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Can washing-pretreatment eliminate the health risk of municipal solid waste incineration fly ash reuse?

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ABSTRACT

Although the reuse of washing-pretreated MSWI fly ash bas been a hot topic, the associated risk is still an issue of great concern. The present study investigated the influence of washing-pretreatment on the total contents and bioaccessibility of heavy metals in MSWI fly ash. Furthermore, the study incorporated bioaccessibility adjustment into probabilistic risk assessment, to quantify the health risk from multi-pathway exposure to the concerned chemicals as a result of reusing washed MSWI fly ash. The results revealed that both water-washing and acid-washing process have resulted in the concentrated heavy metal content, and have reduced the bioaccessibility of heavy metals. Besides, the acid-washing process increased the cancer risk in most cases, while the effect of water-washing process was uncertain. However, both water-washing and acid-washing pretreatment could decrease the hazard index based on bioaccesilbility. Despite the uncertainties accompanying these procedures, the results indicated that, in this application scenario, only water-washing or acid-washing process cannot reduce the actual risk from all samples to acceptable level, especially for cancer risk.

1. Introduction

The reuse of industrial waste in building materials and civil engineering applications has undergone considerable development over a long period. Practices now commonly seen in America, Denmark, Sweden, and the Netherlands are to use coal combustion products, blast-furnace slag and municipal solid waste incineration bottom ash to repair roads and produce asphalt concrete, and ceramic materials (del Valle–Zermeño et al., 2013; Izquierdo et al., 2008; Little et al., 2008). Similarly, the published reports (Guo et al., 2014) have shown that municipal solid waste incineration (MSWI) fly ash has cementitious properties and its main chemical components belong to the system of $CaO–SiO_2–SO_3–Al_2O_3$. Therefore, pretreated MSWI fly ash is now increasingly used for cement manufacturing, roadbed material, and glass ceramics (Aubert et al., 2006, 2007; Francois and Pierson, 2009; Luna Galiano et al., 2011; Wu et al., 2012).

Different from coal fly ash and blast-furnace slag, MSWI fly ash has been listed in the National Hazardous Waste Inventory as HW18 for containing different types of heavy metals, chlorinated organic compounds, dioxins, sulfur compounds, etc., While MSWI fly ash is reused as the civil materials, the issue of potential environmental impact associated with the reuse has emerged. Though pretreatment technologies such as water-washing, acid-washing, have been verified that can remove a part of soluble heavy metals (Anastasiadou et al., 2012; Colangelo et al., 2012), it is still unknown that the potential health risk of pretreated MSWI fly ash reuse is acceptable or not. At present, studies mainly concentrate on using the toxicity characteristic leaching procedure (TCLP) to specify the toxicity and risk of pretreated MSWI fly ash reuse (Lee and Li, 2010; Liu et al., 2009; Yang et al., 2009), but paid little attention to the potential health risk caused by occupational exposure via inhalation, non-dietary ingestion and dermal contact pathways. Besides, approximately 18% of the particle size distributions of fly ashes are under the size of 10 μm, even a fraction of fly ash's particle diameter is around 1 μm (Shi and Kan, 2009), which means the MSWI fly ash particle is easier to adhere to skin, and generate more wind-blown dust emission than soil in the reuse process. Therefore, this paper investigated the health risk of occupational exposure to MSWI fly ash reuse with multi-exposure pathways, in order to get the overall results.

The traditional health risk method based on the total content of heavy metal will lead to over-estimation of risk, since the actual health risk of heavy metals in ingested medium depend on the fraction that is soluble in the gastrointestinal tract available for absorption (Bade et al., 2012; Kördel et al., 2013). To achieve a

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sound evaluation of the health risk, the bioaccessibility or bioavailability of the heavy metals in MSWI fly ash should thus be considered. According to the literatures (Ruby et al., 1999), bioavailability is defined as the fraction of an administered dose that reaches the central (blood) compartment from the gastrointestinal tract, which should be measured by in-vivo studies; while bioaccessibility of a substance is the fraction that is soluble in the gastrointestinal environment and is available for absorption, which can be assessed by in-vitro methods. Considering the cost and time, most researchers preferred to do in-vitro methods, such as the physiologically based extraction test (PBET) (Juhasz et al., 2010, 2011), to evaluate the bioaccessibility of pollutions from matrix such as soils and dust, when assess the actual health risk of heavy metals by ingestion (Hu et al., 2013; Luo et al., 2012; Vasiluk et al., 2011). However, a wealth of studies demonstrated a strong linear relationship between bioaccessibility and exchangeable and soluble fraction, reducible fraction extracted by BCR method (Ahumada et al., 2011; Akkajit and Tongcumpou, 2010; Alvarenga et al., 2009; Baig et al., 2009; Dabek–Zlotorzynska et al., 2005; De La Calle et al., 2013; Karadaş and Kara, 2012; Poggio et al., 2009; USEPA, 2003, 2007). Hence, we used modified four-step BCR method to evaluate the oral bioaccessible fraction (bioaccessibility) in MSWI fly ash.

Additionally, probabilistic approaches, such as Monte Carlo simulation and sensitivity analysis, should be taken into consideration during human health risk assessment process, which could provide the risk assessor with a flexible tool to estimate the uncertainties and stochastic properties of exposure and toxicity (Wu et al., 2011). Nowadays, probabilistic risk assessment has been successfully applied to assess the potential adverse health effects of contaminants from onsite MWS disposal and coal combustion wastes (CCW) practices (Lonati et al., 2007; Lonati and Zanoni, 2012; USEPA, 2010).

Following the discussion above, the purpose of this study was two-fold: (i) to evaluate the effects of water/acid washing pretreatment on the total content and bioaccessibility of heavy metals in MSWI fly ash; (ii) to quantitatively assess the risk of occupational exposure to China MSWI fly ash reuse with a probabilistic approach.

2. Materials and methods

2.1. Sampling and pretreatment

Four kinds of MSWI fly ash samples (FA1, FA2, FA3, and FA4) were collected from four MSWI plants located which all exceeds 300 t MSW/d in four typical regions all over China. Besides, Both FA1 and FA2 were obtained from the grate-type incinerators, while FA3 and FA4 were obtained from the fluidized bed incinerators. The samples were stored in a desiccator at room temperature after the oven-drying process at 105 °C for 24 h.

2.1.1. Water washing pre-treatment

The MSWI fly ash samples were first suspended in distilled water at a liquid–solid ratio (i.e., $cm^3\ g^{-1}$) of 8 in a beaker, and stirred in an agitation apparatus at a rotation speed of 200 ± 2 rpm for 5 h. After washing process, the solid/water mixtures were separated through a vacuum pump filter, and the filter cake was again washed. The resulting material was then oven dried at 105 °C for 24 h, and then stored in a desiccator until analysis.

2.1.2. Acid washing pre-treatment

The MSWI fly ash samples were brought into contact with 0.5 mol/L HNO_3 solutions at a liquid–solid ratio (i.e., $cm^3\ g^{-1}$) of 20 in a beaker, and stirred in an agitation apparatus at a rotation speed of 200 ± 2 rpm for 1 h. After washing process, the solid/water mixtures were separated through a vacuum pump filter, and the solids were dried in an electro-thermostatic blast oven at 105 °C over 24 h. The dried acid-washed MSWI fly ash was collected and stored in desiccators until analysis.

2.2. Chemical analysis

2.2.1. Total contents of heavy metals in raw/pretreated fly ASH

Total content of heavy metal was determined by treating 0.2 g sample with $HNO_3/HClO_4/HF$ acid mixture digestion method at about 120 °C until the digested solution was clear (Sun et al., 2001). Reagent blanks and analytical duplicates were included to ensure the accuracy and precision of analysis.

2.2.2. Chemical speciation of heavy metals in raw/pretreated fly ASH

A modified four-step procedure sequential extraction method (Pan et al., 2013) was adopted to fractionate heavy metals in the exchangeable and acid soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3), and residual fraction (F4). As described above, the bioaccessible fraction of heavy metals have demonstrated a strong linear relationship with the exchangeable and acid soluble fraction (F1) and reducible fraction (F2). Besides, it has been proved that the excess of heavy metal leached in TCLP, was also contributed to the high content of exchangeable and acid soluble fraction (F1) and reducible fraction (F2) of heavy metal (USEPA, 2003). Consequently, we using the F1 and F2 as the oral bioaccessible fraction (bioaccessibility) in MSWI fly ash (Eq. (S1), Table 1).

2.3. Health risk assessment procedures

2.3.1. Evaluation scenario

The major metals, including Zn, Pb, Cu, Cr, Cd, and Ni, are the primary concern of reuse of MSWI fly ash. The risk of worker exposure to pretreated MSWI fly ash open storage pile in landfill site was the focus in this study. The application area was set 15 m × 25 m of the storage pile, and the multiple exposure pathways including non-dietary ingestion, dermal contact and inhalation routes. However, the risk caused by groundwater ingestion was not considered in this study, since the leachate from the storage pile was collected for off-site disposal.

2.3.2. Human exposure and health risk assessment model

Risk assessment is a multi-step procedure comprised data collection and evaluation, exposure assessment, toxicity assessment, and risk characterization. The human exposure to heavy metals in MSWI fly ash can occur via three main pathways: (i) direct oral ingestion of substrate particles ($CDI_{ingestion}$); (ii) inhalation of re-suspended particulates emitted from storage pile ($CDI_{inhalation}$); and (iii) dermal absorption of heavy metals in particles adhered to exposed skin (CDI_{dermal}). The cancer risks were evaluated only for Cr, Cd, and Ni through inhalation exposure pathway, since the metals above are only classified as a known human carcinogen (Group A) or probable human carcinogen (Group B1) via the inhalation route.

According to *Guide for Incorporating Bioavailability Adjustments into Human Health and Ecological Risk Assessments at US Department of Defense Facilities* (USEPA, 2003), modified-mathematical models and exposure parameters were listed in Table S1 and Table S2, respectively (USEPA, 2004; Wang et al., 2013; Wu et al., 2011). We treated BW, EF, ET, AF_d, SA, and IR_{fa} in Eqs. (S2)–(S4) probabilistically, and used Eq. (S5) to incorporate bioaccessibility adjustments into human health risk assessment. In addition, toxicological characteristics in the present study, for each exposure