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Heavy metal source analysis in municipal solid waste (MSW): Case study on Cu and Zn

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ABSTRACT

This research contributes to the knowledge of the heavy metal sources in municipal solid waste (MSW). Samples were collected from 8 cities of Zhejiang province, Eastern China. Cu and Zn, the most two conventional heavy metals with extensive distribution in many kinds of MSW components, were investigated. It shows components of kitchen waste (KW), ash (AS), plastic (PL), and paper (PA) have high universality in MSW and accounted for 55.1–95.5% in each MSW sample. Moreover, these four components are also the main heavy metal sources of MSW, which accounted for 76.3% and 82.3% contribution of the Cu and Zn contents, respectively. The Cu and Zn contents in the gross MSW sample were 41.2–1643.7 mg kg^{-1} and 109.3–1077.9 mg kg^{-1}, respectively, which on different degree exceed the set standard for "environmental quality standard for soil" (Cu, 100 mg kg^{-1}; Zn, 400 mg kg^{-1}) of China and have high potential of environmental risk. The heavy metal contents in the gross MSW do not have spatial variation but present high seasonal variation, significantly higher in summer than winter ($P < 0.01$). Much more attention should be paid on the MSW management in summer to avoid heavy metal pollution.

1. Introduction

As the final MSW disposal method, landfill is a widely accepted technology, especially in developing countries. In China, more than 90% of the MSW is disposed by landfill. However, treatment of MSW by landfill has been, and still is, connected with a risk of pollution [1–3]. The large number of cases of groundwater pollution at landfills and the substantial resources spent in remediation suggests that landfill leachate is a significant source of pollutants [4]. When considering different kinds of contaminants in landfill leachate, heavy metals are especially dangerous because of their persistence and toxicity [5]. Heavy metals can be transferred to the ecosystem components such as underground water or crops, and can thus affect human health through the water supply and food web [6].

Coupled with the improvement of life quality, many kinds of living goods have been entered into people's daily lives. Correspondingly, the MSW components became complicated and disordered, which result in the great variety of heavy metal sources. Batteries, paints, dyes and inks in paper, pesticides and fertilizers in yard waste, which are examples of household and commercial wastes, contain high quantities of heavy metals [7]. However, the MSW components mentioned above are not weighted equally. For example, the battery is always reported to have high heavy metal contents, but it always has very low proportion in MSW. While component like paper seems low but the total amount takes high percentage in MSW. Moreover, it must also be noted that the discharge of metals from a landfill is not limited to a short period. For a long period after the dumping of the MSW at a landfill [8–10], and also after the closure of a landfill, the leaching of heavy metals will continue [11–13]. Obviously, MSW components such as food residual and plastic have different environmental risks on heavy metal leaching. As all kinds of MSW components gathering into a landfill site, heavy metals in them converged and became a new artificial sink. Therefore, the knowledge of the heavy metal contents in MSW components and the origin of these levels are prior objectives. It is also crucial for policy making orientated at reducing heavy metal inputs to surrounding environment like soil and guaranteeing the maintenance or even improvement of soil functions [6].

Nowadays, heavy metal pollution from landfill garners increasing attention but their source analysis has heretofore been widely neglected. The lack of knowledge about heavy metal source in MSW not only limits our insights into environmental risk evaluation, but also confines our understanding of policy making on separation, collection and sustainable management. Considerable effort has thus been expended to better understand the heavy metal source in MSW. Multivariate analysis has been widely used in soil to assist the interpretation of environmental data

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Nomenclature

MSW	municipal solid waste
HZ	Hangzhou
NB	Ningbo
JH	Jinhua
JX	Jiaxing
WZ	Wenzhou
TZ	Taizhou
SX	Shaoxing
QZ	Quzhou
RU	rubber
LE	leaf
KW	kitchen waste
TE	textile
PL	plastic
PA	paper
ME	metal
AS	ash
VE	vegetable
BO	bone
PE	pericarp
GL	glass
TI	timber
CE	cigarette end
HA	hair
VFA	volatile fatty acid

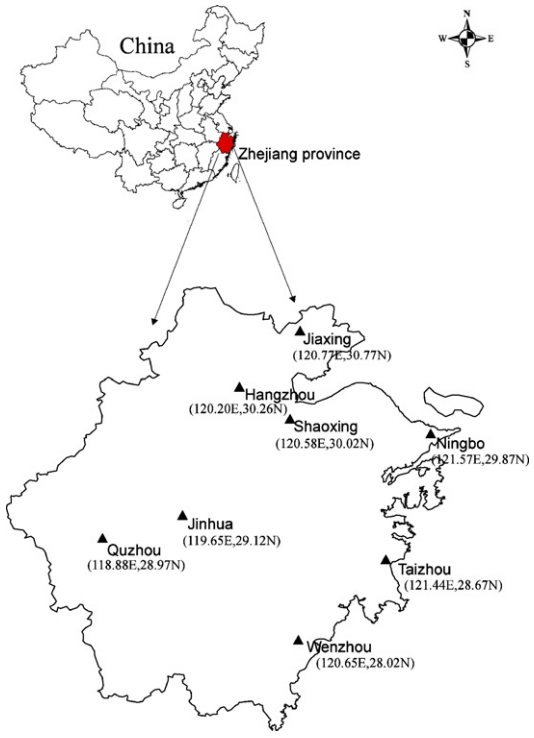

Fig. 1. Map of sampling location.

[14–16]. Many studies also have examined relationships among elements (major and trace) and between elemental concentrations and other soil properties (clay content, cation exchange capacity, pH, soil texture, carbonates) in non-contaminated soils [17–20]. However, MSW is an extremely heterogeneous material independent of its geometry, particle size or chemical composition [21]. Moreover, the generation of MSW also has an obviously seasonal and regional nature. A designated sampling may mask the real information.

This research focused on heavy metal source analysis in MSW. Cu and Zn, the most two conventional heavy metals with extensive distribution in many kinds of MSW components, were investigated. MSW samples were temporally and spatially collected from Zhejiang province in Eastern China. It aims to provide reference information on MSW management and heavy metal pollution control in landfill sites.

2. Materials and methods

2.1. Site characteristics

Zhejiang province is located in the southern part of the Yangtze River Delta on the southeast coast of China (Fig. 1). It lies between 27°12′ to 31°31′ north latitude and 118°00′ to 123°00′ east longitude. It faces the East China Sea on the east and shares borders with Shanghai on the north. The province covers a total land area of 101,800 km^2. Hills and mountains account for 70.4% of the total area in the province. Plains and basins make up 23.2% while the rest 6.4% is water area composed of rivers and lakes. There are 11 cities under the direct jurisdiction of Zhejiang provincial government. The provincial capital city is Hangzhou.

2.2. MSW sampling

Eight cities in Zhejiang province were selected for regional sampling, namely HZ, NB, JH, JX, WZ, TZ, SX, and QZ. Among them, HZ, SX, JH, JX and QZ are inland cities, while TZ, NB and WZ are coastal cities.

In each city, MSW samples were randomly collected from 3 MSW transfer stations. Moreover, in order to achieve a representative sample, MSW were continuously collected at different time in a day, namely, every 4 h from 4:00 AM to 8:00 PM at each transfer station. Subsequently, all sub-samples from each transfer station were completely mixed manually and immediately placed in air-tight plastic bags. 30.0 kg of MSW was finally collected from each city. There were six parallel samples collected in each city.

In addition to regional sampling, we also chose HZ for seasonal variation analysis of MSW. The sampling locations were the same as regional sampling. The MSW sampling process like the procedure mentioned above. MSW samples were sampled monthly and the whole process lasted for a whole year in 2008. There were also six parallel samples collected in each month.

2.3. Sample preparation

Firstly, each MSW sample was carefully mixed and homogenized. Then, around 5.0 kg of each sample was taken for the analysis of the pseudototal contents of Cu and Zn and the moisture content. The rest samples were manually separated as carefully as possible, and the visible and separable components in each MSW sample are all classified and picked out. The specific proportions of each MSW component were characterized by their wet weights. Subsequently, the moisture content of each component was also determined while the rest of MSW (including the gross MSW and each MSW component) were ground by crasher (BB51, Retsch, Germany) after air-drying and sheived with a 40 mesh sheiver.

After air-drying, all MSW samples (including the gross MSW and each MSW component) were ground by crasher (BB51, Retsch, Germany), respectively. The achievable final out-put fineness of the crasher was set as 40 mesh. Then, the ground fraction of each sample was then filled into a plastic flask and homogenized thoroughly by shaking and shaking manually. Sequentially, the sample was

moved out from the flask and placed on a clean watertight board with a taper shape. After quartering, 1 g of the homogenized subsample was taken for determination of the pseudototal contents of Cu and Zn in each component.

2.4. Analysis

The moisture contents of the gross MSW sample and each MSW component were analyzed by drying at 105 °C until a constant weight was achieved. The pseudototal contents of Cu and Zn in the gross MSW sample and each MSW component were determined in accordance with standard methods [22]. 1 g of air-dried representative sample was weighed into the poly tetra fluoro ethylene (PTFE) vessel, 0.5–1.0 ml of deionized water was added to obtain a slurry, then 7.0 ml of 12.0 mol l^{-1} HCl followed by 2.3 ml of 15.8 mol l^{-1} HNO_3 was added drop by drop to reduce foaming. The PTFE vessel was allowed to stand for 16 h (overnight) at room temperature for slow oxidation of the organic matter of the MSW. The temperature of the reaction mixture was slowly raised, until reflux conditions were reached and maintained for 2 h. After cooling the PTFE vessel to room temperature, the digests were filtered into 100 ml in volumetric flasks and diluted to the mark with deionized water. Each sample was handled in triplicate. Cu and Zn were determined by atomic absorption spectrophotometer (SHIMADZU AA-650, Japan).

3. Results and discussion

3.1. Characteristics of the Cu and Zn contents in MSW components

In order to evaluate the heavy metal contribution of each MSW components, MSW samples were firstly separated as carefully as possible. It indicated that the highest possible MSW components included 15 categories, namely RU, LE, KW, TE, PL, PA, ME, AS, VE, BO, PE, GL, TI, CE, and HA. The detailed characteristics of each MSW components are presented in Table 1. Among the collected MSW samples, bulk items like cardboard, paper skin, plastic bottle, woven bag and bulk metalwork were not found indicating that MSW experienced some recovery processes before it was finally discarded.

Table 1
Characteristics of MSW components.

Components	Characteristic
RU	Rubber bag, bicycle tyre, rubber overshoes, rubber band, etc.
LE	Broad leaf, flowers and plants, etc.
KW	Leftovers, hogwash, etc.
TE	Cloth, mitten, fabric, etc.
PL	Various plastic bags except plastic bottle and woven bag
PA	Various paper products like newspaper, toilet paper, food package, cigarette case, etc. Cardboard and paper skin excluded
ME	Pop can, bottle cap, wire, screw, etc. Bulk metalwork excluded
AS	Ash, dust and fine inseparable residue
VE	Vegetable root and leaf
BO	Bone from pork and fish, shell, eggshell, etc.
	Various pericarps
GL	Crushed beer bottle, glass, little oral liquid bottle, etc.
TI	Disposable chopsticks, bamboo stick, broken branch from garden, etc.
CE	Cigarette end including sponge and tobacco
HA	Various animal and human hair

However, this does not mean that the MSW management was well-done because the MSW components were still complicated. For example, the RU component included discard rubber bag, bicycle tyre, and rubber overshoes which can be recycled and should not be directly discarded. Moreover, newspaper in the PA component is also another important resource which should be recycled but not discarded.

Because not all components were found in each MSW sample, comparisons were aided by computing relative fraction specific percentages based on the 15 categories. The separation results of MSW from different regions of Zhejiang province are shown in Fig. 2. It indicates a significant difference on MSW components. For example, there were no VE, BO, PE, GL, TI, GE, CE, and HA found in the sample from JH, while those can be separated from other samples in different degree. However, some components have high

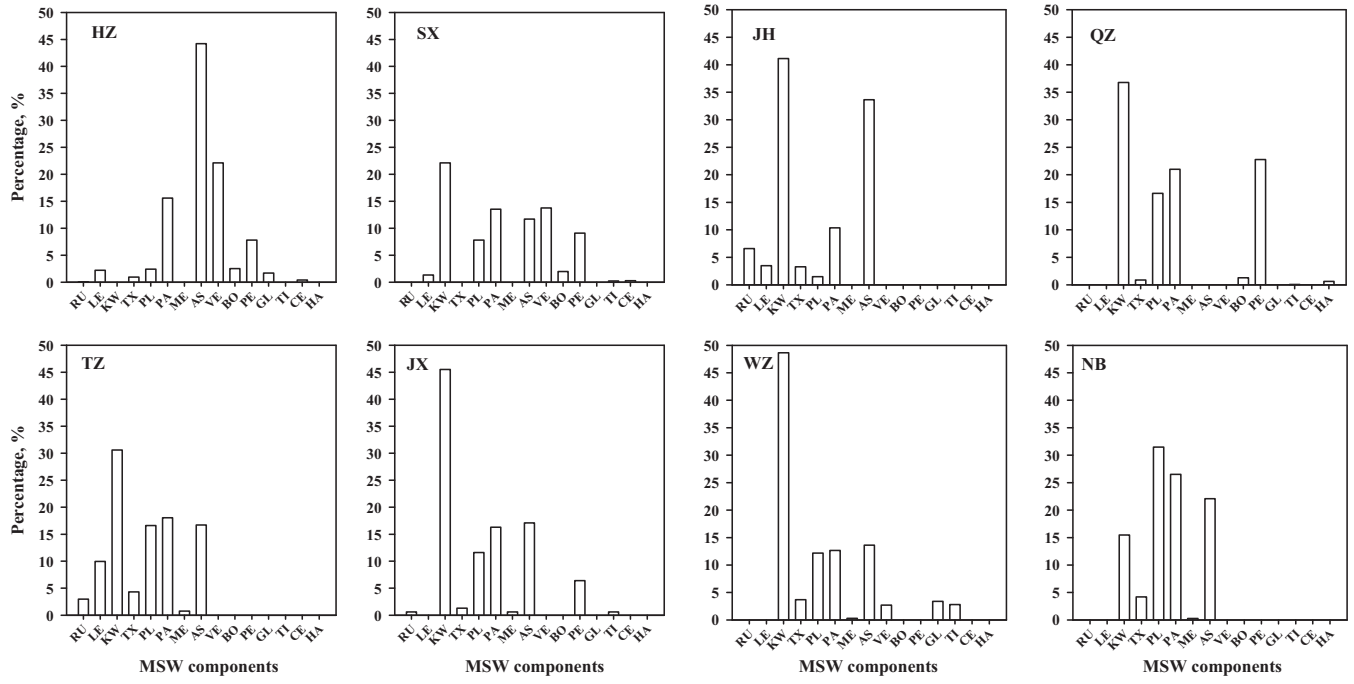

Fig. 2. Percentage of MSW components from different cities of Zhejiang province.

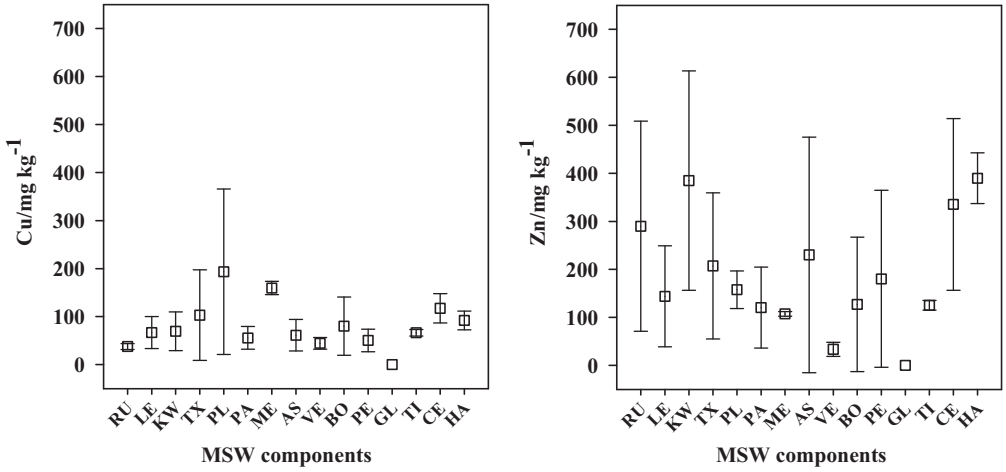

Fig. 3. Cu and Zn contents in different MSW components.

universality in MSW, such as PL, PA, AS, and KW, and accounted for 55.1–95.5% in each MSW sample, while components like CE and HA had very low proportion and were not found frequently. The possible reasons of the MSW component difference might be ascribed to the differences on economic level and life style among the inland cities and coastal cities.

Based on the careful separation, the Cu and Zn contents in each components (GL was excluded) were analyzed. As Fig. 3 shown, the highest average Cu content, 194.5 mg kg^{-1}, was observed in PL, and the lowest one, 37.6 mg kg^{-1}, was found in RU. Moreover, in the MSW components with high percentage including PA, KW and AS, the Cu contents were of 55.6 mg kg^{-1}, 69.4 mg kg^{-1}, and 61.3 mg kg^{-1}, respectively. As for Zn, the highest and the lowest level were observed in HA and LE which accounted for 389.7 mg kg^{-1} and 33.5 mg kg^{-1}, respectively. While the components with relatively high Zn included PA, KW and AS and with Zn content of 120.3 mg kg^{-1}, 385.0 mg kg^{-1}, and 230.2 mg kg^{-1}, respectively.

The total amount of heavy metal in MSW is depend both on the content of heavy metal in certain component and its proportions in MSW. However, these two factors are not logically associated, for instance, the CE and HA have high contents of Cu and Zn, but their proportions in MSW are always very low. While components like PA, KW, and AS were of high proportion but with low Cu and Zn contents. In order to evaluate the contribution of each MSW components on the Cu and Zn contents, their weighting coefficients were calculated. In the calculation, an ideal MSW sample including all the 15 kinds of components were assumed. Some components have their average proportion and contents of heavy metal. Therefore, the weighting coefficient of each MSW component can be calculated by Eq. (1)

$$P_i = \frac{C_i \times wt_i\%}{\sum C_i \times wt_i\%} \quad (1)$$

where P_i means the weighting coefficient of each MSW component, C_i means the average Cu and/or Zn content in each MSW component (Fig. 3), and wt_i% means the average proportion of each MSW component (Fig. 2). The results are shown in Table 2. It indicated that the components of PL, KW, AS, and PA accounted for the most Cu contribution, namely 25.9%, 25.5%, 14.9%, and 10.0%, respectively. Similarly, there are also the same in Zn contribution. Dissimilarly, the contribution order is KW, AS, PL, and PA with the weighting coefficients of 48.2%, 19.1%, 7.6%, and 7.4%, respectively. These four components accounted for 76.3% and 82.3% contribution of Cu and Zn contents, respectively. This was mainly because these four components accounted for the overwhelming majority in MSW sample as mentioned above. Therefore, the heavy metal pollution control should be ascribed to the reasonable management of those components, i.e. PL, KW, AS, and PA. Moreover, the strategies like plastic limit order, paper recycle as well as multipurpose use of kitchen waste and ash can significantly alleviate the potential of heavy metal pollution from MSW.

3.2. Spatial variation of Cu and Zn contents in MSW

In addition to each MSW component's contribution on the heavy metal contents of the gross MSW sample, the spatial variation of Cu and Zn contents in the gross MSW were investigated. Fig. 4 shows the pseudototal contents of the Cu and Zn in the gross MSW from eight cities in Zhejiang province. It indicated that the Cu and Zn contents in MSW are in great spatial variation, namely 41.2–1643.7 mg kg^{-1} and 109.3–1077.9 mg kg^{-1}, respectively. Moreover, the contents of Cu and Zn on different degree exceed the set standard for "environmental quality standard for soil" (Cu, 100 mg kg^{-1}; Zn, 400 mg kg^{-1}) of China at different degree. It thus indicated that heavy metals in MSW have high potential of environmental risk, whose misgoverning may affect the agricultural industry and human health.

In order to evaluate the regional effect, the average contents of Cu and Zn in MSW were statistically analyzed using Tukey method with the software of SAS (SAS 13.1). The results indicated a common feature that the Cu contents in most tested MSW samples fell in between 100 mg kg^{-1} and 200 mg kg^{-1}. The samples from TZ and QZ had the lowest Cu contents of 46.2 mg kg^{-1} and 41.2 mg kg^{-1},

Table 2
Contribution of MSW components on heavy metal content in the gross MSW.

Component	P_i (Cu)	Component	P_i (Zn)
PL	25.9	KW	48.2
KW	25.5	AS	19.1
AS	14.9	PL	7.6
PA	10.0	PA	7.4
PE	6.2	PE	7.2
VE	6.1	RU	2.7
LE	3.0	LE	2.2
TE	2.6	TE	1.8
BO	1.7	VE	1.6
CE	1.5	BO	0.9
RU	1.0	HA	0.5
ME	0.7	TI	0.4
TI	0.7	CE	0.4
HA	0.3	ME	0.2
GL	0	GL	0

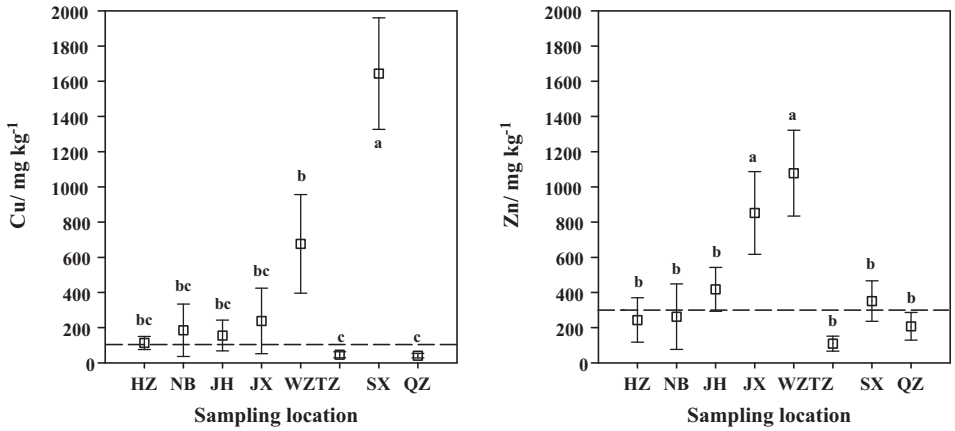

Fig. 4. Cu and Zn contents in MSW from different cities of Zhejiang province (dashed means the environmental standard of soil, and the lowercase stands for the metal content was significant difference at the 0.05 probability level).

respectively, while the sample from SX showed the highest Cu content which accounted for 1643.7 mg kg^{-1}. In terms of statistics, the MSW sample from SX showed significantly different with the other samples on the Cu content ($P < 0.01$). However, to our surprise, there were no significantly difference among JX, NB, JH, HZ, TZ, and QZ though the samples from TZ and QZ had the lowest average contents ($P > 0.05$). The same phenomena can also be found on Zn. Compare to that of Cu, the Zn content in MSW from different regions had much larger variation ranging from 109.3 mg kg^{-1} to 1077.9 mg kg^{-1}, however, there were also no significantly difference among them except WZ and JX ($P > 0.05$). The variation of the Cu and Zn contents in MSW are mainly ascribe to the property of MSW because it is an extremely heterogeneous material independent of its geometry, particle size or chemical composition [21]. Based on the same sampling process, we cannot distinguish the difference among the heavy metal contents in MSW from different regions. It implies that the heavy metal contents in MSW may not show an obvious spatial variation. However, it can be confirmed that their disorder management always threatens to the surround environment because of exceeding the set standard for "environmental quality standard for soil" mentioned above. The reasonable management of MSW is an important strategy of avoiding heavy metal pollution.

3.3. Seasonal variation of Cu and Zn in MSW

In this test, HZ, the capital city of Zhejiang, was chosen for continuously sampling. As Fig. 5 shown, during the period, almost half a years' MSW sample have heavy metal pollution risk because of the Cu and Zn contents exceeding the set standard of "environmental quality standard for soil" mentioned above. It suggests that much attention should be paid on heavy metal in MSW. In general, the Cu and Zn contents were high in the summer, especially in July and August. The highest Cu and Zn contents were 205.5 mg kg^{-1} and 571.6 mg kg^{-1}, respectively. On the contrary, the Cu and Zn contents had the lowest level in the winter, December–February. The lowest Cu and Zn contents were 51.5 mg kg^{-1} and 200.4 mg kg^{-1}, respectively. It indicates that the heavy metal contents in MSW have obviously seasonal nature. The heavy metal content in MSW is significantly higher in summer than winter ($P < 0.01$).

One of the possible reasons is the changing of dietary structure. In summer, a large number of cold drink like coke and ice-cream were consumed, coupled with the discard of various kinds of plastic package and wrapping paper. Moreover, all kinds of fruit and food for relieving summer-heat such as watermelon produce a lot of the kitchen waste and pericarp. As discussed above, those MSW components have high contents of Cu and Zn, and then caused high heavy metal levels. Moreover, microbial degradation is another important factor leading to the seasonal variation. Obviously, the microbiological activity is much higher in summer than in winter. Once the MSW were randomly dumped but not promptly disposed, the putrescible MSW components like KW and PE can easily ferment and generate VFA. VFA, acetic acid for example, can dissociate promptly as shown in Eq. (2). Subsequently, Cu(II) and Zn(II) were released from solid-Me (definition of the original speciation of Cu

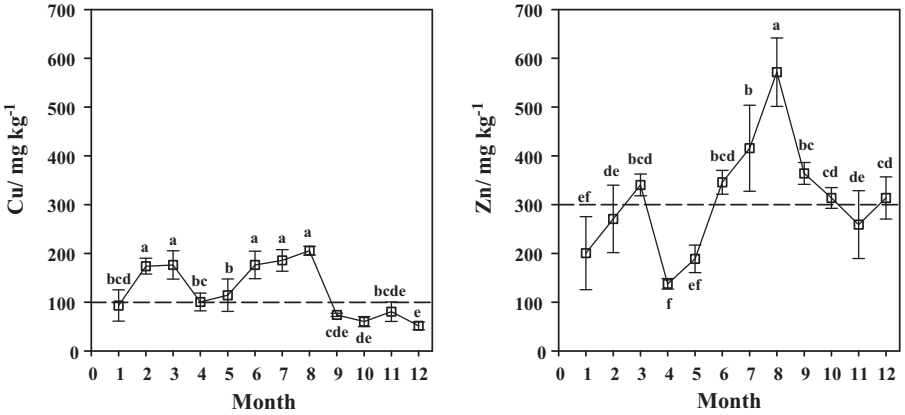

Fig. 5. Seasonal variation of Cu and Zn contents in MSW (dashed means the environmental standard of soil, and the lowercase stands for the metal content was significant difference at the 0.05 probability level).

and Zn in MSW components) in refuse with the action of H^+ from the dissociation of VFA (Eq. (3)).

$$HAc \leftrightarrow H^+ + AC^- \quad (2)$$

$$Solid\text{-}Me + 2H^+ \rightarrow Me^{2+} \quad (3)$$

Obviously, Me^{2+} is more dangerous than its original speciation (solid-Me). By contrast, those phenomena can be greatly avoided in the winter. From the point of view of policy making, much more attention should be paid on the MSW management in summer.

4. Conclusion

The MSW components are complicated and significantly different. Components of PL, PA, AS, and KW have high universality in MSW and accounted for 55.1–95.5% in each MSW sample. These four components are also the main heavy metal sources of MSW, which accounted for 76.3% and 82.3% contribution of the Cu and Zn contents, respectively. The contents of Cu and Zn in the gross MSW sample were 41.2–1643.7 mg kg^{-1} and 109.3–1077.9 mg kg^{-1}, respectively, which on different degree exceeded the set standard for "environmental quality standard for soil" (Cu, 100 mg kg^{-1}; Zn, 400 mg kg^{-1}) of China and have high potential of environmental risk. The heavy metal contents in the gross MSW do not have significant spatial variation but has high seasonal variation, significantly higher in summer than winter ($P < 0.01$). Much more attention should be paid on the MSW management in summer to avoid heavy metal pollution.

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