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Health Risks of Organic Contaminated Soil in an Out-of-Service Oil Refinery Site

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ABSTRACT: Health risks assessment due to exposure to organic contaminated soil at an oil refinery out of service is conducted in this study by integrating health risk methods of U.S. EPA (United States Environmental Protection Agency), ASTM (American Society for Testing and Materials) and VROM (Ministry of Housing, Spatial Planning and the Environment in Netherlands), with the localized parameters using Chinese demographic characteristics and site features. The monitoring results show that the concentration of benzene, benzo(a)pyrene and benzo(a)anthracene in the soil of the site all exceed their risk screening values, respectively, with pollutants concentrated in soil 0.1–5.5 m beneath the surface. To estimate health risks of the site with the 95% upper confidence limit of the pollution, we focus on three major exposure pathways, namely, oral ingestion, dermal contact and breath inhalation, which indicates carcinogenic risk (CR) and non-carcinogenic hazard quotient (HQ) of the contaminated soil reach 9.59×10^{-5} and 15.46, respectively, exceeding the acceptable level of 10^{-5} and 1, posing severe health hazards to the residents at the site.
KEY WORDS: contaminated site, health risk, soil, organic contamination.

INTRODUCTION

In the process of urbanization in China, most factories in the downtown areas in cities or suburbs of cities have been moved to the industry gardens or special zones for industry as planned. The industrial sites have all been polluted due to the operation of the factories to some extent, only different in degree. Bequeathal sites of moved or terminated industrial plants have become an important genre of Chinese contaminated sites (Guo et al., 2010). Because of the health hazards against lives and work of people on site (Castelli et al., 2005; Hao et al., 2004; Robson, 2003), the health and safety problems of the contaminated sites are drawing more and more attention.

Health risks assessment of contaminated sites refers to the qualitative and quantitative estimation of the harm of various pollutants at the site to human health (Chen and Liu, 2006; Chen et al., 2006). Risk assessment of contaminated sites allows us to identify potential harms at the site and spatial distribution of risks, to confirm whether the site needs to be remedied or other actions have to be taken, and to determine the pollutant tolerance level at which public health can be fully protected and serves as the scientific basis for pollution control and management decisions (Kao et al., 2007; Fernández et al., 2006). It facilitates the analysis and comparison of the validity of various remedial measures and provides the basis for reasonable planning of land use and pollution control and allows us to avoid site contamination risks effectively.

The theory concerned with health risks assessment was first proposed by the National Academy of Sciences in America in 1972 (NAS, 1983). Since 1980s, American and European countries have established their own health

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risk assessment systems adapted to the reality in their own countries on the basis of health risk assessment theory (Colin, 1999; U.S. EPA, 1991). China has not developed her own health risks assessment method and approach so far. Chinese scholars usually adopt internationally recognized systems and methods such as RBCA (risk-based corrective action), CLEA (contaminated land exposure assessment), Csoil (contaminants in soil) and other models for the health risks assessment of contaminated sites at present (Li et al., 2011; Zhang H J et al., 2010; Zhang Y et al., 2008). However, the results might be inaccurate and unreliable if they are applied without taking into consideration of particularity of Chinese cases in the site type, the exposure pathway and the parameters of each model.

This article takes a contaminated site of an oil refinery out of service as its research subject and health risks are assessed on the basis of the site investigation and pollutant determination, using the ASTM site health risks assessment method together with some models in the CCME (Canadian Council of Ministers of the Environment) and VROM at the same time, with parameters localized in the model. The pollutant concentration, toxicity, harm and transfer pathways at the site are analyzed in detail and the pollutant transfer model, the acceptable risk level and model parameters are discussed at length so as to provide the technological data and support for the assessment and management of contaminated site risks of the same type.

SITE ENVIRONMENTAL CHARACTERIZATION

Background of the Site

The study site was formerly an oil refinery in a provincial capital city. It was built in the 1950s and was demolished after it ceased its production in 2002 and has been out of service since then. The site, once an industrial zone, was developed into a residential and commercial area after land utilization readjustment. The surrounding area also has been gradually developed into a densely populated commercial and residential area. The site is located at the accumulation plain in the third grade terrace of Yangtze River basin, covering an area of 8 410 m^2, with a ground level of 20.25–22.46 m. Its underlying bed is majorly of Holocene strato-type alluvium (Q_4^{al}) with primarily clay in the upper layer with a burial depth of 0.7–24.6 m and sandy gravel in the bottom layer at a burial depth of 24.3–53.6 m.

Soil Condition Investigation at the Site

On-site investigation shows that water accumulates in low-lying land there and it is apparently red and caramel with a film of oil covering. The site is visibly polluted with petroleum, diffusing pungent odors. According to the production records, the oil refinery mainly produced such products as vaseline, lubricating lipid, calcium base grease, antirust oil, engine oil, coal tar product-benzene, toluene, xylene, crude phenol, cresylic acid and mixed cresol. Based on an environmental chemical behavior analysis on its production technique, and physical and chemical properties of its products as well, it is decided that the environmental monitoring targets at the organic pollutants at site, namely arsenic, beryllium, lead, boron, cadmium, mercury, selenium, chromium, nickel, iron, copper and other heavy metals, sulfates, petroleum, polychlorinated biphenyls, polycyclic aromatic hydrocarbons, phenols, VOCs, organic chloride, and phthalocyanine esters, aiming at an extensive research into the extent and degree of the pollution at the study site.

The study suggests the underlying bed of the site area is mainly clay of the Quaternary system with low permeability coefficient. It is considered a non-aquifer in hydrogeology. In the shallow strata of the quaternary system, there are phreatic water and unsaturated flow, which are insufficient as water supply due to their small volumes. Deep-groundwater is pore-space water in loosening rocks at the bottom. Water-bearing rock formation are composed by Quaternary system Holocene series mild sand clay sandy loam、silty-fine sand and mucky mild clay. The thickness of aquifer is 3.5–6.0 m, buried depth of groundwater reaches 6.75–12.82 m. The pollution is concentrated in the soil layer.

Systematic grid sampling was conducted in accordance with requirements of the Technical Specifications for Environmental Monitoring of Soil. Firstly, Samples were collected in the center of every grid at an interval of 50 m×50 m. At the same time, monitoring in the workshop, warehouse and disposal area were intensified. Four samples were collected from every grid, at depth of 0.5, 1.5, 3 and 5 m respectively under surface. In some grids, the sampling depth reached 10 m. A total of 121 soil samples were collected from the 28 soil monitoring spots (Fig. 1 shows the spots location of soil samples). Then,

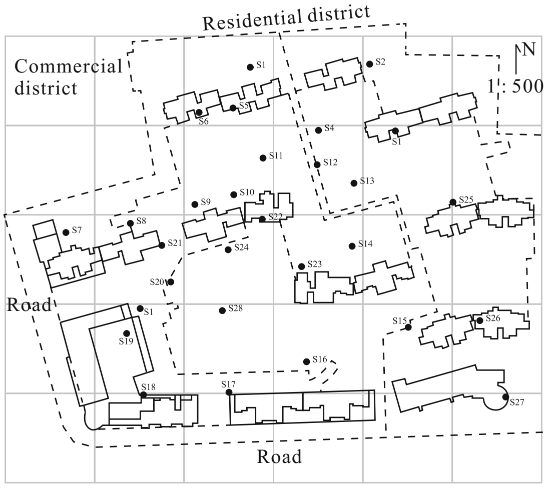

Figure 1. Spots location of soil samples.

soil samples were tested in the State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan, China.

HAZARD IDENTIFICATION

The study site has been planned to be used as a residential area. However, there are not clear-cut standards available for residential land environmental quality assessment covered in the present environment quality assessment standards for soil in relevant documents in China. As a result, the present study has no choice but to apply provisional A level of standard of soil quality assessment for exhibition sites. Comparing the above-mentioned A level standard with the test results, it is found that the study site has been polluted to some extent by such heavy metals as copper, zinc, and nickel, in addition to petroleum hydrocarbons, PAHs (polycyclic aromatic hydrocarbons), PCBs (polychlorinated biphenyls), organic chlorine and phenols. Furthermore, the concentration of benzene, benzo(a)pyrene and benzo(a)anthracene in soil has exceeded starting values of risk for soil pollutants according to the screening values of residential and public land soil contamination stipulated in relevant documents issued by U.S. Environmental Protection Agency (EPA) for health risk assessment of site. The features of pollutants in soil and concentration of representative soil samples are presented in Tables 1, 2 and Fig. 2.

EXPOSURE ASSESSMENTS

Exposure Pathways

The study site shall be exploited as residential land, and naturally the major sensitive group is the residents in the community, including adults and children. The target

Table 1 Features of pollutants in the soil

Pollutants	Concentration range (mg/kg)	Vertical depth (m)	Screen value (mg/kg)	Samples exceeding standard rate
Benzene	0.005–5.86	0.2–5.4	1	26.4%
Benzo(a)anthracene	0.07–24.83	0.1–4.3	0.6	31.4%
Benzo(a)pyrene	0.13–4.91	0.1–5.5	0.06	37.2%

Table 2 Characteristics of contaminated soil

Strata	pH range	Distribution range	Burial depth (m)	Inorganic contaminated constituents	Inclusion compound
Miscellaneous fill	4.34–8.72	All the site	0–1.2	SO_4^{2-}, Cu, Ni, Zn	Building waste and clay, loose structure
Silty clay	4.16–9.80	All the site	0.8–2.9	SO_4^{2-}, Cu, Ni	Iron oxide film in it
Mucky soil	5.86–10.15	All the site	1.0–6.1	SO_4^{2-}, Cu, Ni,	A small amount of organic matter and ferric oxide with thin layer silty, few shell fragments
Clay	7.71–10.40	North of the site	4.5–9.5	Cu, Ni,	A little of ferric oxide with few silty
Silty clay and silt	7.24–10.68	Southwest of the site	1.2–9.8	SO_4^{2-}, Cu, Ni,	Silty with a lot of megohmit, silty clay is plastic

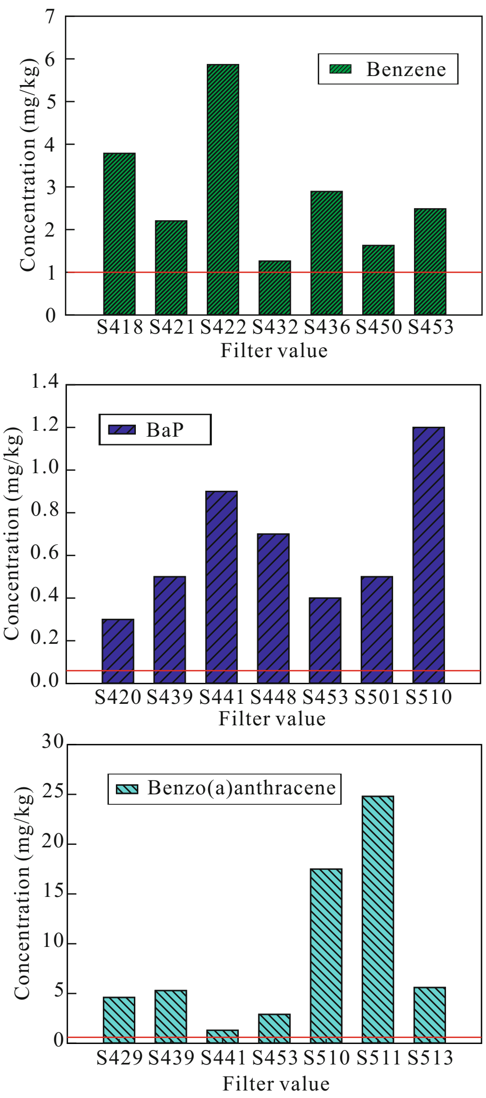

Figure 2. Pollutants density of representative soil samples.

pollutants are mainly volatile organic compounds that are persistent and mobile in soil. They could be desorbed from soil under certain situation (proper temperature, air pressure and land disturbance) (Wu et al., 2005) and inhaled by human beings after evaporation. Therefore the on-site residents are exposed to the pollutants in daily life mainly via contacting polluted soil and inhaling pollutant vapor. By analyzing the daily activities of the residents, typical contacting with the potential pollutants of the sensitive groups include the following scenarios: children put polluted soil into their mouths when playing; people contact with polluted soil and take in pollutants via skin when working on site; inhaling pollution in the soil dust and pollution vapor from soil. The exposure pathways of sensitive groups are identified based on the movement characteristics of pollution in the environment associated with polluted soil as dental intake, skin contact and inhaling soil particles and soil pollution vapor indoor and outdoor.

Exposure Dose Assessments

The assessments of exposure dose involve both exposure dose of carcinogenic pollutants in which life-long health effects should be considered and non-carcinogenic pollutants in which the harmful effects of exposures of children should be taken into account. The exposure dose model shall be established according to the models and methods adopted by U.S. EPA and ASTM (U.S. EPA, 2003, 1996), and by combining the absorption efficiency of human beings through oral intake stipulated by CCME (CCME, 1996), and exposure assessment model for breath inhalation of soil particles established by VROM (Zhao, 2008). The human body parameters should be modified according to Chinese demography (MHPRC, 2006). For exposure dose assessment models and exposure parameters, please refer to Tables 3 and 4.

TOXICITY ASSESSMENTS

According to EPA carcinogen classification standard (Hu, 2009), three pollutants discussed in this paper are all carcinogenic pollutants, with benzene known as a highly potential human carcinogen B1, benzo(a)pyrene known as human carcinogen A and benzo(a)anthracene known as potential human carcinogen C. The International Institute of Cancer Research confirms that benzene is an environmental carcinogen that could cause blood and hereditary damages to human beings. A large amount of epidemiological studies also demonstrates that death rate of workers of benzene-related industries from leukemia, malignant lymphoma and non-proliferative diseases of blood system exceed that of the average level of population These workers are more vulnerable to miscellaneous leukemia and malignant lymphoma (Du and Ge, 2009). Benzo(a)pyrene is the most common carcinogen in the environment. It is strongly teratogenic, mutagenic and endocrine-disrupting. Once taken in, it will damage human organs. Animal experiments reveal that long-term intake of benzo(a)pyrene can lead to malignant tumors such as gastric cancer, skin cancer and lung cancer (Luo et al., 2011). Benzo(a)anthracene is considered to be

Table 3 Exposure dose and risk characterization calculation models

Risk	Exposure pathways		Exposure dose (kg soil/kg weight d)
Cancer effect	Oral ingestion		$OISERca=(\frac{OSIRc\times EDc\times EFc}{BWc\times ATca}+\frac{OSIRa\times EDa\times EFa}{BWa\times ATca})\times ABSo\times 10^{-6}$
	Dermal contact		$DCSERca=\frac{SAEc\times SSARc\times EFc\times EDc\times Ev\times ABSd}{BWc\times ATca}\times 10^{-6}+\frac{SAEa\times SSARa\times EFa\times EDa\times Ev\times ABSd}{BWa\times ATca}\times 10^{-6}$
	Breath inhalation	Soil granule	$PISERca=\frac{TSP\times DAIRc\times EDc\times PIAF\times(fspo\times EFOc+fspi\times EFIc)}{BWc\times ATca}\times 10^{-6}+\frac{TSP\times DAIRa\times EDa\times PIAF\times(fspo\times EFOa+fspi\times EFIa)}{BWa\times ATca}\times 10^{-6}$
		Surface soil ourdoors	$IoVERca1=VF_{suroa}\times(\frac{DAIRc\times EFOc\times EDc}{BWc\times ATca}+\frac{DAIRa\times EFOa\times EDa}{BWa\times ATca})$
		Sublayer soil ourdoors	$IoVERca2=VF_{suboa}\times(\frac{DAIRc\times EFOc\times EDc}{BWc\times ATca}+\frac{DAIRa\times EFOa\times EDa}{BWa\times ATca})$
		Sublayer soil indoors	$IiVERca=VF_{subia}\times(\frac{DAIRc\times EFIc\times EDc}{BWc\times ATca}+\frac{DAIRa\times EFIa\times EDa}{BWa\times ATca})$
Cancer risk (unitless)			$CR_n=(OISER_{ca}\times SF_o+DCSER_{ca}\times SF_d+PISER_{ca}\times SF_i+IoVER_{ca1}\times SF_i)\times C_{sur}+(IoVER_{ca2}+IiVER_{ca})\times C_{sub}\times SF_i$
Non-cancer effect	Oral ingestion		$OISERnc=\frac{OSIRc\times EDc\times EFc\times ABSo}{BWc\times ATnc}\times 10^{-6}$
	Dermal contact		$DCSERnc=\frac{SAEc\times SSARc\times EFc\times EDc\times Ev\times ABSd}{BWc\times ATnc}\times 10^{-6}$
	Breath inhalation	Soil granule	$PISERnc=\frac{TSP\times DAIRc\times EDc\times PIAF\times(fspo\times EFOc+fspi\times EFIc)}{BWc\times ATnc}\times 10^{-6}$
		Surface soil ourdoors	$IoVERnc1=VF_{suroa}\times\frac{DAIRc\times EFOc\times EDc}{BWc\times ATnc}$
		Sublayer soil ourdoors	$IoVERnc2=VF_{suboa}\times\frac{DAIRc\times EFOc\times EDc}{BWc\times ATnc}$
		Sublayer soil indoors	$IiVERnc=VF_{subia}\times\frac{DAIRc\times EFIc\times EDc}{BWc\times ATnc}$
Hazard quotient (unitless)			$HQ_n=(\frac{OISER_{nc}}{RfD_o}+\frac{DCSER_{nc}}{RfD_d}+\frac{PISER_{nc}+IoVER_{nc1}}{RfD_i})\times C_{sur}+(\frac{IoVER_{nc2}+IiVER_{nc}}{RfD_i})\times C_{sub}$
Migration parameters estimates*	Surface soil content of pollutant diffusion in the air outdoors		$VF_{suroa}=\frac{W_{dw}\times\rho_b\times d}{U_{air}\times\delta_{air}\times\tau}\times 10^3$
	Sublayer soil content of pollutant diffusion in the air outdoors		$VF_{suboa}=\frac{H'\times\rho_b}{(\theta_{avs}\times H'+\theta_{wvs}+K_{oc}\times f_{oc}\times\rho_b)\times(1+\frac{U_{air}\times\delta_{air}\times L_s}{D_s^{eff}\times W_{dw}})}\times 10^3$
	Sublayer soil content of pollutant diffusion in the air indoors		$VF_{subia}=\frac{\frac{H'\times\rho_b}{\theta_{avs}\times H'+\theta_{wvs}+K_{oc}\times f_{oc}\times\rho_b}\times\frac{D_s^{eff}/L_s}{ER\times L_B}}{1+\frac{D_s^{eff}/L_s}{ER\times L_B}+\frac{D_s^{eff}/L_s}{D_{crack}^{eff}/L_{crack}\times\eta}}\times 10^3$

*Calculated by practical survey and U.S. EPA recommendation data.

Table 4 Exposure parameters

Exposure parameters		Residential land	
		Children	Children
BW	Body weight (kg)	14.4	53.1
ED	Exposure duration (a)	6	24
EF	Exposure frequency (d/a)	365	365
OSIR	Oral soil ingestion rate (mg/d)	200	100
DAIR	Daily atmosphere inhalation rate (m^3/d)	7.5	15
SAE	Skin surface exposed (cm^2)	2 350	5 700
SSAR	Skin surface adherence rate (mg/cm^2)	0.2	0.07
ATca	Average time of cancer (d)	26 280	
ATnc	Average time of non-cancer (d)	2 190	
EFI	Exposure frequency indoor (d/a)	274	
EFO	Exposure frequency outdoor (d/a)	91	
fspi	Particulate matter proportion in indoor air from soil (unitless)	0.8	
fspo	Particulate matter proportion in outdoor air from soil (unitless)	0.5	
Ev	Event frequency (events/day)	1	
TSP	Total suspended particulates (mg/m^3)	0.30	
PIAF	Internal proportion of soil particulates inhalation (unitless)	0.75	

Table 5 Toxicology parameters of pollutants

Toxicity parameters	Units	Benzene	Benzo(a)anthracene	BaP
Oral carcinogenic slope factor, Sfo	$(mg \cdot kg^{-1} \cdot d^{-1})^{-1}$	5.50E-02	7.30E-01	7.30E+00
Inhalation carcinogenic slope factor, Sfi	$(mg \cdot kg^{-1} \cdot d^{-1})^{-1}$	2.73E-02	3.90E-01	3.90E+00
Dermal carcinogenic slope factor, Sfd	$(mg \cdot kg^{-1} \cdot d^{-1})^{-1}$	5.67E-02	2.35E+00	2.35E+01
Oral reference dose, RfDo	$mg \cdot kg^{-1} \cdot d^{-1}$	4.00E-03	2.00E-04	2.00E-05
Inhalation reference dose, RfDi	$mg \cdot kg^{-1} \cdot d^{-1}$	--	7.00E-07	7.00E-08
Dermal reference dose, RfDd	$mg \cdot kg^{-1} \cdot d^{-1}$	4.00E-03	--	--
Oral absorption fraction, ABSo	--	1	1	1
Dermal absorption fraction, ABSd	--	1.00E-02	1.30E-01	1.30E-01

indirect carcinogen that will cause damages to stomach, lung, liver, skin and other organs. Therefore, the present study aims at assessing site-specific pollution from both carcinogenic risk and non-carcinogenic hazard in the hope of reducing relevant health risks somehow.

Since China has not yet constructed a complete pollutants toxicity database for human health, the major reference of the pollutants toxicity parameters is the Integrated Risk Information System of the U.S. EPA. Table 5 shows the antimony toxicological parameters.

RISK CHARACTERIZATION

Carcinogens risk assessment concerned with factors such as exposure dose and pollutant concentration in surface soil and carcinogenic slope. While non-carcinogenic risks of the contaminated soil can be illustrated with hazard quotient (HQ), which is the specific value between the product of exposure dose pollutant concentration in surface soil and toxicological reference dose. Relevant formulas are shown in Table 3. For the computation, EPA suggests that the 95% upper confidence limit should be adopted as the concentration value. Therefore, the soil pollutants concentration are: benzene 3.48 mg/kg, benzo(a)pyrene 1.88 mg/kg and benzo(a)anthracene 11.79 mg/kg respectively. Human health risks and hazard quotients of each exposure pathway are included in the Table 6.

Because the distribution of pollutants in the soil is uneven, we should deal with pollutants of different points to work out the superposition health risks so as to guide subsequent soil restoration practice effectively. Figure 3 is superposition risks distribution for the pollutants in different areas bounded by the monitoring grids.

The internationally recognized acceptable non-carcinogenic hazard quotient for site-specific risk level is HQ≤1. The acceptable carcinogenic risk varies from country to country. For example, In Netherlands, it is quite a loose one at 10^{-4}. In the UK, 10^{-5} is applied in actual practice. The U.S. ASTM recommends 10^{-6} as the single pollutant risk target and 10^{-4} as the accumulated contaminants risk target. Since China is a developing country, the study uses 10^{-6} and 10^{-5} as sole pollutant risk target and accumulated polluttants risk target, taking into account such factors as the restoration expenses and variations in terms of physique constitution of residents from those of European and American countries. With the above-mentioned standard for health risks assessment, the results, show the carcinogenic risks of benzo(a)anthracene and benzo(a)pyrene exceed 10^{-5}, which means carcinogenic risks and non-carcinogenic hazards of the site both have exceeded the acceptable risk levels, and the site needs restoration before reutilization as a result.

Table 6 Risks assessment results of soil pollutants

Health effect	Cancer risk probability			Non-cancer hazard quotients		
Pathways	Oral ingestion	Dermal contact	Breath inhalation	Oral ingestion	Dermal contact	Breath inhalation
Benzene	3.42E-07	1.03E-08	1.23E-08	1.21E-02	2.84E-04	--
Benzo(a)pyrene	2.45E-05	3.0E-05	6.87E-06	1.305	--	11.45
Benzo(a)anthracene	1.54E-05	1.88E-5	4.9E-08	0.819	--	1.88
Totally	9.59×10^{-5}			15.46		

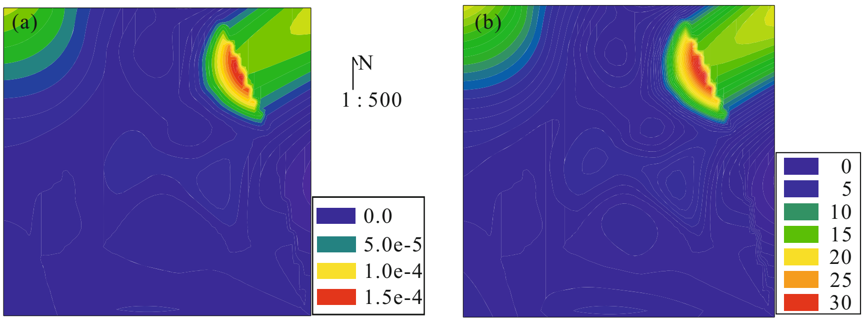

Figure 3. Distribution of site superposition risks. (a) Cancer risk probability; (b) non-cancer hazard quotients.

CONCLUSIONS

The monitoring results show that the soil has been polluted by benzene, benzo(a)pyrene and benzo(a)anthracene due to the long-term production activities. In some parts, the pollutants density in the soil has exceeded Chinese standard of soil quality for exhibition sites and the screening values of health risks assessment for soil contaminants in residential land in standards issued by U.S. EPA.

The study site has been planned to be exploited as residential land, major sensitive group is identified as the residents living on the site in the future. Based on movement characteristics of contaminants in soil environment, the main exposure pathways of the sensitive groups are oral ingestion, dermal contact and breath inhalation of contaminated soil particles, outdoor topsoil pollutants vapor, outdoor subsoil pollutnts vapor and indoor subsoil pollutants vapor.

The cumulative cancer risk and the non-cancer hazard quotient of the site soil are 9.59×10^{-5} and 15.46, exceeding acceptable cumulative risk targets of 10^{-5} and 1. The single cancer risk and the non-cancer hazard quotient of benzo(a)pyrene, benzo(a)anthracene have exceeded acceptable single risk targets of 10^{-6} and 1, respectively. The soil of the site needs restoration before reutilization.

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