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The weathering of municipal solid waste incineration bottom ash evaluated by some weathering indices for natural rock

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ABSTRACT

The weathering of municipal solid waste incineration (MSWI) residues consists of complicated phenomena. This makes it difficult to describe leaching behaviors of major and trace elements in fresh/weathered MSWI bottom ash, which was relevant interactively to pH neutralization and formation of secondary minerals. In this study, mineralogical weathering indices for natural rock profiles were applied to fresh/landfilled MSWI bottom ash to investigate the relation of these weathering indices to landfill time and leaching concentrations of component elements. Tested mineralogical weathering indices were Weathering Potential Index (WPI), Ruxton ratio (*R*), Weathering Index of Parker (WIP), Vogt's Residual Index (*V*), Chemical Index of Alternation (CIA), Chemical Index of Weathering (CIW), Plagioclase Index of Alternation (PIA), Silica–Titania Index (STI), Weathering Index of Miura (Wm), and Weatherability index of Hodder (Ks). Welch's *t*-test accepted at 0.2% of significance level that all weathering indices could distinguish fresh and landfilled MSWI bottom ash. However, *R* and STI showed contrasted results for landfilled bottom ash to theoretical expectation. WPI, WIP, Wm, and Ks had good linearity with reclamation time of landfilled MSWI bottom ash. Therefore, these four indices might be applicable as an indicator to indentify fresh/weathered MSWI bottom ash and to estimate weathering time. Although WPI had weak correlation with leachate pH, other weathering indices had no significant correlation. In addition, all weathering indices could not explain leaching concentration of Al, Ca, Cu, and Zn quantitatively. Large difficulty to modify weathering indices correctly suggests that geochemical simulation including surface sorption, complexation with DOM, and other mechanisms seems to be the only way to describe leaching behaviors of major and trace elements in fresh/weathered MSWI bottom ash.

1. Introduction

In Japan, 79.1% of municipal solid waste (MSW) was combusted and 3.60 million metric tons of municipal solid waste incineration (MSWI) residues were landfilled in 2009 (Ministry of the Environment, Japan, 2011). The leachate from landfill sites must be treated to meet environmental standards. For example, in Japan, environmental standards of leachate from MSW landfill sites are pH (5.8–8.6), BOD (<60 mg/L), concentration of potentially toxic metals for natural (soil and groundwater) environments (e.g. Cd: <0.1 mg/l, Cu: <3 mg/L, Zn: <2 mg/L) (Ministry of Health and Welfare, Japan, 1977). In general, leachate monitoring and treatment is usually necessary for long time even after landfill operation is completed. However, it is quite difficult to estimate necessary period for complete closure of a landfill site because many complex mechanisms are relevant for evaluating the leaching behavior of pollutants. Although some kinds of leaching tests under controlled/uncontrolled pH conditions and geochemical simulation are helpful to investigate leaching mechanisms of metals and other pollutants in MSWI bottom ash (Dijkstra et al., 2002, 2006a,b; Dijkstra et al., 2008; Li et al., 2007; Meima and Comans, 1997, 1998, 1999; Polettini and Pomi, 2004; Van der Sloot et al., 1996), it should be repeatedly stated that necessary period of landfill closure estimated based on leaching tests and geochemical simulation has still large uncertainty owing to complex weathering processes including physical, chemical, and mineralogical reactions.

Although the weathering of MSWI bottom ash preceded many complex reactions, this study focused on leaching of some major and trace elements. Some expected mechanisms in weathering processes, which are pH neutralization, formation of secondary minerals, and sorption to secondary minerals, would affect leaching behaviors of major and trace elements. For example, the neutralization from alkaline pH via carbonation and other reactions decreases

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leaching concentrations of some metals (Johnson et al., 1995; Meima et al., 2002). Some of element leaching behaviors can be explained by the change of solubility-control minerals (Dijkstra et al., 2006a, 2008; Johnson et al., 1995, 1996; Meima and Comans, 1997, 1998, 1999; Meima et al., 2002). Geochemical reactions like hydration and carbonation promoted the formation of secondary minerals like calcite (CaCO3), ettringite ($3CaOAl_2O_3CaSO_4{\cdot}32H_2O$), gypsum ($CaSO_4{\cdot}2H_2O$), anhydrite ($CaSO_4$), and gibbsite ($Al(OH)_3$) (Chimenos et al., 2003; Freyssinet et al., 2002; Piantonea et al., 2004; Polettini and Pomi, 2004; Speiser et al., 2000). Other indentified secondary minerals were reported to be connellite ($Cu_{19}Cl_4(SO_4)(OH)_{32}{\cdot}3H_2O$), halotrichite ($FeAl_2(SO_4)_4{\cdot}22H_2O$), quenstedtite ($Fe(SO_4)_3{\cdot}10H_2O$), melanterite ($FeSO_4{\cdot}7H_2O$), rostite ($Al(SO_4)(OH,F){\cdot}5H_2O$), hematite ($Fe_2O_3$), goethite (FeOOH), weddellite ($CaC_2O_4{\cdot}2H_2O$), and so on (Freyssinet et al., 2002; Piantonea et al., 2004). This seems to enhance the sorption of metals to secondary minerals or inhibit the sorption owing to the conversion of adsorptive minerals (Cornelis et al., 2006; Meima and Comans, 1997; Piantonea et al., 2004). Notwithstanding the complexity of the bottom ash weathering and related leaching processes, previous studies showed that modeling calculations based on sorption constants for hydrous ferric oxide (HFO) were able to describe metals leaching (e.g. Pb, Mo) from weathered MSWI bottom ash (Meima and Comans, 1998). In the present study, a different approach was tried to evaluate element leachability of weathered MSWI bottom ash using simple indices. Considering successful simulations of element leaching behaviors focusing on the sorption to primary/secondary minerals, this study focused on mineralogical weathering indices for natural rock profiles, which were defined as some specific weight/molar ratios of component elements. They were applied to fresh/landfilled MSWI bottom ash. If the net effect of pH, chemical precipitation, and sorption to primary/secondary minerals on element leaching behaviors has certain correlation with specific molar ratios of major components, mineralogical weathering indices might be applicable to describe leaching behaviors of component elements including metals. During the weathering of MSWI bottom ash, mobile/semi-mobile elements are leached and immobile elements remained in MSWI bottom ash. Therefore, specific molar ratios of component elements might reach asymptotically to certain ranges with weathering period. This implies the possibility to describe "weathering time" of landfilled MSWI bottom ash using weathering indices. In addition, this might enable to expect necessary time for chemical/mineralogical stabilization of landfilled MSWI bottom ash through the weathering. This would be helpful to estimate necessary period of landfill closure using only a simple index. In this context, as a first step, the objective of this study is to investigate the relation of these weathering indices to landfill time and leaching concentrations of component elements of fresh/landfilled MSWI bottom ash.

2. Weathering index

2.1. General explanation of weathering indices

During the weathering process, some of the physical and the chemical characteristics of the incinerator bottom ash undergo significant changes including the fragmentation of the big particles into smaller ones and the oxidation of metals in addition to hydrolysis/hydration, dissolution/precipitation of hydroxides and salts of the main cations (Chimenos et al., 2003; Piantonea et al., 2004; Polettini and Pomi, 2004). Although physical and chemical types of weathering should occur in landfilled MSWI residues in parallel, chemical weathering seems to be promoted under wet condition prior to physical weathering if no significant mechanical impact was given to landfilled MSWI bottom ash. In this study, the authors focused on chemical weathering, in particular leaching of major components, as mentioned in Section 1.

In the case of natural rock weathering, the principal assumption of chemical weathering indices is that weathering intensity controlled leaching behavior of major components in natural rocks (Ceryan et al., 2008). Even though the intensity of weathering increases, certain major oxides like Al_2O_3, Fe_2O_3 and TiO_2 are considered as 'immobile'. Therefore, they are assumed to remain constant. On the other hand, SiO_2, Na_2O, K_2O, CaO, and MgO are considered as mobile/semi-mobile. There are different mobilities among these elements. Na, K, Mg, and Ca have higher mobilities than that of Si. The authors expected that specific molar ratios of major components (weathering indices) reached asymptotically to certain ranges owing to leaching of mobile elements. If so, they likely have correlation with "weathering time" of landfilled MSWI bottom ash and are helpful to estimate necessary time for chemical/mineralogical stabilization owing to the weathering. In addition, correlation between weathering indices and net effects of some geochemical mechanisms on element mobilization might be also expectable. This likely enables to describe leaching behaviors of major components and metals using a simple index. Explanations of weathering indices as mentioned below were referred from Price and Velbel (2003), in which more detailed description of each weathering index was reported. In this study, all weathering indices were calculated using weight percent of each element.

2.2. Weathering Potential Index (WPI)

Reiche (1943) suggested weathering index to evaluate potential weatherability of rocks (Weathering index of potential: WPI). It is expressed by percentage ratio of alkalis and alkaline elements to the total earth elements (see Eq. (1)). The denominator of Eq. (1) has mobile, semi-mobile, and immobile element terms (e.g. Na, Si, Al, etc.). In contrast, the numerator has only mobile/semi-mobile element terms. Because values of mobile/semi-mobile element terms decrease owing to the leaching during the weathering, WPI will decrease with the increase of weathering degree. Since Fe(II) oxide (FeO) and Fe(III) oxide (Fe_2O_3) were not analyzed separately in this study, all Fe was assumed to be Fe(III) oxide because hematite (Fe_2O_3) was detected in MSWI bottom ash as Fe oxide crystalline phase (Speiser et al., 2000; Wei et al., 2011). In addition, this study used modified WPI, in which H_2O term was subtracted as suggested by Ceryan et al. (2008).

$$\begin{aligned}\mathrm{WPI} = 100 \cdot (K_2O + Na_2O + CaO + MgO - H_2O^{+})/(SiO_2 \\ + Al_2O_3 + Fe_2O_3 + TiO_2 + FeO + CaO + MgO + K_2O \\ + Na_2O) \end{aligned} \tag{1}$$

2.3. Ruxton ratio (R)

Ruxton (1968) proposed a simple weathering index (*R*), which is expressed by the ratio of silicate oxide to aluminum oxide (see Eq. (2)). *R* decreases with the increase of weathering degree. Because Al is considered as immobile, this assumption made ones expect that *R* would have a correlation with the ratio of silicate loss to the total loss of leached elements. Ruxton (1968) reported that this index is the best applicable to evaluate weathering profiles developed on uniform acid to intermediate bedrock with constant sesquioxide content during the weathering.

$$R = SiO_2/Al_2O_3 \tag{2}$$

2.4. Weathering Index of Parker (WIP)

Parker (1970) suggested weathering index (WIP) for silicate rocks based on element mobility, which was related to bond strength of each element (Na, K, Mg, and Ca) with oxygen. It is ex-