Journal of Hazardous Materials 172 (2009) 316–323

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

The Health Risk Assessment of Pb and Cr leachated from fly ash monolith landfill

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ARTICLE INFO

Article history:
Received 3 February 2009
Received in revised form 3 July 2009
Accepted 3 July 2009
Available online 10 July 2009

Keywords:
Fly ash monolith
Sanitary landfill
Landfill structure
Health risk assessment
Uncertainty

ABSTRACT

As of 2004, nearly two hundred thousand tons of fly ash monoliths are created each year in Taiwan to confine heavy metals for reducing the leaching quantity by precipitation. However, due to abnormal monolith fracture, poorly liner quality or exceeding usage over designed landfill capacity, serious groundwater pollution of the landfills has been reported. This research focuses on Pb and Cr leaching from monolithic landfill to assess the risk of groundwater pollution in the vicinity. The methodology combines water budget simulations using HELP model with fate and risk simulations using MMSOILS model for 5 kinds of landfill structures and 2 types of leaching models, and calculates the risk distribution over 400 grids in the down gradient direction of groundwater.

The results demonstrated that the worst liner quality will cause the largest risk and the most significant exposure pathway is groundwater intake, which accounted for 98% of the total risk. Comparing Pb and Cr concentrations in the groundwater with the drinking water standards, only 14.25% of the total grids are found to be under 0.05 mg/L of Pb, and over 96.5% of the total grids are in the safety range of Cr. It indicates that Pb leaching from fly ash monolithic landfills may cause serious health risks.

Without consideration of the parameters uncertainty, the cancer and noncancer risk of Pb with the sanitary landfill method was 4.23E−07 and 0.63, respectively, both under acceptable levels. However, by considering the parameters uncertainty, the non-carcinogenic risk of Pb became 1.43, exceeding the acceptable level. Only under the sanitary landfill method was the hazard quotient below 1. It is important to use at least the sealed landfill for fly ash monoliths containing lead to effectively reduce health risks.

1. Introduction

Incineration is currently the primary municipal waste treatment method used in a lot of countries. Although incineration can decrease the amount of solid waste, it also produces great amount of solid residue which becomes the main concern of waste management. Bottom ash, the major portion of incineration residue, is considered non-hazardous and can be reused. But the fly ash constitutes a potential health hazard because it contains toxic metals such as lead, cadmium, copper and zinc as well as small amounts of dioxins and furans. Therefore, toxic fly ash has to be solidified/stabilized before final disposal. To the end of 2004, nearly 200 thousand tons of fly ash solidified products were created each year in Taiwan. Even though the Environmental Regulations request the solidified material must meet the quality requirements of 10 kg/cm^2 single axial compressive strength and Toxicity Characteristic Leaching Procedure (TCLP) leaching test before being disposed in the landfill sites, it is necessary to further understand the leaching probability of toxic material from the fly ash monoliths and its long-term impacts on the environment and human health.

There are several papers that present the assessments of environmental contamination (mainly groundwater) and human health risks of waste disposal in recent years. Sophocleous et al. [1] performed a modeling study using Hydrologic Evaluation of Landfill Performance (HELP) model and Multimedia Exposure Assessment Model (MULTIMED) for a small landfill of Wallace County in western Kansas semiarid area. They found that requiring landfill cover, leachate collection system, and compact soil liner will reduce leachate production by 56%. However, the time elapsed between landfill emplacement and leachate pollutant detection at the point of compliance is relatively long, at least 70 years. Zubair [2] used the USEPA's Composite Model for Leachate Migration with Transformation Products (EPACMTP) in conjunction with the HELP and the MINTEQA2 to assess the potential groundwater exposure due to lead associated with disposal of lead-based paint debris managed in landfills. The results suggested the peak receptor well concentration would be between zero and 0.005 mg/l (one-third of the 0.015 mg/L safe drinking water action level for lead in tap water) in approximately 97.38% of the cases and only in less than 4.5% of the cases would the receptor well lead concentration exceed the regulatory action level. Bocanegra et al. [3] conducted the risk assessment for leachate contamination of groundwater at two landfills utilized for the disposal of solid urban wastes in Mar del Plata, Argentina. The HELP and the Visual MODFLOW (Modular Three-dimensional

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doi:10.1016/j.jhazmat.2009.07.013

Groundwater Flow Model) codes were applied to simulate and predict the flux and transport of the chloride ion contaminated plume, based on the border condition for chloride concentration 70 mg/L. The study showed that the concentration of chlorine ion varied from 250 to 900 mg/L and might exceeds 600 mg/L, the allowable maximum level of the World Health Organization (WHO). Ho et al. [4] used the Framework for Risk Analysis in Multimedia Environmental Systems (FRAMES) to develop a probabilistic, risk-based performance-assessment method for selection, design, and monitoring of long-term covers of the landfills. Liu et al. [5] also built an integrated simulation-assessment modeling approach to analyze environmental risks of groundwater contamination at the waste landfills. This approach incorporated an analytical groundwater solute transport model, an exposure dose model, and a fuzzy risk assessment model. It was proved to be useful for supporting the risk management and remediation decisions. Pontedeiro et al. [6] used the multimedia transportation and multiple pathway exposure models to conduct the long-term prediction of the environmental impact and health risks of landfills used for the disposal of solid radioactive wastes resulting from the mineral industry. In 2007, Xiaoli et al. found that the heavy metals became stabilized in the landfill and only a small quantity of heavy metals was released, although the refuse contained high concentrations of potentially toxic heavy metals [7].

Most of the previous studies about the landfill contamination focused on municipal wastes; but the health effects of the toxic materials in the fly ash monoliths leaching problems were rarely explored. Recently, incinerator ash disposed landfill has been brought to public attention. Lo et al. indicated that the concentrations of heavy metals (Cd, Cr, Cu, Ni and Zn) were found to be less than 1 mg/L, except for Pb, which reached 2 mg/L in the incinerator ash co-disposed landfill in Taiwan [8]. Even in Japan, Cu, Pb, As, Zn, Fe, etc. were leached from the co-disposed landfill in Inanc et al.'s study [9]. In order to further quantify the harm from fly ash monolith landfill, the main objective of this paper is to describe an assessment methodology to foresee the risk of landfills used for the disposal of fly ash monoliths. In 2005, the leachate of fly ash monoliths landfills in Taiwan was analyzed in Taiwan EPA. The result clearly showed that Cr is leached the most easily, and Pb concentration is the highest in the leachate. The concentration of Cr in the leachate is about 0.031–0.238 mg/L; Pb is about 0.15–1.46 mg/L. In contrast, Taiwan EPA monitored the leaching from all landfills in Taiwan, showing that Cr concentration was less than 0.002 mg/L, and Pb is 0.01–1.37 mg/L [10]. Therefore the case study in this research focuses on Pb and Cr leachated from the monolithic landfill to assess the health risks of groundwater pollution. The methodology combines water budget simulations using HELP model and risk simulations using MMSOILS model for 5 kinds of landfill structures and 2 types of leaching models, and calculates the risk distribution over 400 grids in the down gradient direction of groundwater. The impacts of parameters uncertainty are also taken into consideration.

2. Methodology

The potential environmental impact of heavy metal leaching from monolithic landfill includes contamination of soil and groundwater. This study integrates simple leaching models, HELP model, and MMSOILS model to analyze human health risks of groundwater contamination caused by waste landfill leakage. The simple leaching model is used for simulating Pb and Cr concentrations in the monolithic leachate, and the HELP model is used for assessing the volume of leachate produced from the landfill. The outputs of this model can be used as the inputs for MMSOILS to estimate the human exposure and health risks associated with releases of Pb and Cr contamination.

Different simulations are performed to evaluate five different landfill structures. The landfill scenarios include the common sanitary landfill (F), the sealed landfill (C), the landfill which used the worst quality liner (W), the landfill which used the best quality liner (B) and the overcapacity landfill (O). Compared to the common landfill, the sealed landfill has stricter conditions including permeability coefficients, thickness of cover, and treatment of leaching. The quality of liners is reflected by the degrees of geomembrane placement quality and installation defects. Finally, the overcapacity landfill means longer landfill duration, which increases the amount of disposal and the landfill area. Relevant parameter information is specified in Table 1. In addition to the variation of landfill types, two kinds of leaching concentrations are considered in this study: the annual average concentration calculated by the simple leaching model and the maximum concentration estimated from the annual landfill quantity of Pb and Cr. The former is close to the real annul average concentration of leachate. However, the health risk based on the latter is the more conservative one. A brief overview of the models and risk quantification methods follows.

2.1. Models

The simple leaching model used to predict long-term average leaching concentration of Pb and Cr from monoliths in this study

Table 1
The parameters of five landfill scenarios used in the HELP model.

Parameter	F	C	W	B	O	Data source
Landfill area (ha)	3.64	3.64	3.64	3.64	5.46	Site-specific
Percent of area where runoff is possible (%)	30%	30%	30%	30%	30%	[15]
Slope (%)	30%	30%	30%	30%	30%	Site-specific
Slope length (m)	150	150	150	150	150	Site-specific
Soil texture[a]	4	4	4	4	4	Site-specific
Vegetation[a]	3	3	3	3	3	Site-specific
Curve number values for runoff simulation	61.1	61.1	61.1	61.1	61.1	Model evaluation
Initial moisture (v/v)	0	0	0	0	0	[15]
Drainage length (m)	500	500	500	500	500	Site-specific
Drain slope (%)	3%	3%	3%	3%	3%	Site-specific
Subsurface inflow (mm/y)	0	0	0	0	0	[15]
Geomembrane pinhole density (#/ha)	10	10	20	1	10	Assumption
Geomembrane installation defects (#/ha)	10	10	20	1	10	Assumption
Geomembrane placement quality	3	3	5	1	3	Assumption
Geotextile transmissivity (cm^2/s)	3.0E−12	3.0E−12	3.0E−12	3.0E−12	3.0E−12	[15]

Denotation: (F) the general sanitary landfill, (C) the sealed landfill, (W) the landfill which used the worst quality liner, (B) the landfill which used the best quality liner, and (O) overcapacity landfill.

[a] The value of soil texture, 4, represents loamy sand; the value of vegetation, 3, represents usual vegetation.

was derived from ANS-16.1 procedure [11]. It uses modified Fick's second law with mass balance concept to describe the physical diffusion and chemical reactions underlying leaching process.

The HELP model was developed by the U.S. Army Engineer Waterways Experiment Station [12]. The model is commonly used to predict the seasonal generation, percolation and drainage of landfill leachate due to precipitation and infiltration. The hydrological results generated by the HELP model include time series information such as precipitation, evapotranspiration, and runoff. Therefore, this study uses HELP model to calculate the volume of leachate produced from monolithic landfill. The parameters required for the model can be divided into two categories: meteorological data and landfill structural data, including monolithic data. Table 1 presents all the parameters of five landfill scenarios used in the HELP model.

MMSOILS was developed by the USEPA [13] and is currently available from EPA's web site in Version 4.0. It can estimate the human exposure and health risks associated with releases of contamination from leachate of landfills. MMSOILS provides a multimedia tool that simulates chemical transport in the atmosphere, soil, surface water, groundwater, and the food chain by using finite difference method. The human exposure pathways considered in MMSOILS include: soil ingestion, air inhalation of volatiles and particulates, dermal contact, ingestion of drinking water, consumption of fish, consumption of plants grown in contaminated soil, and consumption of animals grazing on contaminated pasture. MMSOILS also includes a Monte Carlo mechanism for propagating parameter uncertainties into estimates of exposure and risk.

2.2. Risk quantification

According to Risk Assessment Guidance for Superfund (RAGS), Volume I, Part A [14], the hazards are divided into two groups for calculating numerical estimates of risk: carcinogenic and noncarcinogenic. Although the calculation procedures differ for carcinogenic and noncarcinogenic effects, both sets of procedures assume dose and exposure additivity. The risks or hazard indices for the case of simultaneous exposures to several chemicals from a variety of sources by more than one exposure pathway can be summed up [14].

Pb leaching from fly ash monolithic landfills is a carcinogen and Cr is a noncancer hazard. The slope factors and reference doses (RfD) for Pb and Cr needed to quantify risk is available in California EPA's CalTOX database (California EPA, 1993). The potential for human noncarcinogenic effect of a noncancer hazard through a single exposure pathway is evaluated by the hazard quotient (HQ). The hazard quotient is the ratio of the average daily intake of a contaminant per unit body weight to an acceptable reference dose, as Eq. (1):

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{\mathrm{RfD}} > 1 \tag{1}$$

where HQ is the hazard quotient, CDI is the chronic daily intake for a toxicant averaged over 70 years (mg/kg day), and RfD is the chronic reference dose for a toxicant (mg/kg day). If the hazard quotient is greater than 1, then adverse health effects are possible.

To assess the overall potential for noncarcinogenic effects posed by more than one chemical through several pathways (e.g. ingestion, inhalation, and dermal contact), a hazard index (HI) approach has been adopted, as described below:

$$\mathrm{HI} = \sum \mathrm{HQ}_{\mathrm{ingestion}} + \sum \mathrm{HQ}_{\mathrm{inhalation}} + \sum \mathrm{HQ}_{\mathrm{dermal\ contact}} \tag{2}$$

$$\sum \mathrm{HQ}_{(\mathrm{exposure\ pathway})} = \frac{\sum \mathrm{CDI}_{(\mathrm{exposure\ pathway})}}{\mathrm{RfD}_{(\mathrm{exposure\ pathway})}} \tag{3}$$

where HI is the hazard index, $\Sigma\mathrm{HQ}_{\mathrm{ingestion}}$ the sum of the hazard quotients for ingesting contaminated drinking water, food and soil, $\Sigma\mathrm{HQ}_{\mathrm{inhalation}}$ the sum of the hazard quotients for inhaling toxicants from air, and $\Sigma\mathrm{HQ}_{\mathrm{dermal\ contact}}$ is the sum of the hazard quotients for contacting contaminated soil and water by skin.

If the hazard index exceeds 1, there would be concern for potential noncancer health effects. A hazard index value of 1.0 or less than 1.0 indicates that no adverse human health effects (noncancer) are expected to occur.

For carcinogens, risks are estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen [14]. This carcinogenic risk equation is described below:

$$\mathrm{Risk} = \mathrm{CDI} \times \mathrm{SF} \tag{4}$$

where Risk is a unitless probability of an individual developing cancer, CDI the chronic daily intake averaged over 70 years (mg/kg day), and SF is the slope factor for the concern toxicant $(\mathrm{mg/kg\ day})^{-1}$. Risks for multiple exposure pathways also can be combined for a single exposed individual or group of individuals (5):

$$R_{\mathrm{total}} = \sum R_{\mathrm{ingestion}} + \sum R_{\mathrm{inhalation}} + \sum R_{\mathrm{dermal\ contact}} \tag{5}$$

where R_{total} is total exposure cancer risk, $\Sigma R_{\mathrm{ingestion}}$ the risk estimated for ingesting contaminated drinking water, food and soil, $\Sigma R_{\mathrm{inhalation}}$ the risk estimated for inhaling toxicants from air, and $\Sigma R_{\mathrm{dermal\ contact}}$ is the risk estimated for contacting contaminated soil and water by skin.

In general, total cancer risk value of 10^{-6} is the upper limit of acceptable cancer risk. If total cancer risk exceeds 10^{-6}, meaning at least one in a million individuals will develop cancer due to lifetime exposure to the concerned toxicant, the area evaluated is evidently under the risk of getting cancer.

2.3. Assumptions and limitations

Since this study only considers groundwater contamination of Pb and Cr, several assumptions and limitations of MMSOILS must be acknowledged.

1. A single unconfined aquifer with uniform thickness is modeled. The saturated, porous medium properties are isotropic and homogeneous. The module cannot be used to simulate transport in fractured media unless the fractured medium is represented as an equivalent porous formation.
2. The regional velocity field in the aquifer is constant over time, uniform at all points, and unidirectional in the positive x-direction. This implies that the recharge through the facility and into the groundwater plume is small compared to the natural (regional) flow. However, there will be discrepancy between simulation results and realities, if groundwater aquifer is recharged by direct infiltration of rainwater from the ground surface.
3. Contaminant degradation/transformation follows the first-order rate law and is restricted to biodegradation and hydrolysis. This assumption is conservative since it neglects degradation due to other mechanisms such as oxidation, reduction, etc.
4. Contaminant sorption follows a linear adsorption isotherm. Adsorption takes place instantaneously and the adsorbed phase is in local equilibrium.
5. The initial contaminant concentrations of any contaminants in the aquifer are assumed zero.

The following are the list of assumptions and limitations inherent in the discussion of relationship between the way of ingestion and the amount of intake for food contaminated through groundwater (e.g. consumption of vegetables irrigated by contaminated

Table 2
Pb risk levels with average leaching quantity.

	Drinking water	Meat	Milk	Shower	Avg-Total
Cancer risk					
W	1.25E−05	7.84E−09	1.63E−10	7.23E−09	1.25E−05
O	5.55E−08	3.49E−11	7.27E−13	3.22E−11	5.55E−08
F	3.69E−08	2.32E−11	4.83E−13	2.14E−11	3.69E−08
C	8.67E−09	5.45E−12	1.14E−13	5.03E−12	8.68E−09
B	4.65E−10	2.92E−13	6.09E−15	2.69E−13	4.65E−10
Noncancer risk					
W	1.87E+01	1.17E−02	2.45E−04	1.08E−02	1.87E+01
O	8.30E−02	5.22E−05	1.09E−06	4.81E−05	8.31E−02
F	5.52E−02	3.47E−05	7.23E−E−07	3.20E−05	5.53E−02
C	1.30E−02	8.16E−06	1.70E−07	7.52E−06	1.30E−02
B	6.95E−04	4.37E−07	9.11E−09	4.03E−07	6.96E−04

Denotation: (F) the general sanitary landfill, (C) the sealed landfill, (W) the landfill which used the worst quality liner, (B) the landfill which used the best quality liner, and (O) overcapacity landfill; Avg-Total: the total risks with average leaching quantity.

groundwater or ingestion of meat and milk produced in the same area):

1. The human exposure assessment of the landfill usually faces with two problems unless the landfill has been gone through the environmental impact assessment and detail on-site investigation before its establishment. First, is the downstream flow direction of groundwater constant? Second, it is hard to get the information of population's activities (e.g. irrigation and pasturage with groundwater) at few kilometers downstream of groundwater. Under this circumstance, it is assumed that the residents living around the target landfill do use the contaminated groundwater for consumption of home grown produce and in the livestock industry.
2. Ingestion rates used in exposure assessment are based on the multimedia risk assessment model, CalTOX [15], and Guideline of Health Risk Assessment for Soil and Groundwater Containment Sites in Taiwan [16].
3. The leaching amounts of Pb and Cr calculated from single leaching model and the HELP model are used as the pollutant amounts of groundwater in MMSOILS model to simulate risk distribution over 400 grids (grid spacing is 100 m) in the down gradient direction of groundwater. It is supposed that adult inhabitants living in a grid use local groundwater as the direct source of drinking water 350 days per year in 24 years of their 70-year-life spans [16]. They also consume the meat of beef cattle and the milk of dairy cattle which are raised with local contaminated groundwater. Another exposure route is dermal contact by using groundwater for bathing and showering.

3. Case description

A fly ash monolithic landfill, referred to as SL, in northern Taiwan was chosen as the study object. It is located just beside the Dahan River. The lowest groundwater level contour coincides with the flow direction of Dahan River around Hsinchuang area where it merges with the Dansuie River [17].

Table 3
Noncancer risk levels of Cr with average leaching quantity.

	Drinking water	Meat	Milk	Shower	Avg-Total
W	5.40E−02	6.50E−05	7.78E−06	3.27E−05	5.41E−02
O	2.31E−04	2.78E−07	3.33E−08	1.40E−07	2.31E−04
F	1.60E−04	1.92E−07	2.30E−08	9.66E−08	1.60E−04
C	3.75E−05	4.52E−08	5.41E−09	2.27E−08	3.76E−05
B	2.01E−06	2.42E−09	2.90E−10	1.22E−09	2.01E−06

Denotation: (F) the general sanitary landfill, (C) the sealed landfill, (W) the landfill which used the worst quality liner, (B) the landfill which used the best quality liner, and (O) overcapacity landfill; Avg-Total: the total noncancer risks with average leaching quantity.

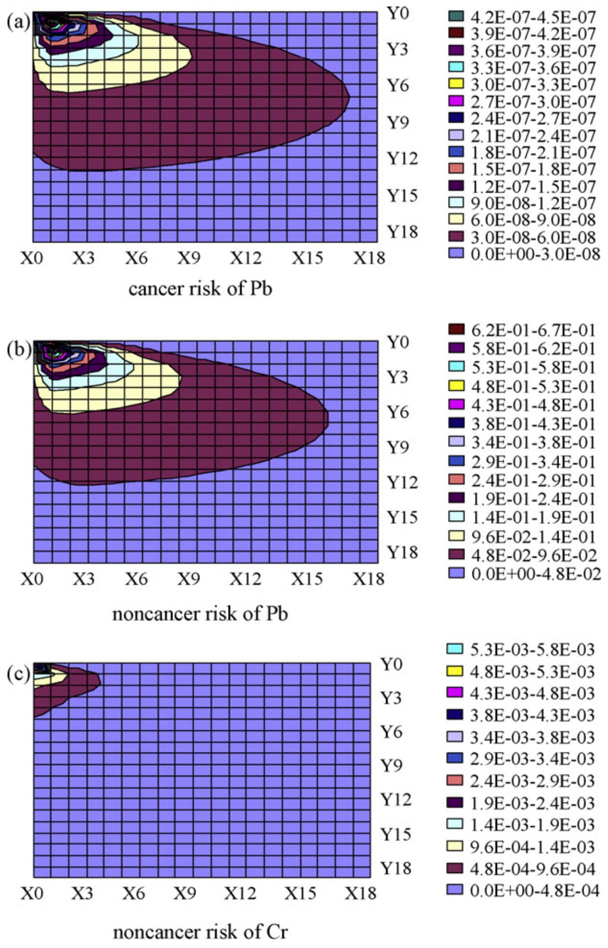

Fig. 1. The cancer risk contours of the sanitary landfill with average leaching quantity.

The landfill covers an area of 3.64 ha and consists of a 78 m × 70 m waste cell. It is assumed that leak position of heavy metal Pb and Cr is just right below the SL. The closest one is in 500 m to SL. Families living there would be exposed to the pollutants by using groundwater as direct source of drinking, showering, and pasturing.

Study boundary is set as a grid consisting of 20 grid points (each is 100 m apart) on y-direction which is parallel to groundwater flow toward downstream direction and 20 grid points on x-direction which is vertical to the groundwater flow. There are totally 400 grid points in the study area and each grid point has a receptor well. Background levels of Pb and Cr in the groundwater of study area are assumed zero before leachate is generated from the SL site. Other sources of Pb and Cr are excluded during 100 years simulation time period.

4. Results

4.1. Risk analysis by using average leaching quantity

Tables 2 and 3 represent the human health risks estimated by using average leaching quantities of Pb and Cr. Fly ash monolithic landfill with bad liner quality obviously has higher health risks. Comparing the average risk of all grids, SL site with the worst liner system has the highest cancer risk level (1.25E−05, which is higher

Table 4
Pb risk levels with the maximum leaching quantity.

	Drinking water	Meat	Milk	Shower	Max-Total
Cancer risk					
W	9.88E−03	6.21E−06	1.29E−07	5.73E−06	9.89E−03
O	3.15E−05	1.98E−08	4.13E−10	1.83E−08	3.16E−05
F	2.88E−05	1.81E−08	3.77E−10	1.67E−08	2.88E−05
C	2.72E−05	1.71E−08	3.56E−10	1.58E−08	2.72E−05
B	3.67E−07	2.31E−10	4.81E−12	2.13E−10	3.67E−07
Noncancer risk					
W	1.48E+04	9.30E+00	1.94E−01	8.57E+00	1.48E+04
O	4.72E+01	2.97E−02	6.18E−04	2.74E−02	4.73E+01
F	4.31E+01	2.71E−02	5.64E−04	2.50E−02	4.31E+01
C	4.07E+01	2.56E−02	5.33E−04	2.36E−02	4.07E+01
B	5.49E−01	3.45E−04	7.19E−06	3.18E−04	5.50E−01

Denotation: (F) the general sanitary landfill, (C) the sealed landfill, (W) the landfill which used the worst quality liner, (B) the landfill which used the best quality liner, and (O) overcapacity landfill; Max-Total: the total risks with the maximum leaching quantity.

Table 5
Noncancer risks of Cr with the maximum leaching quantity.

	Drinking water	Meat	Milk	Shower	Max-Total
W	1.89E+01	2.27E−02	2.72E−03	1.14E−02	1.89E+01
O	5.79E−02	6.98E−05	8.35E−06	3.51E−05	5.81E−02
F	5.56E−02	6.71E−05	8.02E−06	3.37E−05	5.57E−02
C	6.27E−02	7.56E−05	9.04E−06	3.80E−05	6.29E−02
B	7.01E−04	8.44E−07	1.01E−07	4.24E−07	7.02E−04

Denotation: (F) the general sanitary landfill, (C) the sealed landfill, (W) the landfill which used the worst quality liner, (B) the landfill which used the best quality liner, and (O) overcapacity landfill; Max-Total: the total risks with the maximum leaching quantity.

than the acceptable level, 1E−06) and noncancer risk value (18.7, which is also higher than the acceptable value, 1) of Pb. The risk level of the best liner quality is lower than that of the worst liner quality for about 4–5 orders of magnitude. As for the other three landfill structures, using risk levels caused by Pb as an example, overcapacity landfill has the highest risk level (5.55E−08) and sealed landfill has the lowest (8.68E−09). Therefore, multi-layer prevention measure and under design capacity usage can decrease the human health risks of landfill.

Compared to pathways of meat consumption, milk ingestion and showering, direct ingestion of Pb and Cr contaminated groundwater is responsible for the highest health risk. According to the simulation results showed in Table 3, it is clear that Cr in the leachate produces the acceptable health risks to adult inhabitants living in the grid over their 70-year-life spans for all exposure pathways.

Fig. 1 presents the risk contours of the 400 grid points for the sanitary landfill with average leaching quantity. For cancer risk of Pb, most of the grid points have a risk level under 3.0E−08. The highest risk level is 4.5E−07. For noncancer risk of Pb, most of the HQ values are smaller than 4.8E−02. The highest HQ value is 0.67, still less than 1. As for Cr, the area of high HQ values is much smaller than that of Pb. The highest HQ value is only 5.8E−03 for Cr.

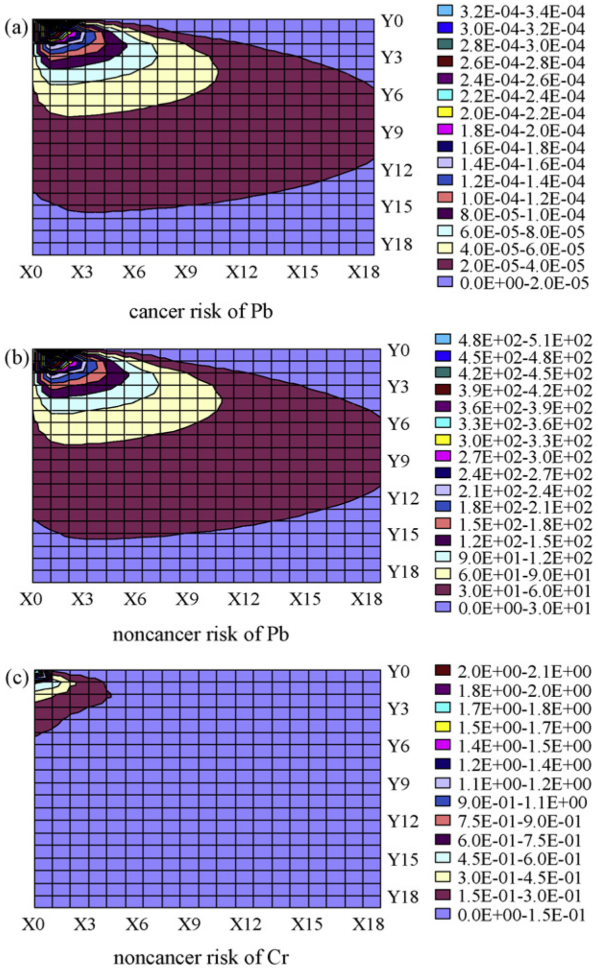

Fig. 2. The noncancer risk contours of the sanitary landfill with the maximum leaching quantity.

4.2. Risk analysis by using the maximum leaching quantity

Tables 4 and 5 list the average human health risks, which were estimated by using the annual maximum leaching quantities of Pb and Cr, of different exposure pathways. SL site with the worst liner system (W) has the highest cancer and noncancer risk levels of Pb (9.89E−03 and 1.48E+04, respectively); same for Cr (18.9). Its risk values are much higher than those of the other kinds of landfill. The results are similar to the ones estimated with average leaching quantity.

Compared to pathways of meat consumption, milk ingestion and showering, direct ingestion of Pb and Cr contaminated groundwater is responsible for the highest health risk. Food chain exposure pathways and dermal contact contribute only a small portion to the overall risk. The risk contribution listed in decreasing order

Table 6
Numbers of grid points of meeting the groundwater drinking standards.

Scenario	Pb-Max		Cr-Max		Pb-Avg		Cr-Avg	
	Grid points	Pass rate	Grid points	Pass rate	Grid points	Pass rate	Grid points	Pass rate
W	18	4.5%	19	4.75%	57	14.25%	386	96.5%
O	29	7.25%	385	96.25%	400	100%	400	100%
F	29	7.25%	386	96.5%	400	100%	400	100%
C	30	7.5%	387	96.75%	400	100%	400	100%
B	400	100%	400	100%	400	100%	400	100%

Denotation: (F) the general sanitary landfill, (C) the sealed landfill, (W) the landfill which used the worst quality liner, (B) the landfill which used the best quality liner, and (O) overcapacity landfill; (Avg) the average leaching quantity, (Max) the maximum leaching quantity.

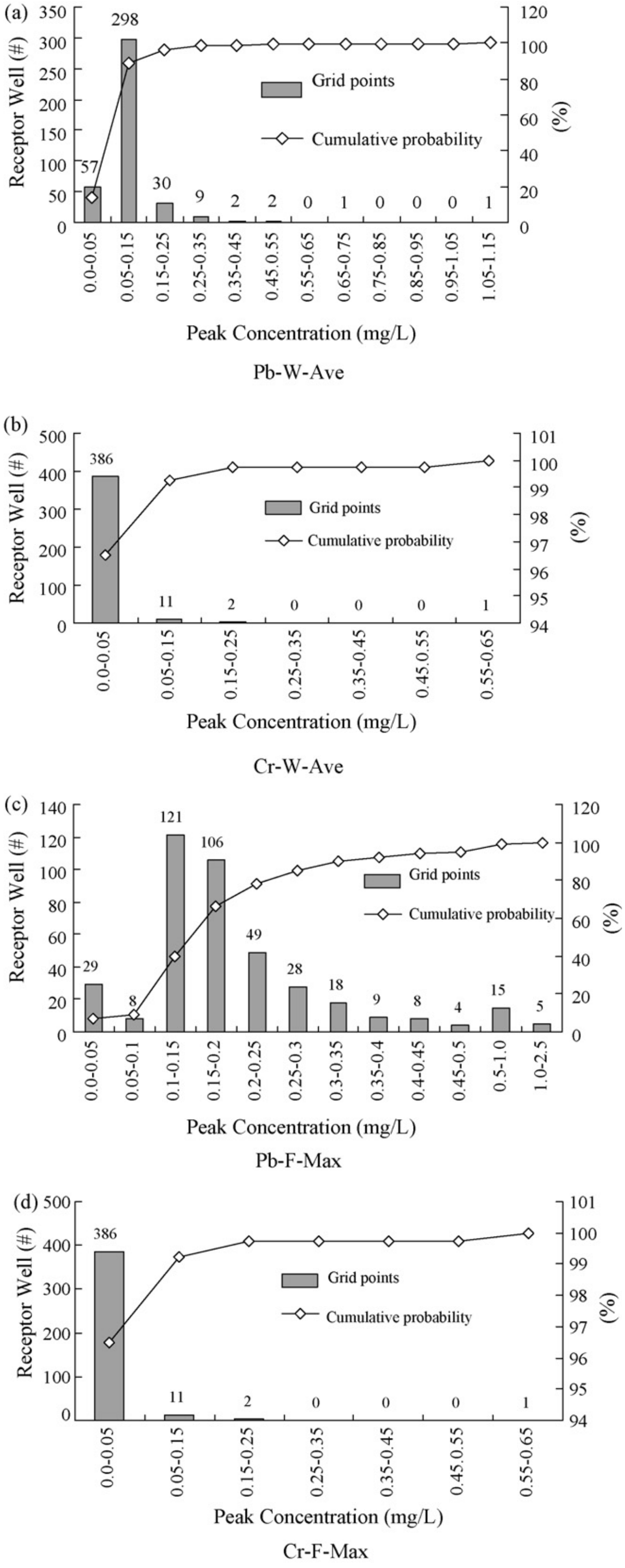

Fig. 3. Distribution of peak concentrations of heavy metals in different landfill scenarios with the averaging and maximum leaching quantity.

is: groundwater drinking, showering, meat consumption and milk ingestion. Except that HQ value (18.9) of W scenario is much higher than the acceptable HQ value 1, simulation results of Cr with the maximum leaching quantity are similar to those with the average leaching quantity.

Fig. 2 presents the risk contours of the 400 grid points for the sanitary landfill with maximum leaching quantity. In terms of Pb cancer risk, all of the risk levels are more than 2.0E−05. The highest value is 3.4E−04, which is three orders of magnitude greater than one calculated with the average leaching quantity. For Pb noncancer risk, the highest HQ value is 510 and the lowest in 30; all are much above the acceptable level. As for Cr noncancer risk, most of the HQ values are below 0.15 and high HQ values are restricted in the area of 3 (*x*-axis) × 3 (*y*-axis), which is 300 m × 300 m.

4.3. Peak concentration

The peak concentration is the highest possible concentration of a metal continuously measured at a receptor well. A comparison of the peak concentrations simulated from average leaching quantities against the groundwater drinking standards (0.05 mg/L for both Pb and Cr) shows that only the scenario W has grid points that cannot meet the groundwater drinking standards (Table 6). Pb concentration in about 85.75% study area exceeds the permissible level, while the exceeding percentage of Cr is 3.5%. Obviously Pb pollution is very serious.

On the contrary, for the results simulated from the maximum leaching quantities, only those of scenario B completely meet the requirements of the groundwater drinking standards (Table 6). The rates of complying with Pb standard for all the other four scenarios are lower than 10%. However, the rates of Cr for all the others are above 96% except the scenario W. The low compliance rates of Pb indicate the importance of adopting the best quality liner and structure for a monoliths landfill.

Fig. 3 illustrates the distribution of possible peak concentrations with the average and maximum leaching quantity. About 50% (298 grid points) of the Pb peak concentrations are between 0.05 and 0.15 mg/L which is higher than the drinking water standard.

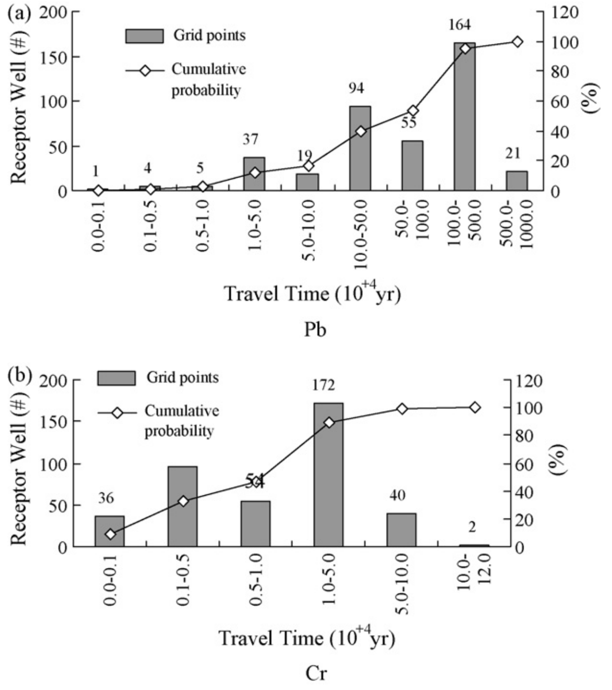

Fig. 4. Travel time of Pb and Cr.

Table 7
The distribution of risks for two landfill scenarios based on average leaching quantity.

Leaching quantity	General sanitary landfill			Sealed landfill		
	Pb		Cr	Pb		Cr
	Cancer risk	HQ	HQ	Cancer risk	HQ	HQ
Original	4.23E−07	0.63	1.6E−03	1.56E−07	0.23	5.54E−04
Avg. of 1000 trials	6.41E−07	0.96	2.17E−03	2.36E−07	0.35	7.51E−04
95th percentile	9.56E−07	1.43	3.23E−03	3.52E−07	0.52	1.12E−03

As shown in Fig. 3, most of the peak concentrations of Pb are in the range of 0.1–0.15 mg/L which is also higher than the standard. However, with very few exceptions, the majority of Cr peak concentrations are lower than 0.05 mg/L.

In this study, travel times of the two heavy metals are also examined. Travel time is the approximate transport time that elapses from when a pollutant leaches from the bottom of SL until it accumulates to the peak concentration in a receptor well. Estimated travel time can help to understand various transport rates and appearance time of peak concentrations of different pollutants. Fig. 4 illustrates the distribution of travel times of Pb and Cr. When 10,000-years is used as the comparison level, only 16.5% (186) grid points of Pb will reach their peak concentrations; while Cr is 46.5%. The transport rate of Cr is three times faster than that of Pb. Most of the grid points reach their peak concentrations at 1,000,000–5,000,000 years interval for Pb and 10,000–50,000 years interval for Cr.

Because the original quantity of Pb in SL is about 15.7 times higher than the quantity of Cr (Pb: 78.36 tons per year; Cr: 4.99 tons per year), Pb's compliance rate is always lower than Cr's in all the scenarios. In addition to the original quantity, the leaching capacity also contributes significantly to the big difference in compliance rates between the two heavy metals. Although Cr's transport rate is higher than Pb's, that does not have much effect on groundwater pollution.

4.4. Uncertainty analysis

In order to understand the effects of parameter uncertainties on the distribution of outcome values, Monte Carlo analysis is performed on 23 parameters by choosing grid point (100, 100) of scenarios F and C as the simulation area. The simulation involves 1000 evaluations to identify 95 percent confidence interval is. Table 7 presents the probability of the distribution of risk for different scenarios based on the averaging leaching quantity. In all the cases, the 95th percentile risk is about 2 times greater than the original risk. Compared with the mean risk of 1000 evaluations, the original risk always falls in the first half low risk area. Using 95th percentile of the noncancer risk value as the criterion, the risk of Cr is within the acceptable range; but for Pb, the noncancer risk of the sanitary landfill (1.43) is obviously greater than that of the sealed landfill (0.52). It is important to consider the use of at least the sealed landfill for fly ash monoliths containing Pb to effectively reduce health risks, although the decision is affected by the degree of conservativeness of decision makers.

5. Summary and conclusion

The potential hazard caused by heavy metal leachate of the monolithic landfills has been always a concerned issue of environmental pollution. This research focused on Pb and Cr leached from 5 kinds of monolithic landfills and characterized the health risks of groundwater pollution in the vicinity. It yields the following conclusions:

1. Comparing the average risk of all grid points, SL site with the worst liner system has the highest risk level, about 5 orders of magnitude higher than the risk level of the best liner quality. The decreasing order of risk of the 5 landfills is the landfill with the worst quality liner, the overcapacity landfill, the common sanitary landfill, the sealed landfill, and the landfill which used the best quality liner. It indicates that multi-layer prevention measure and under design capacity usage can reduce health risks of landfills.
2. In comparison with the pathways of meat consumption, milk ingestion and showering, direct ingestion of Pb and Cr contaminated groundwater is responsible for the highest health risk. Food chain exposure pathway and dermal contact contribute only a small portion to the overall risks.
3. Because the original Pb quantity for landfill treatment is much higher than the Cr quantity, Pb's rates of compliance with the drinking water standard is always lower than Cr's in all the scenarios. Since it takes a long time for Pb to reach the peak concentration in the aquifer, the health effects of Pb on human is slow but greater.
4. Since the original Cr quantity for landfill treatment is low, its pollution area is confined within 500 m around the landfill, even though the Cr's transport rate is higher than Pb's.
5. Without consideration of the parameters uncertainty, the cancer and noncancer risks of Pb with the sanitary landfill method were 4.23E−07 and 0.63, respectively, both within acceptable levels. However, by considering the parameters uncertainty, the noncarcinogenic risk of Pb became 1.43, exceeding the acceptable level. Only under sealed landfill method was the HQ below 1. This may be due to high variability in the Pb concentration in the fly ash monolith.
6. Without consideration of the parameters uncertainty, the noncancer risks of Pb with the general sanitary landfill and the sealed landfill methods were 0.23 and 0.63, respectively, both under acceptable levels. However, by considering the parameters uncertainty, only the HQ of the sealed landfill scenario is below 1. It shows that uncertainty of parameters may change the simulation results significantly.

This paper describes an assessment methodology to foresee the health risks of different fly ash monoliths landfills in order to inform the management of fly ash. Pb was found to be of concern deserving further investigation. However, because Pb and Cr are not the only hazardous substances in the fly ash monoliths, future studies should develop other substances (such as cadmium, mercury and arsenic, etc.) leaching estimation methods to fully assess the risks of landfills. In addition, linear additivity of health effects across multiple exposure pathways and chemicals has been widely used in risk assessment; the resultant uncertainties and the quality and uncertainties of parametric data must be examined carefully so that the simulation results can be used properly to inform the relevant management decisions.

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