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RESEARCH ARTICLE

Two-step calculation method to enable the ecological and human health risk assessment of remediated soil treated through thermal curing

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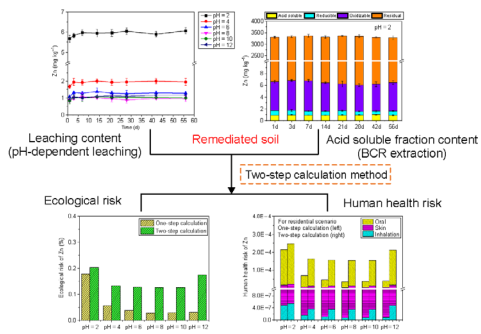
HIGHLIGHTS

• Remediated soil treated by thermal curing exhibited strong inherent resistance to acidic attack with the formation of $ZnCr_2O_4$ spinel.
• A two-step method to calculate the sum of the leaching and acid-soluble fraction contents of Zn in remediated soil for risk evaluation have been proposed.
• Compared with the traditional one-step calculation method, this two-step calculation method can effectively avoid underestimating the risk of remediated soils.

GRAPHICAL ABSTRACT



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ABSTRACT

The centralized utilization of heavy-metal-contaminated soil has become the main strategy to remediate brownfield-site pollution. However, few studies have evaluated the ecological and human health risks of reusing these remediated soils. Considering Zn as the target metal, systematic pH-dependent leaching and the Community Bureau of Reference (BCR) extraction were conducted at six pH values (pH = 2, 4, 6, 8, 10, 12) for the remediated soil treated through thermal curing. The pH-dependent leaching results showed that with the formation of $ZnCr_2O_4$ spinel phases, the remediated soil exhibited strong inherent resistance to acidic attack over longer leaching periods. Furthermore, the BCR extraction results showed that the leaching agent pH value mainly affected the acid-soluble fraction content. Moreover, a strong complementary relationship was noted between the leaching and acid-soluble fraction contents, indicating that the sum of these two parameters is representative of the remediated soil risk value. Therefore, we proposed a two-step calculation method to determine the sum of the two heavy metal parameters as the risk value of remediated soil. In contrast to the traditional one-step calculation method, which only uses the leaching content as the risk value, this two-step calculation method can effectively avoid underestimating the risk of remediated soil.

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1 Introduction

With the acceleration of urbanization and industrial transformation, many old factories are facing closure or relocation, leaving behind soils with significant heavy metal contamination, and such soils have attracted increasing research attention worldwide (Khalid et al., 2017; Xie et al., 2019). The hazardous metals, such as zinc (Zn), chromium (Cr), lead (Pb), copper (Cu) and cadmium (Cd), present in these site soils can be attributed to the industrial emissions, traffic emissions, and mining activities (Luo et al., 2012). These heavy metals are nonbiodegradable and thus persist in soils, thereby affecting the yield and quality of farm crops and posing a significant risk to human health (Fu et al., 2012; Sun et al., 2019). However, in the context of ensuring sustainable development and alleviating the shortage of soil resource, these brownfield sites must be necessarily subjected to secondary development (Wang et al., 2018; Vareda et al., 2019). Therefore, effective remediation technologies and corresponding risk evaluation methods must be identified to evaluate the potential ecological and human health risks of remediated soils in different actual reuse scenarios.

Until now, many techniques has been developed to remediate heavy-metal-contaminated brownfield-site soils, such as physical, chemical and biological remediation (Kumpiene et al., 2008; Yao et al., 2012). Thermal curing, based on the fraction transformation through high-temperature sintering, is a notable heavy-metal-contaminated soil remediation technology because of its high efficiency and practical advantages (Samaksaman et al., 2016; Li et al., 2019). Through thermal curing, the hazardous metals in contaminated soils can be incorporated into spinel crystal structures and reused as building materials such as bricks in residential and industrial scenarios (Tang et al., 2011; Guo et al., 2017). Spinels are usually expressed using the general formula "AB_2O_4," where "A" represents divalent metals such as Zn, Cu, Cd or Ni, and "B" represents trivalent matrix metals such as Cr or Al (Marinković et al., 2004). Spinels have been recognized as a promising crystal structure in which a variety of divalent metals can be incorporated and exist stably in the obtained sintered products for a long time (Shih et al., 2006; Tang et al., 2016).

After heavy metals are stabilized in a spinel structure, their leaching potential is significantly reduced (Taha et al., 2018; Ding et al., 2019). For example, when ZnO is sintered (to simulate zinc-laden ash) with kaolinite and mullite ceramic precursors, both zinc aluminate spinel ($ZnAl_2O_4$) and willemite (Zn_2SiO_4) phases can be observed in the products. The leachability of the potential product phases indicates that the zinc contents in ZnO and Zn_2SiO_4 leachates are approximately two orders of magnitude higher than those in $ZnAl_2O_4$ leachate (Shih and Tang, 2012). Spinels demonstrate a considerably higher inherent resistance to acidic attack than metal oxides under leaching, and thus, the spinel incorporation strategy has been noted to be beneficial in stabilizing hazardous metals (Tang et al., 2014). However, the leaching content only reflects the cation-proton exchange mechanism in the metal leaching of remediated soil. Certain doubts remain regarding the potential release of heavy metals due to geochemical fraction changes in the complex reuse environment, as well as the subsequently generated ecological and human health risks during the long-term reuse process (Malviya and Chaudhary, 2006; Liu et al., 2018). Stakeholders in the reuse process of remediated soil are mostly concerned about the fate of heavily contaminated soil and the evaluation of its environmental effect because of the potential detrimental effects on the ecological security and human health. Therefore, environmental risk assessment guidelines for remediated soils from brownfield sites are necessary and becoming increasingly stringent.

The risk values associated with remediated soil reuse in actual complex environmental conditions are key to evaluate the corresponding ecological and human health risks. Almost all the existing ecological and human health risk evaluation models are based on the use of risk values to determine the risk level (Yang et al., 2018). In recent decades, several one-step methods have been reported, in which the calculated leaching content (determined through leaching tests) is used to represent the risk value of remediated soils (Ding et al., 2019). These methods mainly include the toxicity characteristic leaching procedure (TCLP), synthetic precipitation leaching procedure (SPLP) and multiple extraction procedure (MEP), which were issued by the US Environmental Protection Agency (USEPA) and are widely used in the current methods to evaluate the effect of soil remediation technologies (Abbas et al., 2018; Mahedi and Cetin, 2019). If the leaching contents of heavy metals are lower than the relevant toxicity standards under specific experimental conditions, the method is considered to be in accordance with the remediation requirements (Gupta et al., 2019). The risk of heavy metals in remediated soils includes both the short-term leaching risk and long-term release risk. However, the traditional one-step calculation methods are usually based on the leaching contents of heavy metals released under strong acid conditions, such as those involving a specific pH of 2–3, and thus represent only the short-term leaching risk of the remediated soil. It is difficult to characterize the long-term release risk of heavy metals under complex environmental pH conditions, and the environmental risk may be underestimated by simply considering the leaching content as the risk value (Taha et al., 2019). In addition, these methods focus only on the effect of the remediation technology and do not extensively consider the ecological and human health risk implications of these remediated soils in the reused environment. Therefore, it is particularly important and urgent to establish a more suitable risk value calculation method to evaluate the ecological and human health risks of remediated soil in different actual reuse scenarios.

In this study, considering Zn as the target metal, systematic pH-dependent leaching and Community Bureau of Reference (BCR) extraction were conducted at six pH values (pH = 2, 4, 6, 8, 10 and 12) for remediated soil treated through thermal curing. The leaching and fraction distribution stability were examined, and a suitable risk value calculation method for the ecological and human health risk evaluation of remediated soil was proposed. Specifically, the objectives of this study were to 1) explore the leaching and fraction distribution stability characteristics of Zn in remediated soil at different pH values; 2) propose a suitable risk value calculation method for environmental risk assessment; and 3) compare the differences in the risk assessment results between the new risk value calculation method and traditional one-step calculation method.

2 Materials and methods

2.1 Thermal curing

Among the hazardous metals found in brownfield-site soils, Zn is one of the most concerning and in need of remediation (Li et al., 2014; Zhou et al., 2017). In a previous study, we observed that Zn could be incorporated into a $ZnCr_2O_4$ spinel structure by sintering Zn-enriched artificial soils (Wu et al., 2019). Thus, considering Zn as the target metal, soil that was critically polluted with Zn was artificially prepared, and coal gangue and shale (at a mass ratio of 1:3) were added as auxiliary materials to sinter the mixed polluted soil into bricks. The polluted soil was made of 10 g of a $ZnO + Cr_2O_3$ mixture (Zn:Cr molar ratio of 1:2) and 90 g of uncontaminated soil powder. All the powder mixtures were airdried and pressed into pellets under a pressure of 350 MPa. Subsequently, the samples were thermally treated at 1300°C for a dwell time of 3 h with a controlled heating and cooling rate of 10°C min^{-1} in a muffle furnace. Moreover, all the original raw materials were digested with HCl-HNO_3-$HClO_4$, and the initial Zn content was determined through the inductively coupled plasma optical emission spectrometry (ICP-OES, Optima 8000, Perkin Elmer). It indicates that clean soil was used to prepare contaminated soil, thereby eliminating the influence of Zn and Cr in the original soil on the later experiment. The results are summarized in Table 1.

2.2 pH-dependent leaching procedure

All the sintered samples were ground to pass through a 0.149 mm sieve, and pH-dependent leaching procedures were performed. A total of six pH values (pH = 2, 4, 6, 8, 10 and 12) were considered. According to the solid waste extraction procedure for leaching toxicity of China (HJ/T299-2007), the leaching solution was prepared from a mixture of concentrated sulfuric acid (H_2SO_4) and concentrated nitric acid (HNO_3) with a mass ratio of 2:1 (leaching solution pH = 2, 4, 6), and the pH of the leaching solution was adjusted using sodium hydroxide (NaOH) and deionized water (leaching solution pH = 8, 10, 12) (Zhou et al., 2018; Zhang et al., 2020). Each leaching vial was filled with 20 mL of the leaching solution and 1 g of the sample powder, and each treatment was repeated three times. The leaching vials were rotated end-over-end at 30 r min^{-1} for agitation periods ranging from 1 to 56 d. For each sample series, a total of eight samplings were extracted at time intervals of 1 d, 3 d, 7 d, 14 d, 21 d, 28 d, 42 d and 56 d. Next, the leachates were filtered through 0.45 μm cellulose membranes, and the leaching content of Zn was determined through ICP-OES.

2.3 BCR extraction

The heavy metal fraction was analyzed using the BCR three-step extraction method proposed by the European Community Standards Division (Sahuquillo et al., 1999; Pardo et al., 2004). Specifically, four geochemical fractions of heavy metals were determined, including the acid-soluble fraction, reducible fraction, oxidizable fraction, and residual fraction (Pueyo et al., 2008; Sutherland, 2010). After all the samples were subjected to pH-dependent leaching, all the residual samples were washed with deionized water to ensure the same pH value conditions, and next, the BCR extraction procedure was conducted. The specific details regarding the BCR extraction method implemented in this study are as follows. After the leaching experiment, each sample was transferred to a 50 mL centrifuge tube, and different reagents were added to enable continuous extraction. To obtain the acid-soluble fraction, 20 mL of 0.11 mol L^{-1} CH_3COOH was added to the sample, which was shaken and extracted for 18 h and later centrifuged at 9000 r min^{-1} for 20 min. The supernatant was stored for testing. To obtain the reducible fraction, 20 mL of 0.5 mol L^{-1} $NH_2OH \cdot HCl$ was added to the abovementioned residue, the sample was shaken for 18 h and centrifuged, and the supernatant was stored for testing. To obtain the oxidizable fraction, 10 mL of 8.8 mol L^{-1} H_2O_2 was slowly added to the abovementioned residue; subsequently, the sample was shaken for 2 h and heated at 85°C in a water bath. After cooling, 40 mL of 1.0 mol L^{-1} CH_3COONH_4 was added, the sample was shaken for 18 h and centrifuged, and the supernatant was stored for testing. To obtain the residual fraction, the remaining solid residue was washed with deionized water and then digested with HCl-HNO_3-$HClO_4$ for testing. All the geochemical fraction contents of Zn were determined through ICP-OES.

2.4 Risk value calculation

The risk value is key to evaluate the ecological and human health risks of remediated soils. Almost all the current ecological and human health risk evaluation models are

Table 1 Zn content in raw experimental materials

Heavy metal (mg kg^{-1})	Uncontaminated soil	Coal gangue	Shale	Mixed soil sample
Zn	27.7±3.8	23.0±1.7	90.6±5.7	3413.7±205.6

based on the risk values that the heavy metals may produce under complex environmental conditions (Yang et al., 2018). For example, the most widely used ecological risk assessment model (RAC) considers the acid-soluble fraction content of heavy metals as the risk value and the proportion of this content relative to the total amount of heavy metals as the potential ecological risk value of heavy metals (Nemati et al., 2011; Li et al., 2018). In the human health risk evaluation model recommended by the technical guidelines for the risk assessment of the soil contamination of land for construction in China (HJ 25.3-2019), the total amount of heavy metals is considered as the risk value, and the sum of the risks generated by the total amount of heavy metals in various exposure pathways related to the human body is considered as the potential human health risk value. In general, the leaching content of remediated soils treated through thermal curing is usually considered as the risk value to calculate the corresponding environmental risks when using the traditional one-step calculation method (Gupta et al., 2019). However, the geochemical fraction distribution of remediated soils is significantly different from that of natural soil. The environmental risk may be underestimated if only the leaching content is considered as the risk value. Therefore, according to the leaching and fraction distribution stability characteristics summarized in this study, the risk value calculation includes two components: the leaching content and the content that may be released due to changes in the fraction distribution of heavy metals. The calculated risk values can be applied in the existing ecological risk and human health risk models to realize risk assessment.

3 Results and discussion

3.1 Leaching and fraction distribution stability characteristics of Zn under different pH value leaching conditions

Based on the theory of AB_2O_4 spinel formation, the molar ratio of Zn (a divalent ion) to Cr (a trivalent ion) was set as 1:2, which is stoichiometrically consistent with the ratio of the two metals in the product phase. In the initial stage of $ZnCr_2O_4$ spinel formation, a solid-state reaction occurred between ZnO and Cr_2O_3 due to a nucleation process, leading to the formation of the $ZnCr_2O_4$ spinel with a cubic face structure (Stephen et al. 2007; Dixit et al. 2015). In our previous work, by combining advanced analytical technologies such as X-ray photoelectron spectroscopy and transmission electron microscopy equipped with energy dispersive X-ray spectroscopy (EDX), $ZnCr_2O_4$ spinel was identified in the sintered mixture of ZnO + Cr_2O_3. Furthermore, 70.55% of the available Zn was included in a $ZnCr_2O_4$ spinel phase at the lowest temperature (700°C), while the transformation ratio value of Zn increased continuously with the temperature until it reached nearly 100% at 1300°C after 3 h. All the incorporation mechanisms of Zn and Cr into $ZnCr_2O_4$ spinel have been presented in our previous study and are thus not repeated in this article (Wu et al., 2019).

Figure 1 demonstrates the leaching performance of Zn under different pH value leaching conditions, as evaluated through the pH-dependent leaching. The experimental results showed that the leaching contents of Zn in sintered samples increased with the extraction time and gradually leveled off after one week. Specifically, after one week, the average leaching contents of Zn were 5.98, 1.95, 1.29, 0.96, 1.02 and 1.10 mg kg^{-1} as the pH increased from 2 to 12. The leaching content of Zn under acidic conditions was significantly higher than that under alkaline conditions, and the difference was not significant under the alkaline condition. In general, strong acid conditions are more conducive to the release of divalent metal cations such as Zn^{2+} , and pH neutral conditions impede the release. The leaching content of Zn was consistently lower than the risk screening value of soil remediation of heavy-metal-contaminated sites in China for residential land (Zn < 500 mg kg^{-1}, DB43/T1165). This result implies that the Zn in remediated soils is well consolidated through thermal curing to realize brownfield-site heavy metal pollution

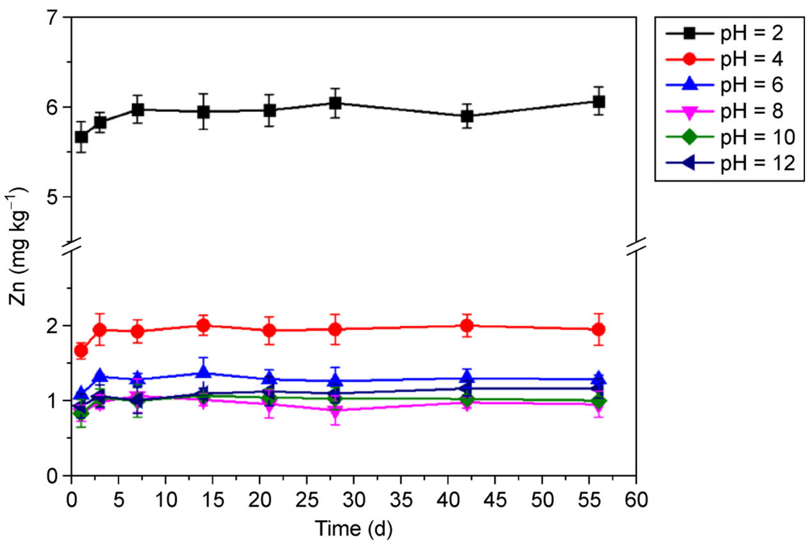

Fig. 1 Leaching characteristics of Zn in remediated soil at different values of the leaching agent pH.

remediation. With the continuous formation of $ZnCr_2O_4$ spinel, the leachability of Zn^{2+} was reduced significantly due to its incorporation into the spinel crystal structure (Tang et al., 2011; Snellings, 2015). Additionally, with a decrease in the leaching agent pH value, the leaching amount of Zn increased significantly, which reflected the short-term leaching characteristics and inherent resistance to acidic attack of the remediated soil. Moreover, strong acid conditions could promote the release of Zn^{2+} ; however, such a release did not change significantly as the leaching time elapsed, which reflected the leaching stability of the remediated soil subjected to thermal curing.

The geochemical fraction distribution characteristics of Zn in the samples after the completion of the pH-dependent leaching procedure are presented in Fig. 2. The BCR sequential extraction procedure results showed that the contents of Zn in the acid-soluble fraction, reducible fraction, oxidizable fraction, and residual fraction were

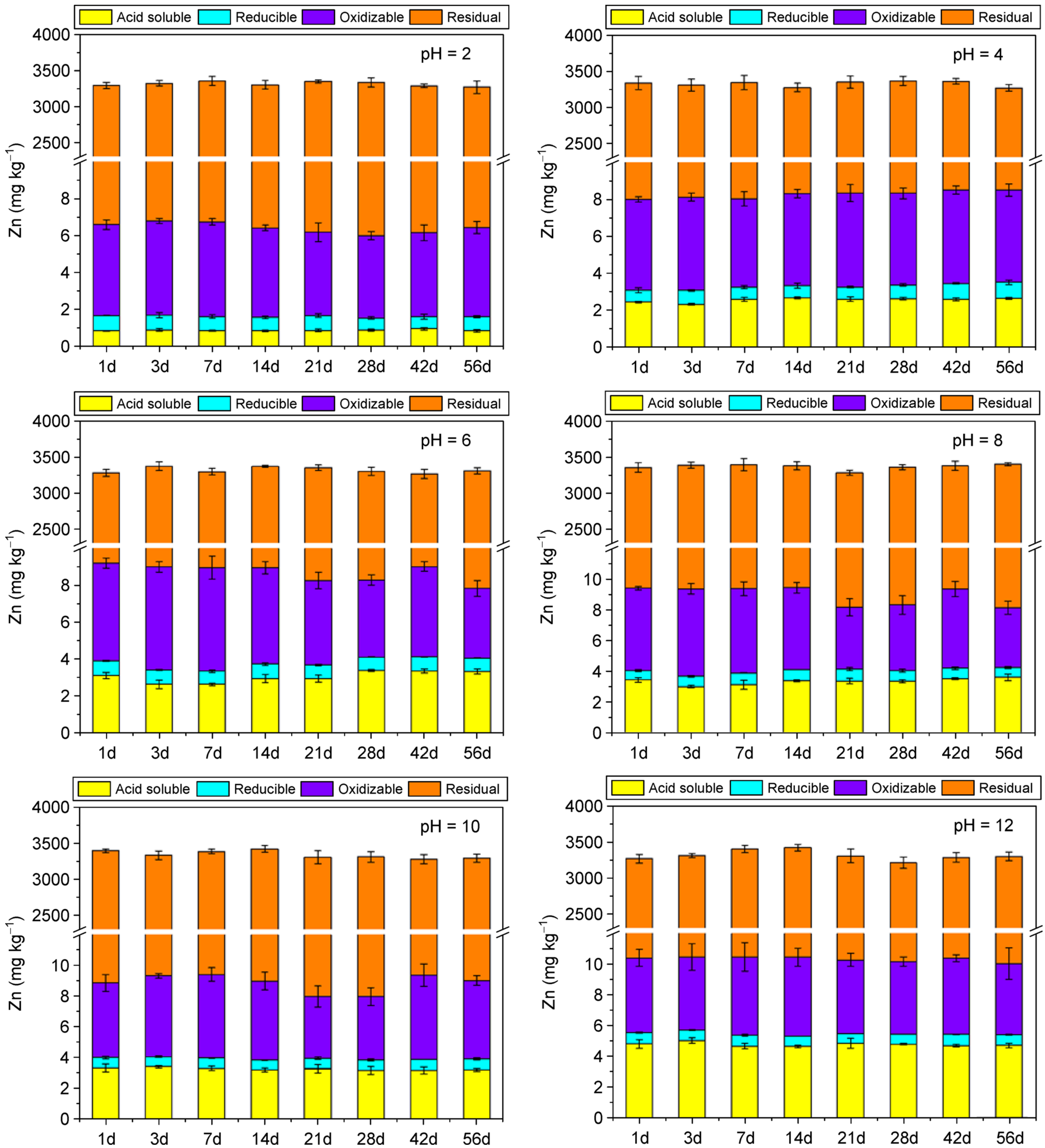

Fig. 2 Fraction distribution characteristics of Zn in remediated soil.

0.85–4.77 mg kg^{-1}, 0.69–0.76 mg kg^{-1}, 4.82–5.00 mg kg^{-1} and 3308.20–3357.42 mg kg^{-1}, respectively, at different leaching agent pH values. The composition of the four Zn fractions in the remediated soil showed that the content of the residual fraction was significantly higher than those of the other three fractions. With the formation of $ZnCr_2O_4$ spinel during the thermal curing sintering process, most of the acid-soluble fraction in the samples was converted into the three other, more stable fractions.

Furthermore, the Pearson correlation analysis showed that the leaching content ($R^2 = -0.750$) and acid-soluble fraction content ($R^2 = 0.915$) of Zn in the samples exhibited a strong relationship with the pH value of the leaching agent (Table 2). Moreover, a strong inverse correlation was observed between the leaching content and acid-soluble fraction content ($R^2 = -0.850$), which reflected a strong complementary relationship between these two parameters. More importantly, the pH value of the leaching agent mainly affected the leaching content and acid-soluble fraction content and did not significantly influence the other three fractions. This effect reflected the long-term release and fraction distribution characteristics of Zn in the remediated soil. Under the action of a strong acid leaching agent, the large amount of Zn^{2+} adsorbed on the surface of the remediated soil leached out rapidly in the first leaching step, which included a certain amount of Zn^{2+} in the form of the acid-soluble fraction. Furthermore, in the second extraction step, the content of the acid-soluble fraction in the remediated soil correspondingly decreased. Conversely, the decrease in the leaching content in the first leaching step led to an increase in the acid-soluble content in the second extraction step. Therefore, these two parameters exhibited a complementary relationship, which also provided a theoretical basis for the environmental risk assessment in remediated soils.

3.2 Two-step calculation method to determine the risk value of remediated soil

The pH value is the most important factor in the natural environment and can affect the release of heavy metals in soil (Komonweeraket et al., 2015). Numerous studies have demonstrated that extremely acidic or alkaline conditions promote the release of heavy metals in soil (Kogbara et al., 2012; Król et al., 2020). In accordance, the experimental results of this study showed that the leaching agent pH value directly affected the leaching content and acid-soluble fraction of remediated soil subjected to thermal curing (Figs. 1 and 2).

The biological toxicity of heavy metals is related not only to their leaching amount but also to the geochemical fraction distribution. This distribution directly affects the migration and circulation of heavy metals in the environment (Palleiro et al., 2016). Consequently, analyzing the heavy metal geochemical fraction distribution is valuable and can help distinguish the bioavailability and effect of heavy metals in remediated soil (Arunachalam et al., 1996; Saleem et al., 2018). Among the four geochemical fractions, the acid-soluble fraction metal, which is adsorbed on the soil component, poses the highest risk to the environment but is most often ignored. Therefore, it is desirable to account for this parameter in the present remediation soil risk assessment system. The other three fractions are relatively stable and unlikely to be released from samples even under extreme conditions (Pérez-Moreno et al., 2018). Therefore, it is necessary to propose a more suitable method than the traditional one-step method for risk value calculation by incorporating the acid-soluble fraction distribution characteristics of remediated soil in the calculation system.

Notably, as the pH increases from 2 to 12, the sum of the leaching content and acid-soluble fraction content becomes 6.78, 4.46, 4.29, 4.26, 4.25 and 5.84 mg kg^{-1} (Fig. 3). The sum of the leaching content and acid-soluble fraction content is significantly higher than the leaching content. This phenomenon is even more pronounced under alkaline conditions. Especially under strong alkali conditions (pH = 10 and pH = 12), the sum of the leaching content and acid-soluble fraction content is 4.2 and 5.3 times as high as the leaching content, respectively. In such a scenario, if we use the traditional one-step calculation method, which only calculates the leaching content of heavy metals to evaluate the risk of remediated soils, the risk may be significantly underestimated. In addition, under the condition of a strong alkali environment, the heavy metals in remediated soils do not leach over a short period of time, although they are still stored in the soils in the form of the acid-soluble fraction. These heavy metals may be released in the future, considering a long time scale; therefore, it is necessary to consider the potential release risk of such

Table 2 Correlation relationship of the leaching agent pH values, leaching contents and four fractions of Zn.

Correlation relationship	Leaching agent pH value	Leaching content	Acid soluble fraction	Reducible fraction	Oxidizable fraction	Residual fraction
Leaching agent pH value	1	–0.750**	0.915**	–0.378**	0.020	0.088
Leaching content	–0.750**	1	–0.850**	0.231	–0.079	–0.143
Acid soluble fraction	0.915**	–0.850**	1	–0.309*	–0.031	0.023
Reducible fraction	–0.378**	0.231	–0.309*	1	0.120	–0.206
Oxidizable fraction	0.020	–0.079	–0.031	0.120	1	0.223
Residual fraction	0.088	–0.143	0.023	–0.206	0.223	1

** Significantly correlated at the 0.01 level (both sides). * Significantly correlated at the 0.05 level (both sides).

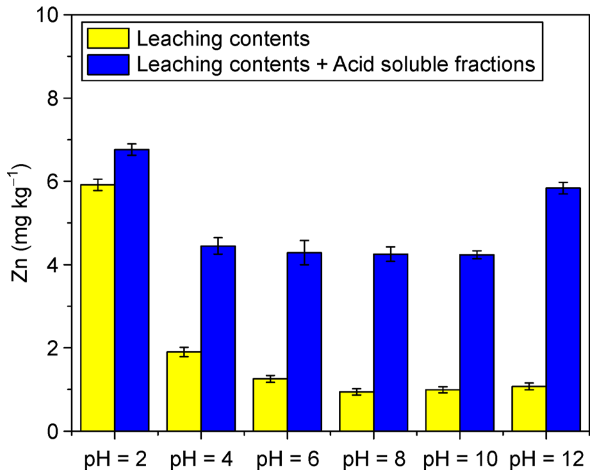

Fig. 3 Leaching and acid soluble fraction content characteristics of remediated soil at different values of the leaching agent pH.

metals. Therefore, a two-step calculation method to calculate the sum of the leaching content and acid-soluble fraction content of heavy metals may be a better strategy to reasonably assess the potential risk value in a complex actual environment.

The risk value can be calculated as follows:

$$C_{\text{risk}} = C_{\text{leaching}} + C_{\text{acid}} \tag{1}$$

where C_{risk}, C_{leaching} and C_{acid} represent the risk value, leaching content and acid-soluble fraction content of the remediated soil samples, respectively. Thus, the proposed approach can effectively avoid the underestimation of the risk, as in the traditional one-step calculation method.

3.3 Evaluation of ecological risk for remediated soil

By incorporating the variation characteristics of the leaching content and acid-soluble fraction content, the risk value (C_{risk}) was obtained through pH-dependent leaching and the BCR extraction procedure. Subsequently, the risk assessment code model (RAC), which has been widely applied with the BCR sequential extraction scheme to assess the ecological risks of heavy metals in soil, was modified and adopted to evaluate the ecological risks of remediated soil subjected to thermal curing (Nemati et al., 2011; Li et al., 2018). In this study, the modified ecological risk calculation model (MRAC) based on the RAC was used to calculate the level of risk. The MRAC can be calculated as follows:

$$\mathrm{R}_{\text{ecological}} = C_{\text{risk}}/(C_{\text{risk}} + C_{\text{reducible}} + C_{\text{oxidizable}} + C_{\text{residual}}) \tag{2}$$

In the formula, $\mathrm{R}_{\text{ecological}}$ denotes the ecological risk of the sample; and C_{risk}, $C_{\text{reducible}}$, $C_{\text{oxidizable}}$ and C_{residual} represent the risk value and reducible fraction, oxidizable fraction and residual fraction contents, respectively. The five classifications in MRAC include a safe level (less than 1%), low-risk level (1%–10%), medium-risk level (10%–30%), high-risk level (30%–50%) and very high-risk level (over 50%) (Jain, 2004; Tong et al., 2020).

Based on the one-step and two-step calculation methods, the ecological risks of the remediated soil were evaluated using the MRAC method. The results showed that in the case of the one-step calculation method, the ecological risks of Zn in the remediated soil samples were 0.18%, 0.06%, 0.04%, 0.03%, 0.03% and 0.03% at pH = 2 to pH = 12. However, when the two-step calculation method was used, the ecological risks of Zn were 0.20%, 0.13%, 0.13%, 0.13%, 0.13% and 0.18% at pH = 2 to pH = 12 (Fig. 4). The ecological risks of Zn in the remediated soil calculated using the two calculation methods corresponded to a safe level. However, the ecological risk results calculated using the two-step calculation method were significantly higher than those calculated using the one-step calculation method. This discrepancy was especially true under strong alkali conditions (pH = 10 and pH = 12), in which the ecological risk value increased by 4.3 times and 6 times, respectively. Compared with the one-step calculation method, the results of the two-step calculation method were more conservative and conducive to facilitate the environmental protection of remediated soils at reuse sites.

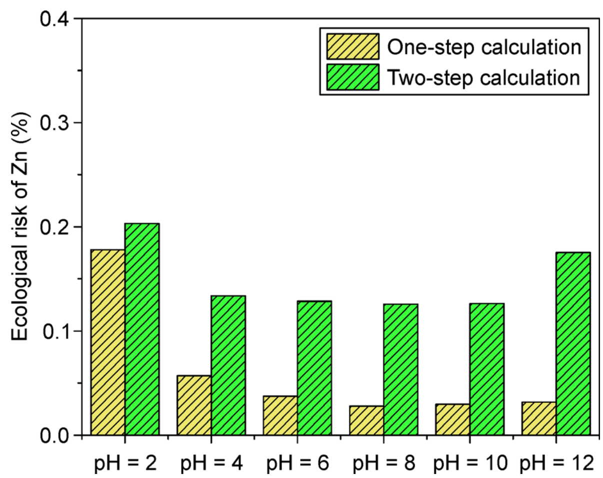

Fig. 4 Ecological risk of Zn in remediated soil.

3.4 Evaluation of the human health risk of remediated soil

The risk value (C_{risk}) obtained through the systematic pH-dependent leaching and BCR extraction procedure can be combined with a human health risk evaluation model to calculate the human health risk of remediated soil. In this study, the human health risk evaluation model proposed by the technical guidelines for the risk assessment of soil contamination of land for construction in China (HJ 25.3-2019) was used. This model is usually divided into two parts: cancer and noncancer submodels (Hu et al., 2017; Wang et al., 2020). Since heavy metals are nongaseous pollutants, the human health risks of heavy metals in the remediated soil are primarily attributed to oral ingestion, skin contact, and direct inhalation. We considered two types of reuse issues represented by residential and industrial scenarios.

For the residential scenario, the carcinogenic effect of heavy metals and lifetime hazards of exposure to children and adults were considered. For the noncarcinogenic effect of heavy metals, the harm to the children upon exposure was considered. The calculation formulas related to the oral intake, skin contact and direct inhalation are as follows:

Risk from oral intake of remediated soil:

$$\mathrm{OISER_{ca}} = C_{\mathrm{risk}} \times S_{\mathrm{Fo}} \times \left(\frac{OSIR_{\mathrm{c}} \times ED_{\mathrm{c}} \times EF_{\mathrm{c}}}{BW_{\mathrm{c}} \times AT_{\mathrm{ca}}} + \frac{OSIR_{\mathrm{a}} \times ED_{\mathrm{a}} \times EF_{\mathrm{a}}}{BW_{\mathrm{a}} \times AT_{\mathrm{ca}}}\right) \times \mathrm{ABS_o} \times 10^{-6} \quad (3)$$

$$\mathrm{OISER_{nc}} = \frac{C_{\mathrm{risk}}}{RfD_{\mathrm{o}} \times SAF} \times \frac{OSIR_{\mathrm{c}} \times ED_{\mathrm{c}} \times EF_{\mathrm{c}}}{BW_{\mathrm{c}} \times AT_{\mathrm{nc}}} \times \mathrm{ABS_o} \times 10^{-6} \quad (4)$$

Risk from skin contact of remediated soil:

$$\mathrm{DCSER_{ca}} = C_{\mathrm{risk}} \times S_{\mathrm{Fd}} \times \left(\frac{SAE_{\mathrm{c}} \times SSAR_{\mathrm{c}} \times EF_{\mathrm{c}} \times ED_{\mathrm{c}} \times E_{\mathrm{v}} \times ABS_{\mathrm{d}}}{BW_{\mathrm{c}} \times AT_{\mathrm{ca}}} \times 10^{-6} + \frac{SAE_{\mathrm{a}} \times SSAR_{\mathrm{a}} \times EF_{\mathrm{a}} \times ED_{\mathrm{a}} \times E_{\mathrm{v}} \times ABS_{\mathrm{d}}}{BW_{\mathrm{a}} \times AT_{\mathrm{ca}}} \times 10^{-6}\right) \quad (5)$$

$$\mathrm{DCSER_{nc}} = \frac{C_{\mathrm{risk}}}{RfD_{\mathrm{d}} \times SAF} \times \frac{SAE_{\mathrm{c}} \times SSAR_{\mathrm{c}} \times EF_{\mathrm{c}} \times ED_{\mathrm{c}} \times E_{\mathrm{v}} \times ABS_{\mathrm{d}}}{BW_{\mathrm{c}} \times AT_{\mathrm{nc}}} \times 10^{-6} \quad (6)$$

Risk from direct inhalation of remediated soil particles:

$$\mathrm{PISER_{ca}} = C_{\mathrm{risk}} \times S_{\mathrm{Fi}} \times \left(\frac{PM_{10} \times DAIR_{\mathrm{c}} \times PIAF \times ED_{\mathrm{c}} \times (fspo \times EFO_{\mathrm{c}} + fspi \times EFI_{\mathrm{c}})}{BW_{\mathrm{c}} \times AT_{\mathrm{ca}}} \times 10^{-6} + \frac{PM_{10} \times DAIR_{\mathrm{a}} \times PIAF \times ED_{\mathrm{a}} \times (fspo \times EFO_{\mathrm{a}} + fspi \times EFI_{\mathrm{a}})}{BW_{\mathrm{a}} \times AT_{\mathrm{ca}}} \times 10^{-6}\right) \quad (7)$$

$$\mathrm{PISER_{nc}} = \frac{C_{\mathrm{risk}}}{RfD_{\mathrm{i}} \times SAF} \times \frac{PM_{10} \times DAIR_{\mathrm{c}} \times PIAF \times ED_{\mathrm{c}} \times (fspo \times EFO_{\mathrm{c}} + fspi \times EFI_{\mathrm{c}})}{BW_{\mathrm{c}} \times AT_{\mathrm{nc}}} \times 10^{-6} \quad (8)$$

For the industrial scenario, for the carcinogenic and noncarcinogenic effects of heavy metals, the lifetime hazard and harm of human exposure in adulthood were considered, respectively.

Risk from oral intake of remediated soil:

$$\mathrm{OISER_{ca}} = C_{\mathrm{risk}} \times S_{\mathrm{Fo}} \times \frac{OSIR_{\mathrm{a}} \times ED_{\mathrm{a}} \times EF_{\mathrm{a}}}{BW_{\mathrm{a}} \times AT_{\mathrm{ca}}} \times \mathrm{ABS_o} \times 10^{-6} \quad (9)$$

$$\mathrm{OISER_{nc}} = \frac{C_{\mathrm{risk}}}{RfD_{\mathrm{o}} \times SAF} \times \frac{OSIR_{\mathrm{a}} \times ED_{\mathrm{a}} \times EF_{\mathrm{a}}}{BW_{\mathrm{a}} \times AT_{\mathrm{nc}}} \times \mathrm{ABS_o} \times 10^{-6} \quad (10)$$

Risk from skin contact of remediated soil:

$$\mathrm{DCSER_{ca}} = C_{\mathrm{risk}} \times S_{\mathrm{Fd}} \times \frac{SEA_{\mathrm{a}} \times SSAR_{\mathrm{a}} \times EF_{\mathrm{a}} \times E_{\mathrm{v}} \times ABS_{\mathrm{d}}}{BW_{\mathrm{a}} \times AT_{\mathrm{ca}}} \times 10^{-6} \quad (11)$$

$$\mathrm{DCSER_{nc}} = \frac{C_{\mathrm{risk}}}{RfD_{\mathrm{o}} \times SAF} \times \frac{SEA_{\mathrm{a}} \times SSAR_{\mathrm{a}} \times EF_{\mathrm{a}} \times ED_{\mathrm{a}} \times E_{\mathrm{v}} \times ABS_{\mathrm{d}}}{BW_{\mathrm{a}} \times AT_{\mathrm{nc}}} \times 10^{-6} \quad (12)$$

Risk from direct inhalation of remediated soil particles:

$$\mathrm{PISER_{ca}} = C_{\mathrm{risk}} \times S_{\mathrm{Fi}} \times \frac{PM_{10} \times DAIR_{\mathrm{a}} \times PIAF \times ED_{\mathrm{a}} \times (fspo \times EFO_{\mathrm{a}} + fspi \times EFI_{\mathrm{a}})}{BW_{\mathrm{a}} \times AT_{\mathrm{ca}}} \times 10^{-6} \quad (13)$$

$$\mathrm{PISER_{nc}} = \frac{C_{\mathrm{risk}}}{RfD_{\mathrm{i}} \times SAF} \times \frac{PM_{10} \times DAIR_{\mathrm{a}} \times PIAF \times ED_{\mathrm{a}} \times (fspo \times EFO_{\mathrm{a}} + fspi \times EFI_{\mathrm{a}})}{BW_{\mathrm{a}} \times AT_{\mathrm{nc}}} \times 10^{-6} \quad (14)$$

$$\mathrm{SAE_c} = 239 \times H_c^{0.417} \times BW_c^{0.517} \times SER_c \quad (15)$$

$$\mathrm{SAE_a} = 239 \times H_a^{0.417} \times BW_a^{0.517} \times SER_a \quad (16)$$

In the formulas, $OISER_{ca}$, $DCSER_{ca}$ and $PISER_{ca}$ represent the carcinogenic risk from oral intake, skin contact and direct inhalation, respectively, and $OISER_{nc}$, $DCSER_{nc}$, and $PISER_{nc}$ represent the noncarcinogenic risk from oral intake, skin contact and direct inhalation, respectively. For Zn, which is a noncarcinogen, only the noncarcinogenic effects were calculated in this evaluation. In general, the level of harm caused by human exposure to noncarcinogenic heavy metals through a single route is ultimately characterized by the hazard quotient. Furthermore, the level of human exposure to noncarcinogenic heavy metals is characterized by the sum of the hazard quotients of a population exposed to a single

Table 3 Exposure parameters and reference values for human health risk calculation.

Exposure parameter	Description	Unit	Reference value	
			Residential scenario	Industrial scenario
$OSIR_c$	Daily intake of contaminated soil by children	mg d^{-1}	200	–
$OSIR_a$	Daily intake of contaminated soil by adults	mg d^{-1}	100	100
ED_c	Exposure duration of Children	a	6	–
ED_a	Exposure duration of adults	a	24	25
EF_c	Childhood exposure factor	d/a	350	–
EF_a	Adult exposure factor	d/a	350	250
BW_c	Childhood body weight	kg	19.2	–
BW_a	Adult body weight	kg	61.8	61.8
ABS_o	Dermal absorption factor	–	1	1
AT_{ca}	Average time of carcinogenic effects	d	27740	27740
AT_{nc}	Average time of noncarcinogenic effects	d	2190	9125
$SSAR_c$	Soil adhesion coefficient of skin surface in children	mg/cm	0.2	–
$SSAR_a$	Soil adhesion coefficient of skin surface in adults	mg/cm	0.07	0.2
ABS_d	Skin contact absorption efficiency factor	–	1	1
E_v	Frequency of daily skin contact events	n/d	1	1
SAE_c	Exposed skin surface area for children	cm^2	2848	–
SAE_a	Exposed skin surface area for adults	cm^2	5374	3023
SER_c	Area ratio of exposed skin for children	–	0.36	–
SER_a	Area ratio of exposed skin for adults	–	0.32	0.18
SAF	Reference dose coefficient for exposure to soil	–	1	1
H_c	Average height of children	cm	113.15	–
H_a	Average height of adults	cm	161.5	161.5
PM_{10}	Amount of particulate matter in the air	mg cm^{-3}	0.119	0.119
$DAIR_c$	Daily air breathing volume for children	m^3 d^{-1}	7.5	–
$DAIR_a$	Daily air breathing volume for adults	m^3 d^{-1}	14.5	14.5
$PIAF$	Retention rate of particulate matter in the body	–	0.75	0.75
$fspi$	Indoor air comes from the proportion of particulate matter	–	0.8	0.8
$fspo$	Outdoor air comes from the proportion of particulate matter	–	0.5	0.5
EFO_c	Outdoor exposure frequency for children	d/a	87.5	–
EFO_a	Outdoor exposure frequency for adults	d/a	87.5	62.5
EFI_c	Indoor exposure frequency for children	d/a	262.5	–
EFI_a	Indoor exposure frequency for adults	d/a	262.5	187.5

Table 4 Noncarcinogenic reference dose and carcinogenic slope factor for different heavy metals.

Heavy metal	Noncarcinogenic reference dose (mg kg^{-1} d^{-1})			Carcinogenic slope factor (mg kg^{-1} d^{-1})		
	Oral (RfD_o)	Skin (RfD_d)	Inhalation (RfD_i)	Oral (S_{Fo})	Skin (S_{Fd})	Inhalation (S_{Fi})
Cu	4.00E-02	4.00E-02	–	–	–	–
Zn	3.00E-01	3.00E-01	3.00E-01	–	–	–
Pb	1.40E-04	1.40E-04	–	–	–	–
Cd	3.00E-03	2.50E-05	5.71E-05	–	–	6.30E + 00
Ni	2.00E-02	8.00E-04	2.06E-02	1.70E + 00	4.25E + 00	9.01E-01
As	3.00E-04	3.00E-04	3.00E-04	1.50E + 00	3.66E + 00	1.50E + 00
Cr	1.50E + 00	1.95E-02	2.86E-05	–	–	–
Hg	1.60E-04	1.60E-04	8.75E-05	–	–	–

heavy metal in multiple ways, namely, the hazard index. Finally, the acceptable hazard index for a single heavy metal must be less than 1. All the exposure parameters and reference values are presented in Tables 3 and 4.

Based on the one-step and two-step risk value calculation methods, the human health risk of remediated soil was evaluated. The calculated human health risks of the remediated soil samples are shown in Fig. 5. The human health risks of remediated soils treated by thermal curing are less than 1, which corresponds to the safe range. When using the one-step risk value calculation method, as the leaching agent pH value gradually increased, the human health risks of Zn gradually decreased. For the residential scenario, the hazard index of Zn was 2.15E-04, 6.94E-05, 4.58E-05, 3.44E-05, 3.61E-05 and 3.88E-05, and for the industrial scenario, the hazard index of Zn was 2.61E-05, 8.42E-06, 5.56E-06, 4.17E-06, 4.38E-06 and 4.72E-06 at pH = 2 to pH = 12. When the two-step risk content calculation method was used, for the residential scenario, the hazard index of Zn was 2.45E-04, 1.61E-04, 1.55E-04, 1.54E-04, 1.54E-04 and 2.12E-04, and for the industrial scenario, the hazard index of Zn was 2.98E-05, 1.96E-05, 1.89E-05, 1.87E-05, 1.87E-05 and 2.57E-05 at pH = 2 to pH = 12.

The human health risks determined using the two-step calculation method were higher than those obtained using the one-step calculation method. This aspect was especially true under strong alkali conditions (pH = 10 and pH = 12), in which the human health risk value increased by 4.3 and 5.4 times for the residential and industrial scenarios, respectively. Notably, the parameter requirements of the industrial scenario are not as strict as those of the residential scenario in the latest technical guidelines for risk assessment of soil contamination of land for construction in China (HJ 25.3-2019). The calculated human health risks of the industrial scenario were lower than those of the residential scenario. The human health risk evaluation results of the two-step calculation method were more conservative than those of the one-step method. The two-step method can thus facilitate the protection of human health against remediated soils at reuse sites.

4 Conclusions

Remediated soil treated by thermal curing exhibited strong

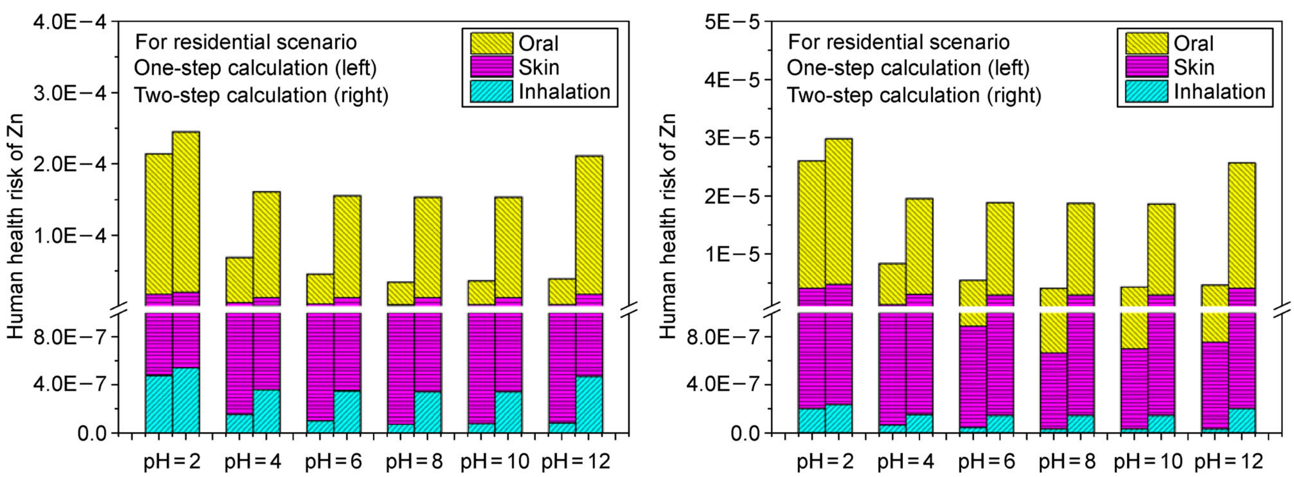

Fig. 5 Human health risk against Zn in remediated soil for residential and industrial scenarios.

inherent resistance to acidic attack with the formation of $ZnCr_2O_4$ spinel. The fraction distribution characteristics of Zn in the remediated soil showed that the leaching agent pH value mainly affected the acid-soluble fraction content but did not significant influence the other three fractions. Moreover, a strong complementary relationship was observed between the leaching content and acid-soluble fraction content, which indicated that the sum of these two parameters is more representative than either individual parameter of the risk value of the remediated soil. Based on this characteristic, we proposed a two-step calculation method to calculate the sum of the leaching and acid-soluble fraction contents of heavy metals as the risk value of remediated soil treated by thermal curing. This method was further combined with the modified ecological risk evaluation model and human health risk evaluation model proposed by the technical guidelines for the risk assessment of soil contamination of land for construction in China to evaluate the ecological and human health risk of remediated soil. Compared with the traditional one-step calculation method, which considers only the leaching content as the risk value, this two-step calculation method can effectively avoid underestimating the risk of remediated soils, especially in alkaline conditions.

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