and bioremediation, must be taken to reclaim and recover the soil quality at the ruin of abandoned chemical plant prior to reconstruction.

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