Cement-fly ash stabilisation/solidification of contaminated soil: Performance properties and initiation of operating envelopes

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Abstract

This study was aimed at evaluating the mechanical and pH-dependent leaching performance of a mixed contaminated soil treated with a mixture of Portland cement (CEMI) and pulverised fuel ash (PFA). It also sought to develop operating envelopes, which define the range(s) of operating variables that result in acceptable performance. A real site soil with low contaminant concentrations, spiked with 3,000 mg/kg each of Cd, Cu, Pb, Ni and Zn, and 10,000 mg/kg of diesel, was treated with one part CEMI and four parts PFA (CEMI:PFA = 1:4) using different binder and water contents. The performance was assessed over time using unconfined compressive strength (UCS), hydraulic conductivity, acid neutralisation capacity (ANC) and pH-dependent leachability of contaminants. With binder dosages ranging from 5 – 20% and water contents ranging from 14 – 21% dry weight, the 28-day UCS was up to 500 kPa and hydraulic conductivity was around 10^{-8} m/s. With leachant pH extremes of 7.2 and 0.85, leachability of the contaminants was in the range: 0.02 - 3,500 mg/kg for Cd, 0.35 – 1,550 mg/kg for Cu, 0.03 – 92 mg/kg for Pb, 0.01 – 3,300 mg/kg for Ni, 0.02 – 4,010 mg/kg for Zn, and 7 – 4,884 mg/kg for total petroleum hydrocarbons (TPH), over time. Design charts were produced from the results of the study, which show the water and/or binder proportions that could be used to achieve relevant performance criteria. The charts would be useful for the scale-up and design of stabilisation/solidification (S/S) treatment of similar soil types impacted with the same types of contaminants.

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Research Highlights

- Cement and fly ash were used for treatment of a mixed contaminated soil.
- The stabilisation treatment was evaluated by mechanical and leaching tests.
- The binder effectively reduced the leachability of most metals.
- The binder has the potential to maintain acceptable leachability levels over time.
- Design charts were produced to assist in optimisation of treatment process design.

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1 Introduction

Soil contamination is a widespread problem, often arising from different industrial activities on the site, leading in many cases to a mixture of contaminants in the soil. Contaminated land is typically remediated to address environmental risks and risks to site users. The remediation of contaminated land is generally perceived as a sustainable process as it encourages the recycling of land and regeneration of urban areas, thus minimising greenfield development (Harbottle et al., 2005). Stabilisation/solidification (S/S), which usually employs the addition of cementitious binders to the contaminated soils, has emerged as a cost effective and efficient remedial measure for the treatment of contaminated soils. However, the performance of S/S treated soils depends on a large number of variables, including the characteristics of the waste and binder, and the formulation. Therefore, this work has attempted to develop operating envelopes, that is, the range of operating conditions that result in acceptable performance, for S/S treatment of contaminated soils using different cementitious binder systems such as ordinary Portland cement (CEMI), mixtures of lime or CEMI and ground granulated blast furnace slag (slag) (Kogbara et al., 2010, 2011; Kogbara and Al-Tabbaa, 2011). This paper deals with evaluation of the mechanical and pH-dependent leaching performance of contaminated soil treated with a mixture of CEMI and pulverised fuel ash (PFA), which is also known as fly ash, and initiates the development of operating envelopes for S/S treatment with the binder.

PFA is commonly blended with cement for geotechnical soil stabilisation. As PFA is a by-product, it is much cheaper than cement. Hence, the more cement can be replaced by PFA for satisfactory soil stabilisation, the more economical the operation becomes (Bell, 1994). In comparison with sand-cement grouts, the final properties of PFA grouts take longer to develop, but include reduced hydraulic conductivity and increasing compressive strength and durability (UK Quality Ash Association (UKQAA), 2006). Very few published studies have deployed cement-PFA mixtures for treatment of contaminants in soils although they have been used to treat other hazardous waste streams and for ground improvement works. In a certain study, PFA has been activated by lime for treatment of contaminated soil (Dermatas and Meng, 2003). Consequently, information on the granular leaching behaviour of cement-PFA-treated contaminated soils under different pH conditions is rare in the literature. Leachability as a function of pH is important as many contaminants are less mobile under alkaline conditions, but the initial alkalinity of treated soils is reduced over time by acidic influences in the environment. The main finding from most of the studies that have used CEMI-PFA mixtures to treat contaminated soils is that metal leachability decreases with curing age in contaminated soils treated with 10 and 20% binder dosage of the total dry mass (Akhter et al., 1990; Al-Tabbaa et al., 1998; Antemir et al., 2005; Perera and Al-Tabbaa, 2005; Moon et al., 2010).

Therefore, the aim of the present study was to investigate the range of operating conditions for effective S/S treatment of contaminated soil using a CEMI–PFA mixture. Specifically, the study sought to investigate the effect of the water and binder proportions used in the treatment process (i.e., the primary independent variables) on selected mechanical and leaching properties of the treated soil, with an emphasis on the leaching behaviour of six contaminants commonly found in soils, under different pH conditions.

2 Materials and methods

2.1 *Contaminated soil and binder*

The study sought to employ a real site soil contaminated with metals and organics, in order to have something close to a typical contaminated soil, which is usually characterized by the presence of both types of contaminants. However, the site soil used, which was obtained from a service station in Birmingham, UK, contained low levels of metals and total petroleum hydrocarbons (TPH). The soil was clayey silty sandy gravel comprising 65% gravel, 29% sand, 2.8% silt and 3.2% clay; with a natural water content of ~ 12% and very low (0.22%, dry weight basis) organic C content. Since the soil did not contain significant concentrations of metals, it was, therefore, spiked in small batches of ~3 kg with five metals, added as reagent grade chemical compounds (all supplied by Fisher Scientific), and diesel to increase the contaminant levels to relatively high values for monitoring during the course of the study. The choice of 3,000 mg/kg of metals and 10,000 mg/kg of TPH was generally on the relatively high side compared to typical contamination levels found in soils (Breckenridge and Crockett, 1995). Table 1 details the contaminant compounds spiked. The concentrations of contaminants recovered from the spiked soil (particle size < 20 mm), which was crushed to < 2 mm for analysis, through acid digestion for metals and solvent extraction for TPH are also shown in Table 1. The pH of the contaminated soil was highly alkaline at 9.83, probably due to the association between sodium and carbonate species in the soil. The determinants of high soil alkalinity have been extensively discussed (Brautigan, 2010), and alkaline soils make up one third of the world's soils (Guerinot, 2007).

A blend of CEMI (Lafarge, UK) and PFA (supplied by the UKQAA) was used as the binder. The mix ratio was one part CEMI and four parts PFA (i.e., CEMI:PFA = 1:4, w/w). The mix proportion was chosen based on binder screening for a parallel study on S/S of metal treatment sludges (Stegemann and Zhou, 2008), and also work by Arora and Aydilek (2005). Table 2 shows the pertinent physico-chemical properties of the soil and binder components. The PFA is a by-product from coal fired power station electricity generation, conforming to BS EN 450-1 (BSI, 2005) and is classified as low lime PFA or Class F fly ash according to ASTM C618-03 (ASTM, 2003). Information on the concentration of trace metals in PFA is not available in the technical data sheet. However, the UKQAA states that trace metals contained in fly ash are bound into a glassy matrix preventing them from leaching from the material. Moreover, less than 1% of fly ash is soluble in water, of which the water soluble material consists of SO_4^{2-} from gypsum and limited amounts of alkalis.

2.2 *Preparation of treated contaminated soil samples*

The procedure for preparing the contaminated soil entailed thorough mixing of the diesel and the soil, and addition of solutions of the metallic compounds made with de-ionised water. Further mixing was carried out until the mix appeared homogenous. Thereafter, the contaminated soil was stored in a sealed container for about 2 h. Soil only samples were taken for analysis at this point before binder addition. The binder constituents, CEMI and PFA, were then mixed together and de-ionised water added to form a paste. The binder paste was then applied to the contaminated soil in 5, 10 and 20% dosages (dry weight), and mixed thoroughly.

Standard Proctor compaction test (BS 1377-4: BSI, 1990) with a 2.5 kg rammer was used to determine the density-moisture content relationship and the optimum moisture content (OMC) of the contaminated soil-binder mixtures. The compacted mixtures were then broken up and cast into cylindrical moulds, 50 mm diameter and 100 mm high. The stabilised/solidified products described earlier were then prepared at the maximum dry density (MDD) and OMC and at other density-moisture content points determined in the compaction test. The moulded samples were demoulded after 3 days and cured at 95% relative humidity and 20°C until tested.

2.3 *Testing methodologies*

The main tests carried out on the stabilised/solidified contaminated soil samples were unconfined compressive strength (UCS) (ASTM D1633-00, 2000), hydraulic conductivity (ASTM D5084-03, 2003) and acid neutralisation capacity (ANC) (Stegemann and Côté, 1991) with determination of contaminant leachability at 0, 1 and 2 meq/g acid additions. The choice of only three acid additions was for consistency with parallel projects on development of operating envelopes for different waste types (Stegemann and Zhou, 2008; Lampris et al., 2009). Furthermore, to help in assessment of chemical immobilisation of metals within a given pH zone, the ANC test on the untreated contaminated soil included two more acid and two base additions (NaOH) in addition to the three acid additions used for the treated soils. This was done in order to cover the full pH range attained by the treated soils.

The curing time before testing was 7, 28, 49 or 84 days. Testing started with low binder dosage (5%) with assessment of granular leachability of contaminants in the ANC test until most granular leaching waste acceptance criteria (WAC) for different types of landfill (UK Environment Agency, 2006) were met. Hence, performance parameters were not determined on the highest binder dosage (20%) used, at all of the above curing ages. The UCS before immersion and ANC tests were determined on triplicate samples, but hydraulic conductivity and UCS after immersion were determined on duplicate samples. The detailed procedures used for the tests are described in previous related publications (Kogbara and Al-Tabbaa, 2011; Kogbara et al., 2011).

2.4 *Statistical analysis and contour map plotting*

One and two-way analysis of variance (ANOVA) were used to test for statistically significant differences in performance parameters due to differences in water and binder proportions used, acid addition in the ANC, and curing age. The Kolmogorov-Smirnov test was used to check for normal distribution of data prior to use of ANOVA. Contour maps (design charts) were plotted from experimental results to represent operating envelopes for selected performance parameters. These were produced using Origin 8.6 software (OriginLab Corporation, Northampton, USA) and the kriging correlation, which best fits the data points among the gridding methods provided by the software package, was used to determine the contour intervals.

3 Results and discussion

Where applicable, the discussion of results here focuses on the values of performance parameters obtained at 28 and 84 days, which represent standard and extended curing ages, respectively, in the cement and concrete industry. The 7 and 49-day values are presented to show the early-age performance of the stabilised/solidified products and the development of a performance parameter over time. Performance parameters were determined only for the OMC mixes at 49 and 84 days since initial results of the 7 and 28-day old samples of 5 and 10% binder dosage mixes showed that the best performance was obtained around the OMC. It is for the same reason that only the OMC mix was used for the 20% binder dosage. Hence, the 20% dosage mix has fewer points than the lower binder dosages in the following graphs. Where applicable, the error bars in the graphs represent the standard deviation from the mean values.

3.1 *Compaction behaviour*

Figure 1 shows the compaction behaviour of the stabilised/solidified contaminated soils in relation to the untreated soil. Binder addition to the soil increased the MDD of the soil but there was no significant effect of binder addition on the soil's OMC. The OMC was 0.16 for 5 and 10% binder dosages and 0.165 for 20% binder dosage, respectively. There were no clearly apparent trends in the variation of OMC and MDD with water content and binder dosage. In the absence of literature on CEMI-PFA-treated contaminated soils, these results differ from those observed for CEMI-PFA uncontaminated clay soils (Bell, 1994), where the OMC increased and the MDD decreased with CEMI-PFA addition. As might be expected, the compaction behaviour of soil upon CEMI-PFA addition depends on soil type and composition.

3.2 *Unconfined compressive strength*

The variation of UCS with water content at 7 and 28 days is shown in Figure 2a. The UCS values were generally very low with most values < 100 kPa. This is because PFA addition does not result in high strength in this timeframe. UCS values largely depend on the cement addition. Moreover, the presence of contaminants can be expected to decrease strengths. Interestingly, the highest water content of the 10% binder mix exhibited a 28-day UCS value about five times the average value for 10% binder mixes and even slightly higher than the 20% binder OMC mix (Fig. 2a). Generally, there was significant increase in 28-day UCS with increase in binder ($P < 0.001$) and water proportions. However, although the UCS of 10% dosage mixes increased significantly ($P < 0.001$) with water content, that of 5% dosage mixes was not statistically significant ($P = 0.44$). On one hand, this differs from the position of Arora and Aydilek (2005) who observed a higher UCS on the dry side of OMC for uncontaminated silty sand. Nevertheless, it tends to follow the observation of Horpibulsuk et al. (2009) in which the maximum strength of uncontaminated clay treated with fly ash blended cement was observed on the wet side of OMC (i.e. at 1.2 times the OMC), thereafter, strength decreased with increasing water content. It is likely that the interaction of the contaminants, especially the diesel, with the soil and binder caused the change in the normal strength-moisture content relationship for uncontaminated sandy soils. On the other hand, it is consistent with a previous study (Al-Tabbaa et al., 2000) that recorded increase in UCS with increase in CEMI-PFA dosage.

It is also interesting to note that the difference between 7 and 28-day UCS values widens with increasing water content (Fig. 2a). There was relatively little strength gain between 7 and 28-days in 5 and 10% binder products, except for the wettest mix of 10% dosage (Fig. 2a, mix with w/s ratio of 0.2), but the UCS of the 20% binder product quadrupled between the said periods (see Fig. 2a and Fig. 2b). The UCS of 5% binder products at 49 and 84 days could not be determined as the OMC products crumbled. Figure 2b also shows the UCS after immersion of only the 49-day old 10% binder product, since the 5% binder product could not be determined. The UCS was halved with immersion in water. The above findings suggest that with lower binder dosages, more water simply made the soil-binder particles sticky enough to stay put although there was improper hydration in such samples. This led to a decrease in strength and immersion in water has more deleterious effect on the samples.

3.3 *Hydraulic conductivity*

Figure 3 shows the hydraulic conductivity of some of the products. Those not shown could not be determined because these products crumbled. The hydraulic conductivity of the 10% binder product decreased slightly with time unlike in the work of Al-Tabbaa and Evans (2000), which recorded increase in hydraulic conductivity over time. As the present observation was not statistically significant and to obtain further data on this interesting possible trend, the hydraulic conductivity of the 10 and 20% binder OMC products was continued after 1 year, and has been included in Figure 3.

The hydraulic conductivity of the 20% binder product after 1 year increased significantly by about an order of magnitude over its 28-day value; that of the 10% binder product remained lower than its 28-day value, but the latter observation was not statistically significant. Generally, the hydraulic conductivity fluctuated around 10^{-8} m/s, which is typical of cement-based stabilised/solidified products, but higher than the UK and United States Environment Protection Agency (USEPA) 10^{-9} m/s limit (Al-Tabbaa and Stegemann, 2005). Hydraulic conductivity generally correlated with the UCS as the Pearson correlation of available data revealed a moderate negative correlation ($r = -0.65$). It appears the hydraulic conductivity was better (lower) at higher than OMC just as was the case for the UCS.

3.4 *ANC and leachability of contaminants*

Leachability testing was carried out on only three of the four water contents studied: the OMC and the next successive water contents on the dry and wet side of the OMC. Figures 4 – 6 show contaminant concentration as a function of leachate pH, for the various stabilised/solidified products tested after 28 days curing. Three points are shown for each solidified product, from left to right representing the leachate pH values measured at 2, 1 and 0 meq/g HNO_3 additions. There is a smooth curve in each graph, except the TPH graph in Figure 6b. It is the theoretical solubility profile of the metal hydroxide (Stegemann, 2005) based on data in the MINTEQ database, a chemical equilibrium model for predicting metal speciation and solubility (Allison et al., 1990). The leachability of the untreated contaminated soil at different acid and base additions (apart from TPH) is also shown. For comparison, the pHs of the binder alone (without contaminated soil) at 0, 1 and 2 meq/g acid additions were 12.8, 12.4 and 11.0, respectively.

The following general observations can be made from the graphs in Figures 4 – 6. There was no significant effect of w/s ratio on leachate pH and leachability of contaminants ($p > 0.1$ in all cases) at all three acid additions. There was no influence of binder dosage on TPH leachability (Fig. 6b) similar to previously reported observations for lime-stabilised soil (Schifano et al., 2005). In addition to the above, some more general observations are summarised as follows:

- In most cases, treatment with 10 and 20% binder dosage increased the leachate pH and hence, reduced the leachate concentrations of the contaminants below that of the untreated soils, but leachate pH and leachate concentrations in 5% dosage mixes were similar to those in the untreated soil.

- With acid addition, the potential for leachability reduction declined due to the relatively low buffering capacity of the binder and only 20% dosage was still effective at 2 meq/g acid addition. Thus, the results showed that the only benefit of additional binder addition is the ANC.

As expected, the leachability of the CEMI-PFA stabilised/solidified soil was largely pH-controlled. The leaching behaviour of the contaminants in the soil-binder system was largely similar to those in the authors' previous related publications on slag-cement and lime-slag binders, where they were discussed in depth (Kogbara and Al-Tabbaa, 2011; Kogbara et al., 2011). Hence, only a phenomenological description of the controlling factors for contaminant leachability is made here. The results in Figure 4a show that Cd leachability was generally similar to that of the untreated soil under the influence of acid or base addition, especially with lower binder dosage. This demonstrates the influence of pH on leachability of the metal. Moreover, at pH 11.5, there were slight differences in leachability between the untreated soil, and the OMC mixes of 10 and 20% dosage. Copper leachability more closely followed its hydroxide profile: this is typical of samples with low organic matter, such as this one (Li et al., 2001).

The trend of Pb leachability in the treated soils in the high pH zone (9.5 – 11.5) did not closely follow the theoretical solubility profile of $Pb(OH)_2$ (Stegemann, 2005) where Pb leachability increases between pH 10 and 11 (Fig. 5a). Although, with NaOH addition, the Pb leachability of the untreated soil did not increase within the said pH zone, it increased between pH 11.5 and 12.5 in line with the $Pb(OH)_2$ solubility profile. The leachability of most mixes was less than that of the untreated soil at the same pH. This confirms the position of Dermatas and Meng (2003) that PFA addition to CEMI increases the immobilisation pH region for Pb. However, in some cases, there seems to be no influence of the binder on Pb immobilisation as Pb leachability in the untreated soil was similar to, or even less than those of S/S soils. Nevertheless, the leachability of the majority of the mixes in the pH zone, 10 – 11.5 was less than that of the untreated soil in the same pH zone, which suggests the potential for Pb immobilisation by CEMI-PFA at high pH (Fig. 5a). The treated soil mixes leached out higher concentrations of Ni at alkaline pH than the estimated solubility limits for $Ni(OH)_2$ (Stegemann, 2005) but the leachate concentrations between pHs 5 and 7 were about the estimated limits (Fig. 5b). Although the exact mechanism responsible for the above was not investigated, it is probably because most of soluble Ni is present as carbonate complexes, which are more soluble than $Ni(OH)_2$ (Christensen et al., 1996). A similar observation was made for cement-slag mixtures used to treat the same soil (Kogbara and Al-Tabbaa, 2011). There was evidence of chemical immobilisation of the metal at pH 10 and 11.5, where Ni has minimum solubility (Fig. 5b) similar to the observation for Cd.

Zinc also showed a similar behaviour to Ni at pH 10 and 11.5, which supports the earlier position on the similarity of the leaching trends for the more mobile metals. Furthermore, the amphoteric behaviour of Zn where leachability increases at > pH 11 was not observed in the treated soil mixes due to the pH regime of the binder. Moreover, Zn concentration in the 20% dosage mix was less than that in the untreated soil for all three acid additions (Fig. 6a), which suggests the potential for chemical immobilisation of the metal by the binder with the formation of more crystalline phases at higher binder dosage. The leachability of TPH in the mixes was independent of pH (Fig. 6b) as was observed for the same soil treated with slag-cement and lime-slag binders (Kogbara and Al-Tabbaa, 2011; Kogbara et al., 2011).

The variation of contaminant leachability with pH over time in OMC mixes of the stabilised/solidified soil is shown in Figures 7 – 9. The graphs are essentially similar to those described in Figures 4 – 6 except that, here, OMC mixes alone are compared over time, since the results showed no significant difference in leachability due to moisture content. There was generally a decrease in pH of the OMC mixes between 7 and 28 days, no clear trend between 28 and 49 days and a further decrease in the majority of the samples between 49 and 84 days. This is expected as the initial alkalinity of stabilised/solidified materials is neutralised by acidic influences in the environment. Generally, there was no significant difference ($p > 0.1$ in all cases) in the leachate concentrations of the metals beyond 28 days curing age, except for the leachability of Cd, Ni and Zn in the driest mix of the 10% binder dosage between 28 and 49 days (Figs 7a, 8b and 9a). Zinc leachability decreased between 7 and 28 days in the 20% binder dosage mix in the high pH (~12) zone, which is in line with the observations of Moon et al (2010). Similarly, there were slight decreases in Cu and Pb leachability over time in the high pH zone in line with the findings of Chitambira (2004) and Perera and Al-Tabbaa (2005).

3.5 *Design charts representing operating envelopes for UCS, leachate pH and leachability*

Design charts based on contour maps for two major performance parameters considered in this work, namely UCS and leachability, are discussed in this section in order to illustrate the operating envelopes for the parameters. Hydraulic conductivity is not included here due to paucity of experimental data points.

Figure 10a shows the design chart for 28-day UCS. The horizontal bars on the chart represent the range of water contents at which laboratory data exists for the binder dosages studied, and shows the limit of accuracy of the contouring process. Figure 10b shows how the chart works. The arrows on the figure show how to deduce the operating envelope for 28-day UCS of CEMI-PFA S/S soil based on the Environment Canada 440 kPa UCS performance threshold (Stegemann and Côté, 1996) for controlled utilisation. None of the mixes met the 1 MPa UK Environment Agency (2006) UCS standard for landfill disposal. Detailed performance thresholds for mechanical and leaching behaviour that can be used with the charts to deduce operating envelopes have been provided in previous related publications (Kogbara and Al-Tabbaa, 2011; Kogbara et al., 2011). Figure 10b shows that, based on the data obtained, the minimum binder dosage that would be required to achieve the 440 kPa UCS threshold is ~ 9% CEMI-PFA dosage and the w/s ratio must be around 0.2. If a lower w/s ratio is to be used, the binder dosage required will significantly increase.

The results of the study show that the influence of water content on leachability is insignificant within the workable range of water contents. Hence, contaminant leachability is mainly determined by the binder dosage applied and the pH of the leachate. However, the leachate pH is a function of binder addition and is not a controllable parameter as it is rather an output of the system, which cannot be known before the binder dosage to be used is determined. Thus the contaminant concentration in the leachate cannot be extrapolated unambiguously.

Consequently, in the context of a design chart for leachability, the pH of the leachant (i.e. the deionised water and acid solutions) is the major parameter to consider since it is independent of binder dosage and is a controllable parameter, which can be set by the designer through suitable modulation of acid concentration. In fact, in practice, the leachant pH represents ‘acidic influences’ in the environment, examples include carbonation by CO2 uptake and natural leachants like rain water and landfill leachate. The pHs of the leachants used in the leaching tests were 7.2, 1.2 and 0.85 for deionised water with 0, 1 and 2 meq/g acid addition, respectively.

In the light of the above, Figure 10c shows the design chart for the leachate pH extrapolated based on its operating parameters, leachant pH and binder dosage. The construction(s) in the figure illustrates how it works. Assuming a natural leachant such as pristine rain water (leachant pH widely accepted as 5.6), will consistently get into contact with soil treated with 9% CEMI-PFA dosage, over a long time frame, it can be deduced from the construction with solid lines in Figure 10c that the pH of the treated soil will drop to ~9.9. This will in turn determine the concentration of contaminants leaching from it; hence, the design charts for leachate pH are to be used together with those for leachability of contaminants (Fig. 11). It should be noted that the design charts for leachate pH as well as those for contaminant leachability were produced using data from all water contents and curing ages studied since there were no significant differences in the performance properties due to the effect of both parameters.

In the design charts for leachability of contaminants shown in Figure 11, each of the six contour maps in the figure represents one of the contaminants studied. The contour map in Figure 11a has arrows unlike the other contour maps as it is used as an example to illustrate how the chart can be used to deduce operating envelopes. Although there are no performance thresholds for contaminant leachability under different pH conditions (or acid additions), as such thresholds only exist for leaching with deionised water only, Figure 11a shows how the design chart can be used to determine contaminant leachability. With the 9% minimum binder dosage being considered appropriate to satisfy the strength criteria above, consistent leaching by a leachant with pH of 5.6 will lead to a Cd concentration < 1 mg/kg (Fig. 11a, construction with solid lines). For instance, this will satisfy the existing granular leachability criteria of 1.0 mg/kg Cd for stable non-reactive hazardous waste in non-hazardous landfill waste acceptance criteria. For the 9% binder dosage and leachant pH of 5.6, the other leachabilities are: Cu < 1 mg/kg, Pb < 0.1 mg/kg, Ni ~ 100 mg/kg, Zn < 1 mg/kg, and TPH ~ 150 mg/kg (see Figs 11b – f).

Comparing Figures 10c and 11a (constructions with dashed lines), it can be deduced that the leachate pH of the soil treated with 10% binder dosage must be lowered to ~8.8 before its Cd leachability will reach the aforementioned performance threshold of 1.0 mg/kg. That would require a leachant with pH of ~3.8. Thus, an insight into the long-term leaching performance of the binder can also be deduced from the design charts as the above example is similar to what happens when progressive carbonation eventually reduces the pH of stabilised materials to about 8 - 9 (Arickx et al., 2010). It is worth mentioning that spiking of contaminants used here would make the contaminants, especially the metals, more mobile than if they were present in a real contaminated soil. Thus, in using the design charts, it should be noted that the leaching results here provide higher estimates of typical leachability values encountered in real environments.

Furthermore, a design chart for contaminated soil is soil-specific as it depends on the properties of the particular soil used. However, it is documented that even in widely different classes of waste materials, including contaminated soil, leaching trends show similar characteristics and are controlled by very few parameters such as pH, redox potential and complexation (van der Sloot et al., 1996). Hence, the charts here can be used to estimate likely contaminant leachability ranges in other soils, and would also provide conservative estimates for the UCS since a real contaminated soil with weathered hydrocarbons would be stronger than one with fresh contamination used here (Al-Sanad and Ismael, 1997).

4 Conclusions

This research has provided valuable data and useful insight into the performance properties of a mixed contaminated soil treated with cement-fly ash and has attempted to develop operating envelopes for cement-fly ash S/S treatment of a particular type of contaminated soil that can broadly apply to similar soil types impacted with the same type of contaminants. Although, the soil available for use in the study was alkaline and the initial pH of the soil has a significant effect on leachability, the findings of this study would still be very useful due to the extent of alkaline soils around the world. The strengths and weaknesses of the binder formulation used have been shown. The results show that compacting samples around the OMC gives improved performance parameters. Since granular leachability is the most important practical performance parameter from an industrial perspective, the results suggests that with ≥ 20% CEMI-PFA dosage, the binder would significantly reduce the granular leachability of common contaminants in the soil to acceptable levels. The binder also has the potential to maintain acceptable leachability levels over a long period when the pH of the treated material is ultimately lowered by acidic influences in the environment. The results further suggest that performance criteria not met could be satisfied with higher binder dosages and carefully selected water contents.

This work attempted to produce design charts from laboratory data generated. Crucial operating parameters affecting the performance of S/S technology were identified and experimentally investigated. Thereafter, a kind of ‘black box’ model that facilitates the extrapolation of the system’s response to different values of the operating parameters was developed, which would be useful for the scale-up and design of S/S technology. However, since this method is at its initial stage of development, the design charts may not be very reliable for certain water and binder proportions and leachant pH due to limited data for these parameters. Hence, further studies will continue along the lines of improving the reliability of the design charts produced in this study with data from different soil types, ranges of operating variables and performance parameters. Moreover, future studies may also consider validation of the research findings for other contaminant concentrations since contaminant leachability from soil also varies with its initial concentration.

Acknowledgments

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Table 1. Contaminant concentrations in the spiked contaminated real site soil

Contaminant	Compound	Amount of metal or diesel spiked (mg/kg, dry mass)	Amount of contaminant recovered (mg/kg)
Cadmium	$Cd(NO_3)_2.4H_2O$	3,000	3,500 ± 150*
Copper	$CuSO_4.5H_2O$	3,000	3,200 ± 230
Lead	$PbNO_3$	3,000	3,700 ± 210
Nickel	$Ni(NO_3)_2.6H_2O$	3,000	3,600 ± 150
Zinc	$ZnCl_2$	3,000	4,200 ± 290
TPH	Diesel	10,000	6,300 ± 1500

*Results indicate mean ± standard deviation of three replicates

Table 2. Physico-chemical properties of binder constituents

Binder material	Bulk density (kg/m^3)	Specific gravity	Specific surface area (m^2/kg)	pH*
CEMI	1,300 – 1,450	3.15	400	12.80 ± 0.10
PFA	1,100 – 1,700	1.80 – 2.40	3,430	10.22 ± 0.11

Binder material	%								
	CaO	SiO_2	MgO	Al_2O_3	Fe_2O_3	TiO_2	K_2O	SO_3	Cl
CEMI	63.6	13.9	0.6	10.2	2.7	0.8	0.9	6.9	0.02
PFA	1 - 5	45 - 51	1 - 4	27 - 32	7 – 11	0.8 – 1.1	1- 5	0.3 – 1.3	0.05 - 0.15

The data here (except pH) were taken from materials data sheets provided by the suppliers of the binder materials.

* The pH of triplicate samples was determined in a 1:10 cement/PFA: water (deionised water) suspension following BS EN 12457 (BSI, 2002).

Figure captions

Figures 1 – 11 are shown on pages 17 - 27 (PDF file numbering), respectively.

Figure 1. Compaction behaviour of CEMI-PFA stabilised/solidified soil

Figure 2. UCS of CEMI-PFA stabilised/solidified soil (a) at 7 and 28 days (b) in OMC mixes at different curing ages

Figure 3. Hydraulic conductivity of CEMI-PFA stabilised/solidified soil

Figure 4. Leachability in CEMI-PFA stabilised/solidified soil at 28 days for (a) Cd and (b) Cu

Figure 5. Leachability in CEMI-PFA stabilised/solidified soil at 28 days for (a) Pb and (b) Ni

Figure 6. Leachability in CEMI-PFA stabilised/solidified soil at 28 days for (a) Zn and (b) TPH

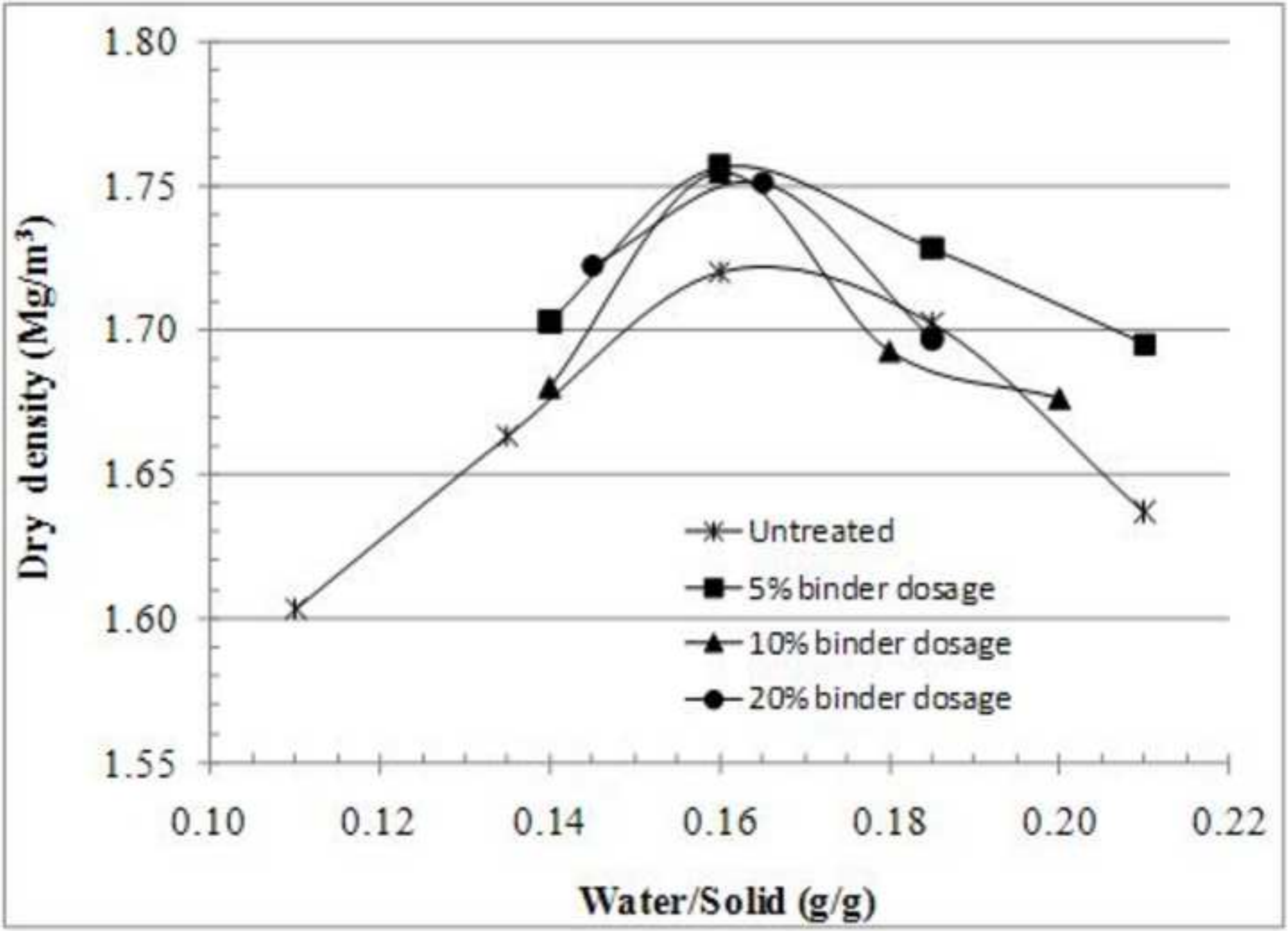
Figure 7. Leachability of (a) Cd and (b) Cu in OMC mixes of CEMI-PFA stabilised/solidified soil at different curing ages

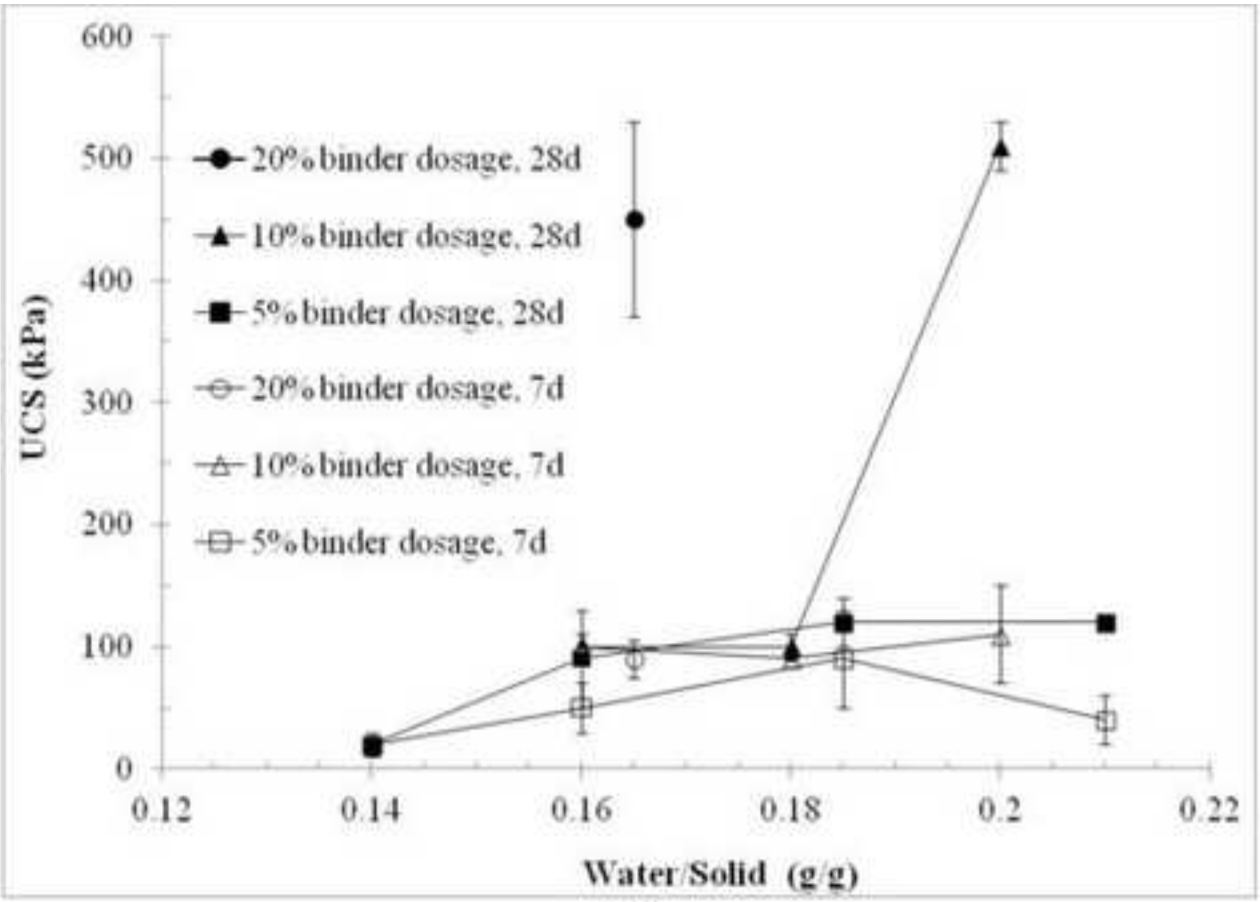
Figure 8. Leachability of (a) Pb and (b) Ni in OMC mixes of CEMI-PFA stabilised/solidified soil at different curing ages

Figure 9. Leachability of (a) Zn and (b) TPH in OMC mixes of CEMI-PFA stabilised/solidified soil at different curing ages

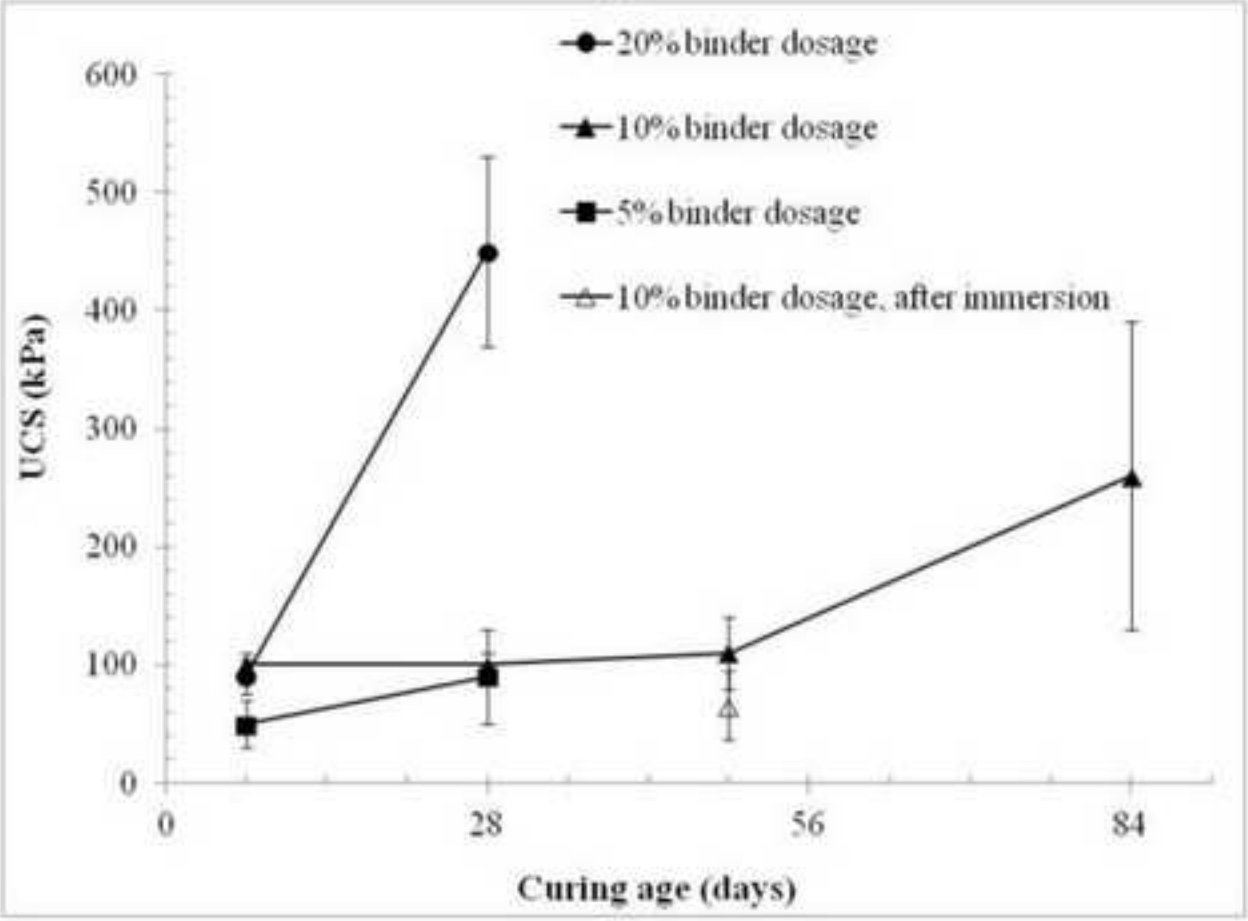
Figure 10. (a) Design chart for 28-day UCS (b) example of how the chart works and (c) leachate pH design chart

Figure 11. Design charts for leachability in CEMI-PFA stabilised/solidified soil of (a) Cd, (b) Cu, (c) Pb, (d) Ni, (e) Zn and (f) TPH

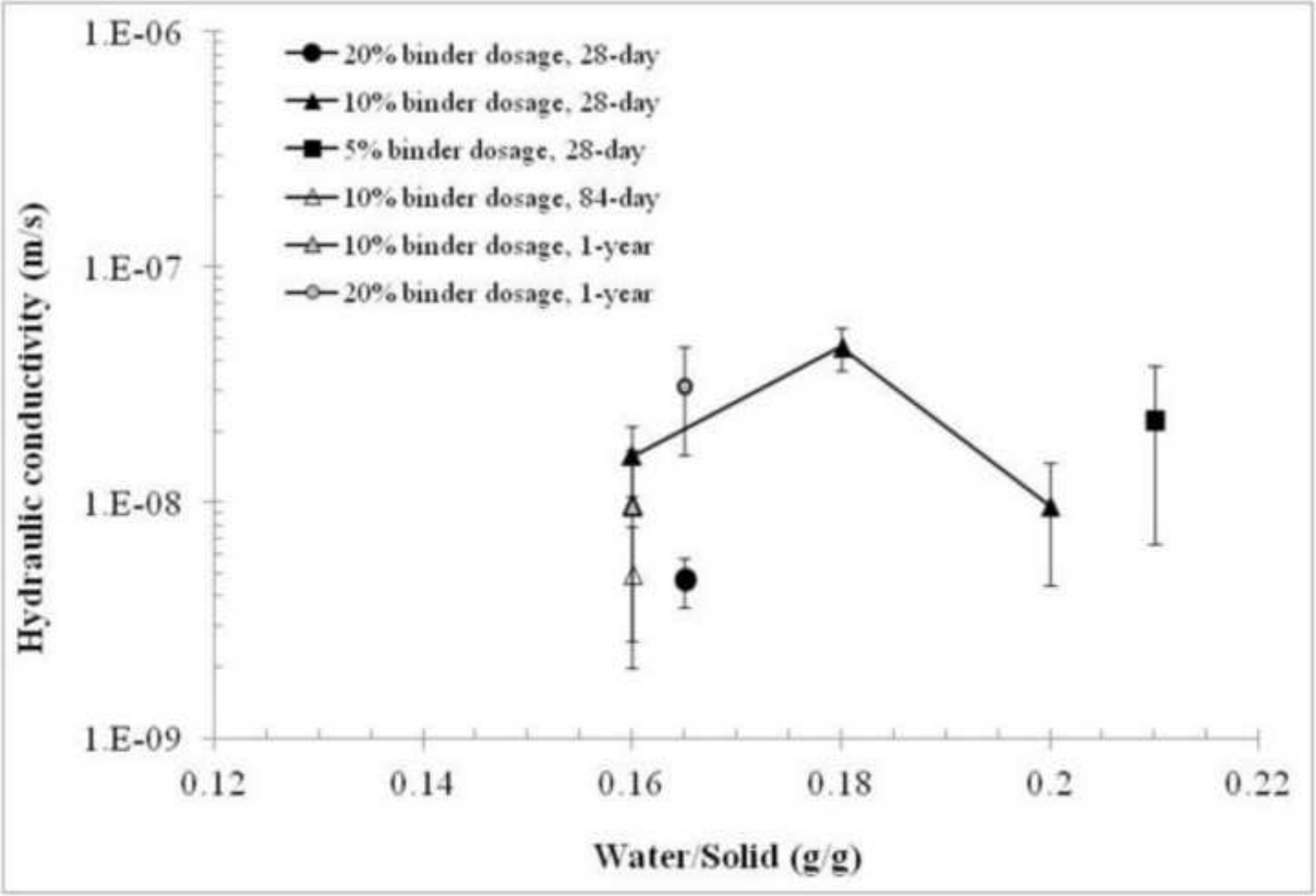

1.80
1.75
1.70
1.65
1.60
1.55
Dry density (Mg/m³)
0.10
0.12
0.14
0.16
0.18
0.20
0.22
Water/Solid (g/g)
Untreated
5% binder dosage
10% binder dosage
20% binder dosage

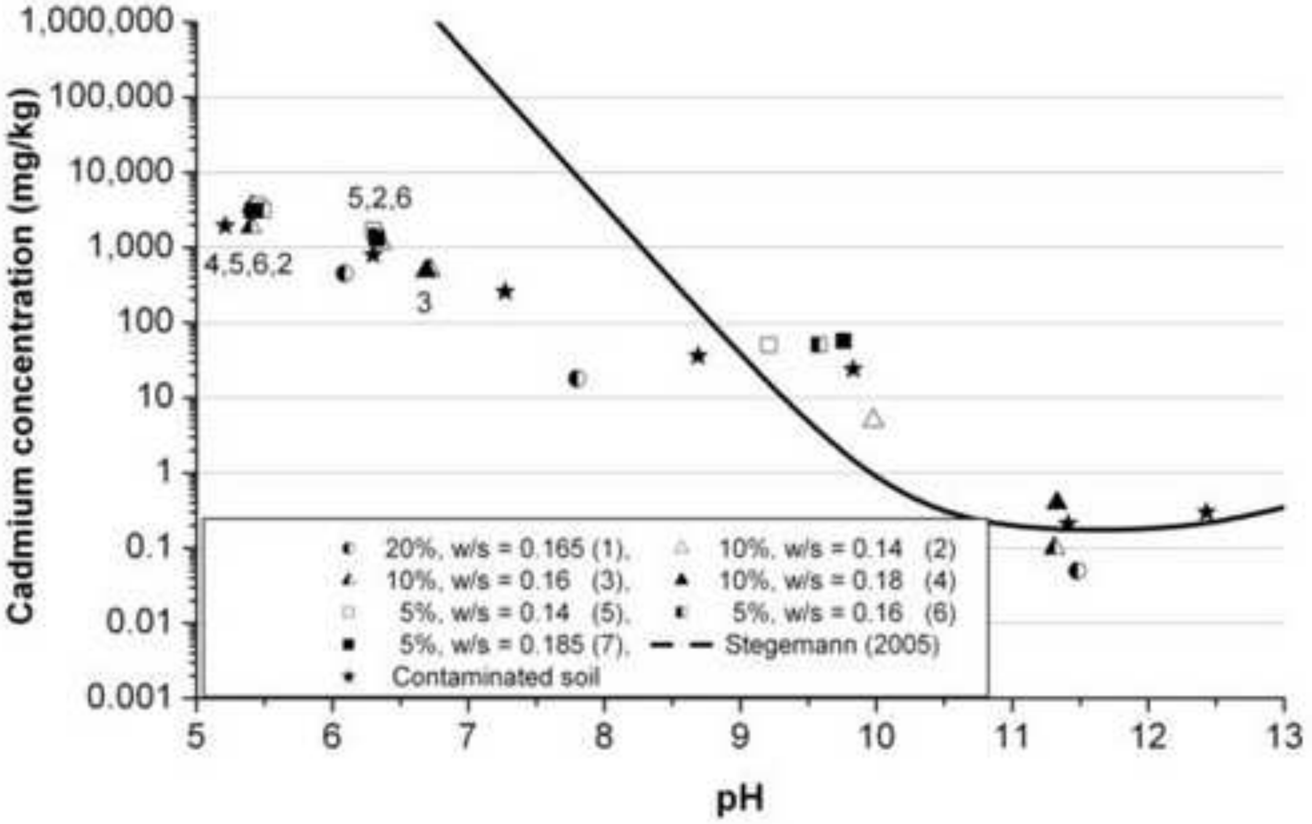

UCS (kPa)
600
500
400
300
200
100
0
0.12
0.14
0.16
0.18
0.2
0.22
Water/Solid (g/g)
20% binder dosage, 28d
10% binder dosage, 28d
5% binder dosage, 28d
20% binder dosage, 7d
10% binder dosage, 7d
5% binder dosage, 7d

(a)

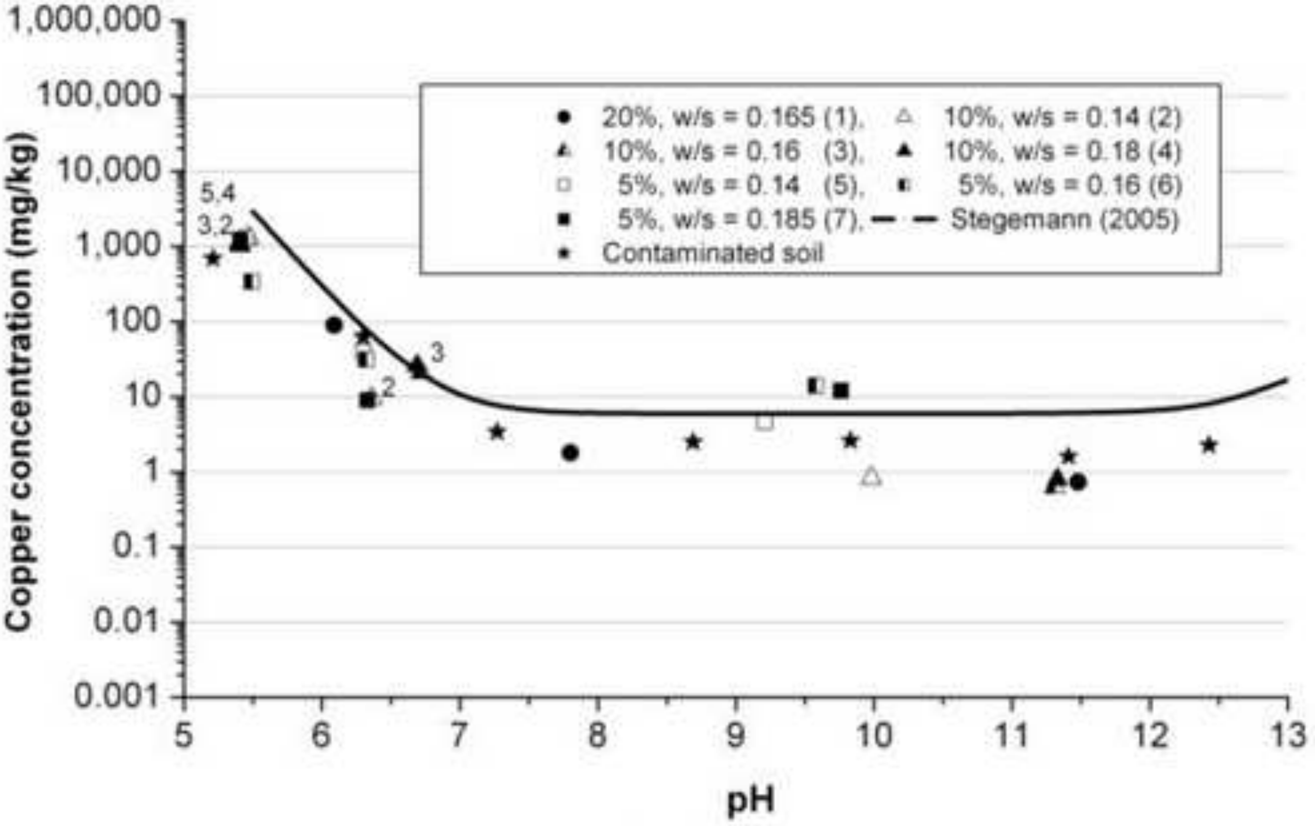

UCS (kPa)
600
500
400
300
200
100
0
0
28
56
84
Curing age (days)
20% binder dosage
10% binder dosage
5% binder dosage
10% binder dosage, after immersion

(b)


1.E-06
1.E-07
1.E-08
1.E-09
Hydraulic conductivity (m/s)
0.12
0.14
0.16
0.18
0.2
0.22
Water/Solid (g/g)
20% binder dosage, 28-day
10% binder dosage, 28-day
5% binder dosage, 28-day
10% binder dosage, 84-day
10% binder dosage, 1-year
20% binder dosage, 1-year

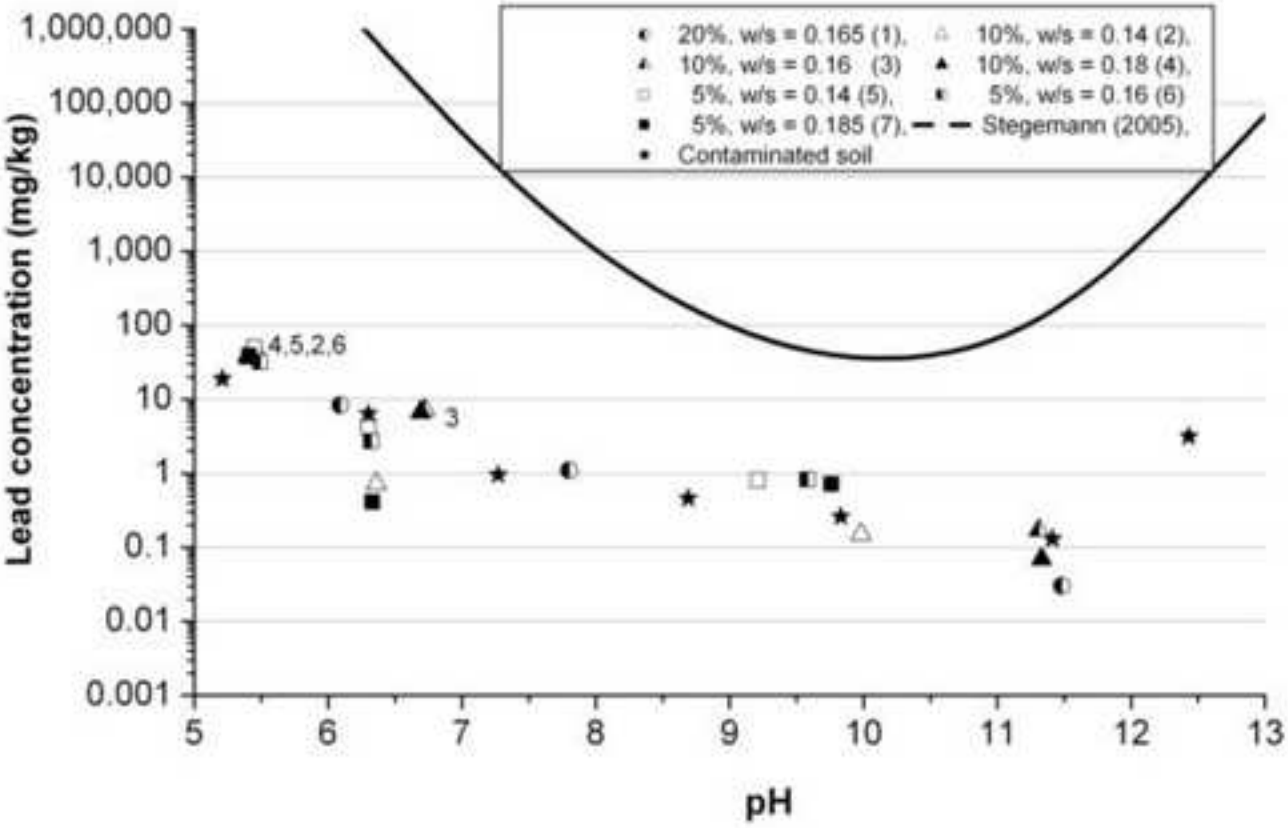

1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
Cadmium concentration (mg/kg)
5
6
7
8
9
10
11
12
13
pH
5,2,6
4,5,6,2
3
20%, w/s = 0.165 (1),
10%, w/s = 0.14 (2)
10%, w/s = 0.16 (3),
10%, w/s = 0.18 (4)
5%, w/s = 0.14 (5),
5%, w/s = 0.16 (6)
5%, w/s = 0.185 (7),
Stegemann (2005)
Contaminated soil

(a)

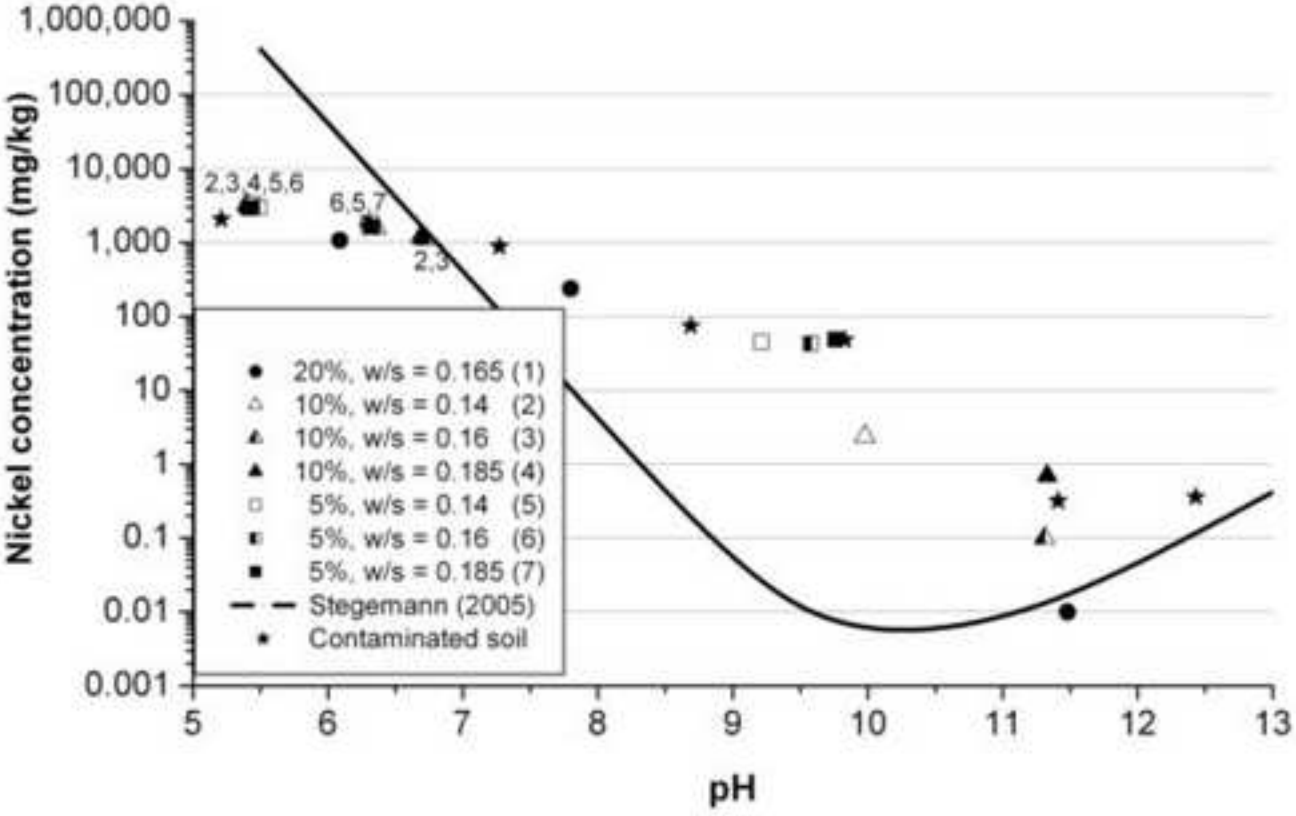

1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
Copper concentration (mg/kg)
5
6
7
8
9
10
11
12
13
pH
5,4
3,2
3
2
20%, w/s = 0.165 (1),
10%, w/s = 0.14 (2)
10%, w/s = 0.16 (3),
10%, w/s = 0.18 (4)
5%, w/s = 0.14 (5),
5%, w/s = 0.16 (6)
5%, w/s = 0.185 (7),
Stegemann (2005)
Contaminated soil

(b)

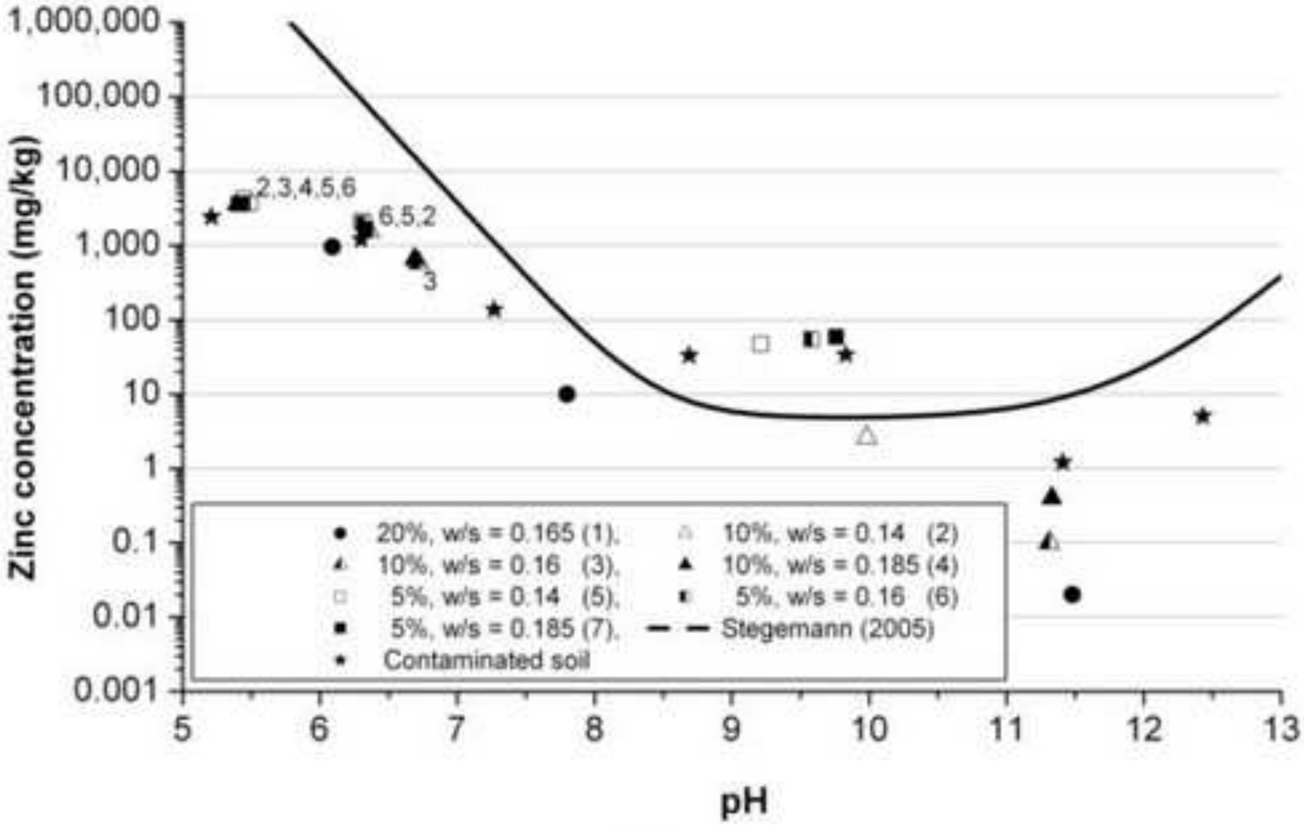
Figure


Lead concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
5
6
7
8
9
10
11
12
13
pH
4,5,2,6
3
20%, w/s = 0.165 (1),
10%, w/s = 0.14 (2),
10%, w/s = 0.16 (3)
10%, w/s = 0.18 (4),
5%, w/s = 0.14 (5),
5%, w/s = 0.16 (6)
5%, w/s = 0.185 (7),
Stegemann (2005),
Contaminated soil

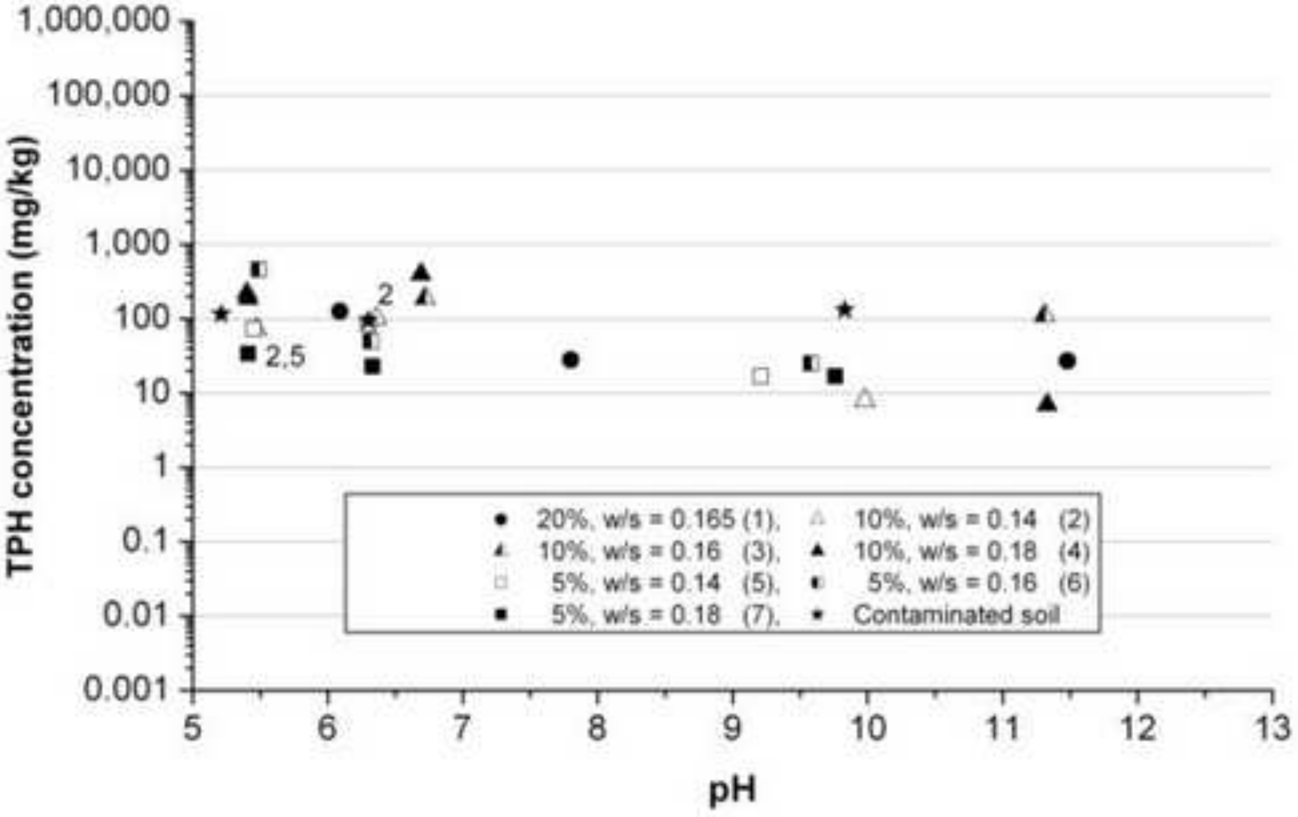
(a)


Nickel concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
5
6
7
8
9
10
11
12
13
pH
2,3,4,5,6
6,5,7
2,3
20%, w/s = 0.165 (1)
10%, w/s = 0.14 (2)
10%, w/s = 0.16 (3)
10%, w/s = 0.185 (4)
5%, w/s = 0.14 (5)
5%, w/s = 0.16 (6)
5%, w/s = 0.185 (7)
Stegemann (2005)
Contaminated soil

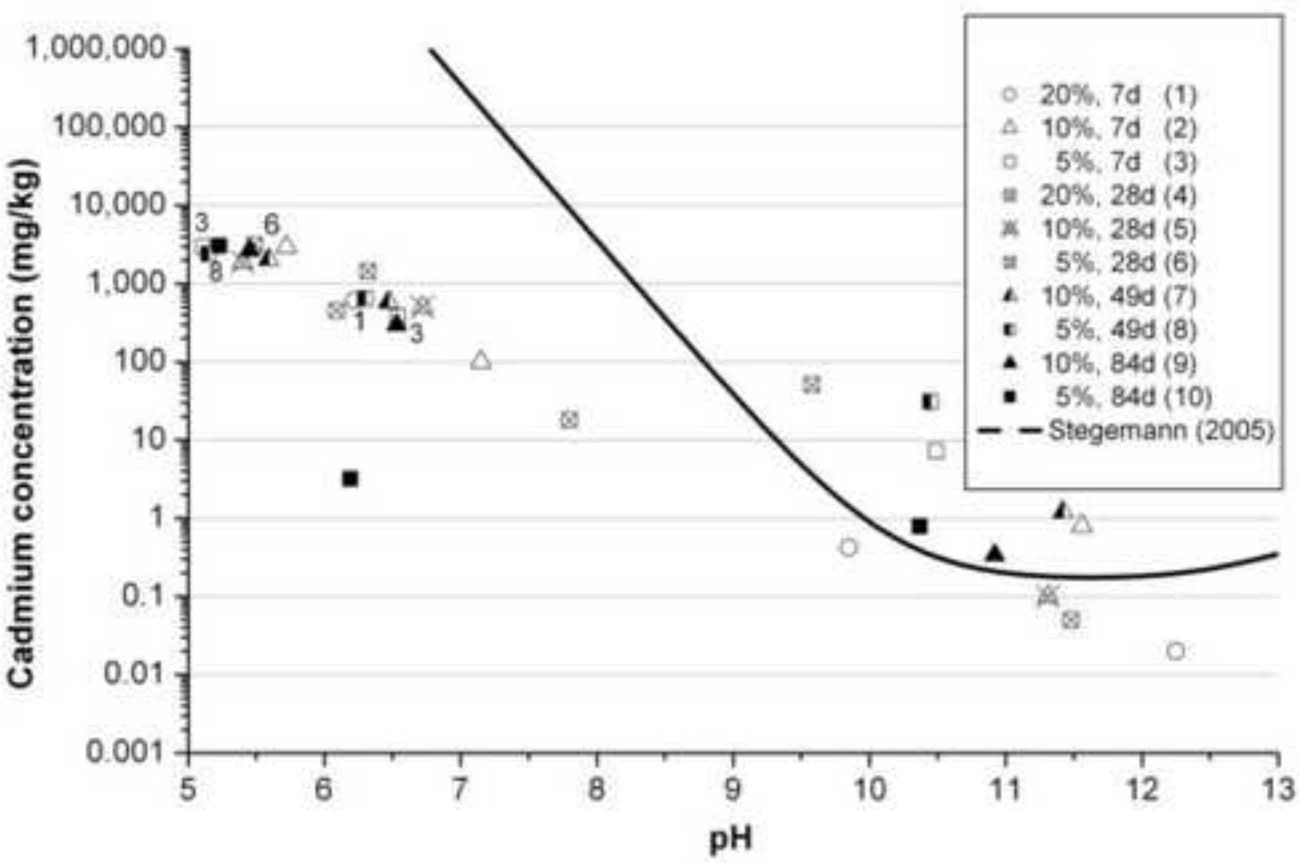
(b)


Zinc concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
pH
5
6
7
8
9
10
11
12
13
2,3,4,5,6
6,5,2
3
● 20%, w/s = 0.165 (1),
△ 10%, w/s = 0.14 (2)
◭ 10%, w/s = 0.16 (3),
▲ 10%, w/s = 0.185 (4)
□ 5%, w/s = 0.14 (5),
◧ 5%, w/s = 0.16 (6)
■ 5%, w/s = 0.185 (7),
— Stegemann (2005)
★ Contaminated soil

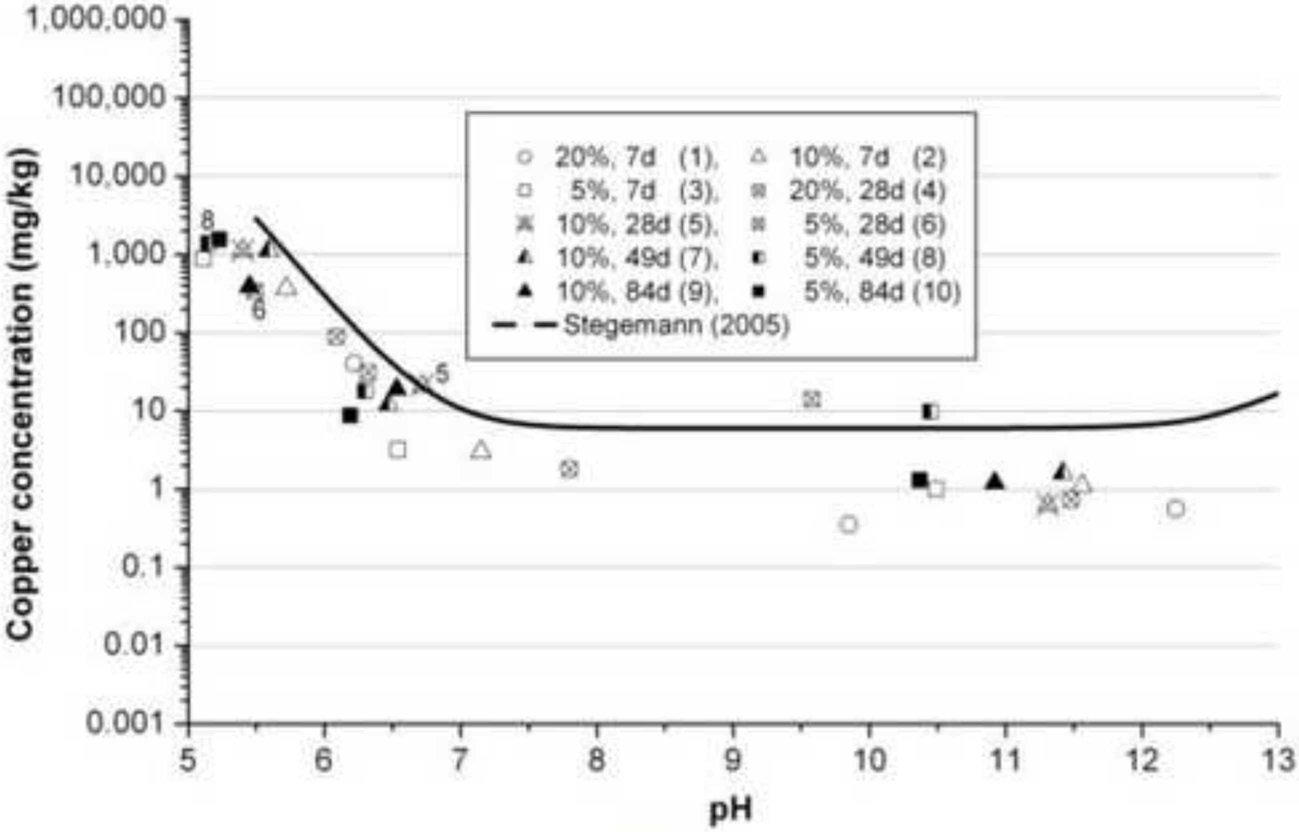
(a)


TPH concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
pH
5
6
7
8
9
10
11
12
13
2
2,5
● 20%, w/s = 0.165 (1),
△ 10%, w/s = 0.14 (2)
◭ 10%, w/s = 0.16 (3),
▲ 10%, w/s = 0.18 (4)
□ 5%, w/s = 0.14 (5),
◧ 5%, w/s = 0.16 (6)
■ 5%, w/s = 0.18 (7),
★ Contaminated soil

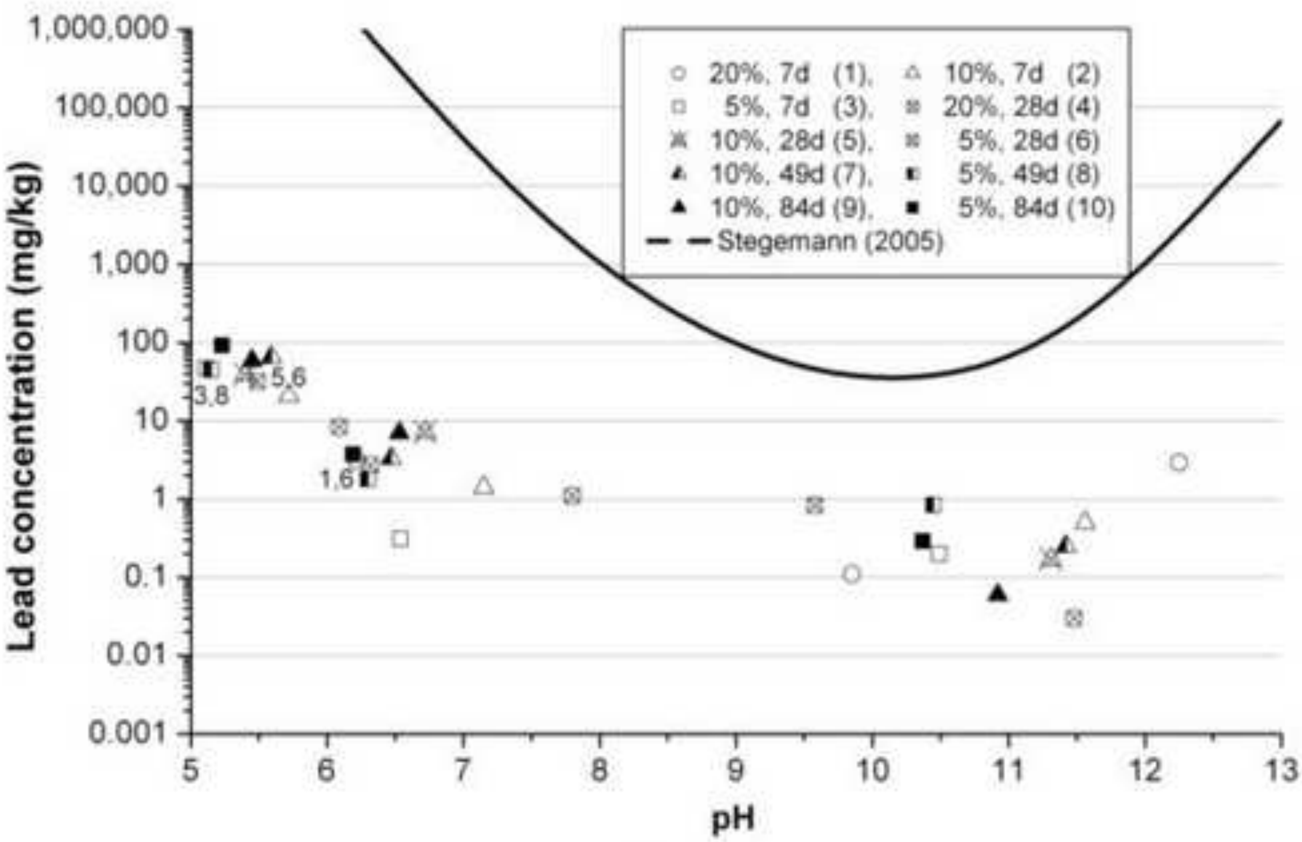
(b)


Cadmium concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
5
6
7
8
9
10
11
12
13
pH
3
6
8
1
3
○ 20%, 7d (1)
△ 10%, 7d (2)
□ 5%, 7d (3)
20%, 28d (4)
10%, 28d (5)
5%, 28d (6)
10%, 49d (7)
5%, 49d (8)
▲ 10%, 84d (9)
■ 5%, 84d (10)
Stegemann (2005)

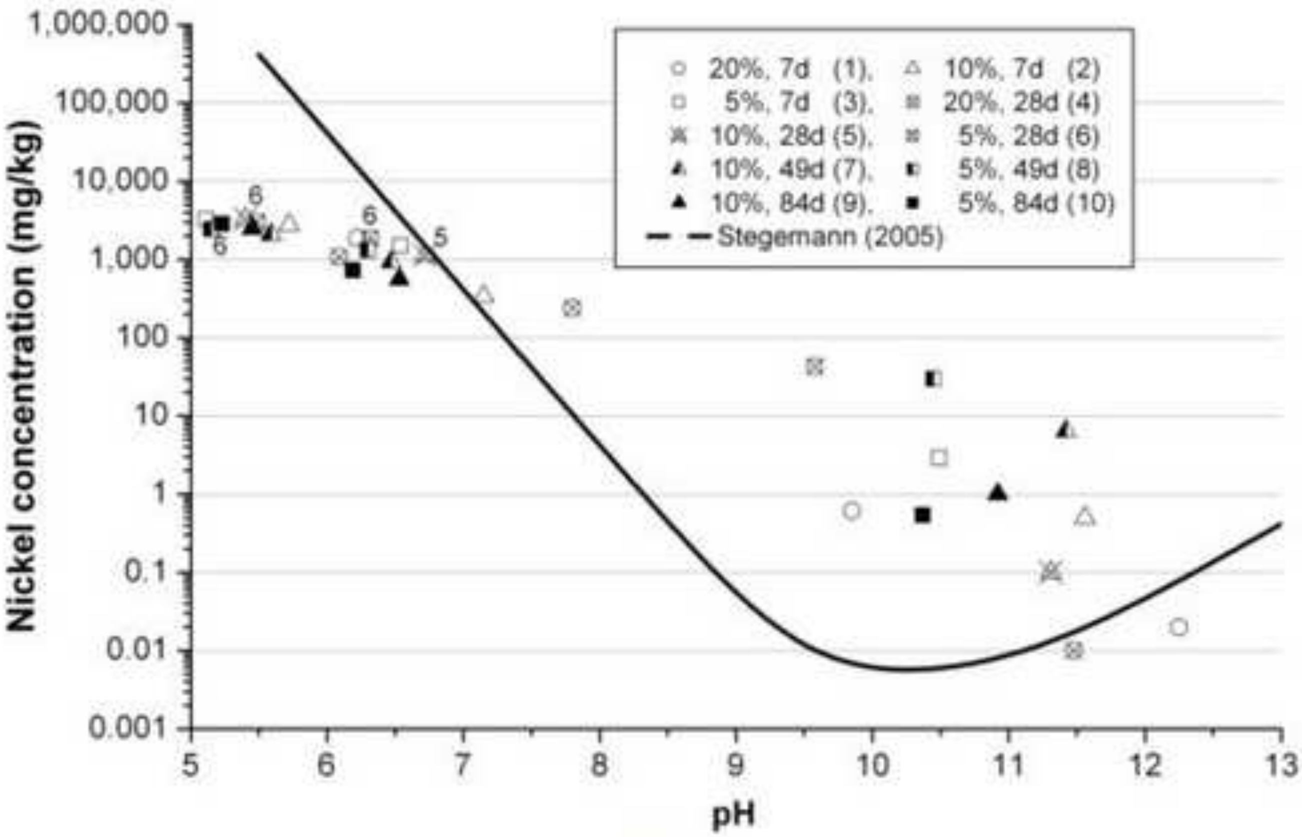
(a)


Copper concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
5
6
7
8
9
10
11
12
13
pH
8
6
5
○ 20%, 7d (1),
△ 10%, 7d (2)
□ 5%, 7d (3),
20%, 28d (4)
10%, 28d (5),
5%, 28d (6)
10%, 49d (7),
5%, 49d (8)
▲ 10%, 84d (9),
■ 5%, 84d (10)
Stegemann (2005)

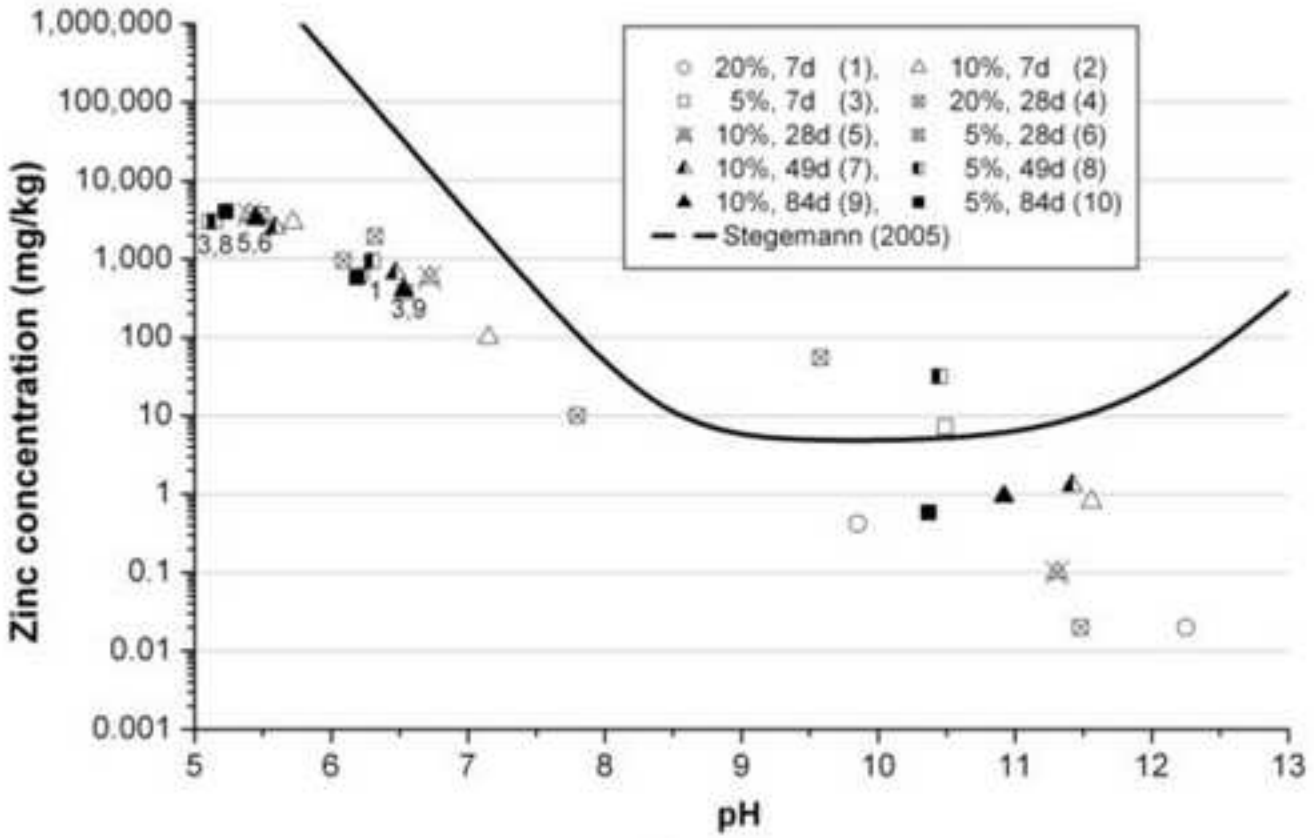
(b)


Lead concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
pH
5
6
7
8
9
10
11
12
13
20%, 7d (1),
10%, 7d (2)
5%, 7d (3),
20%, 28d (4)
10%, 28d (5),
5%, 28d (6)
10%, 49d (7),
5%, 49d (8)
10%, 84d (9),
5%, 84d (10)
Stegemann (2005)
3,8
5,6
1,6

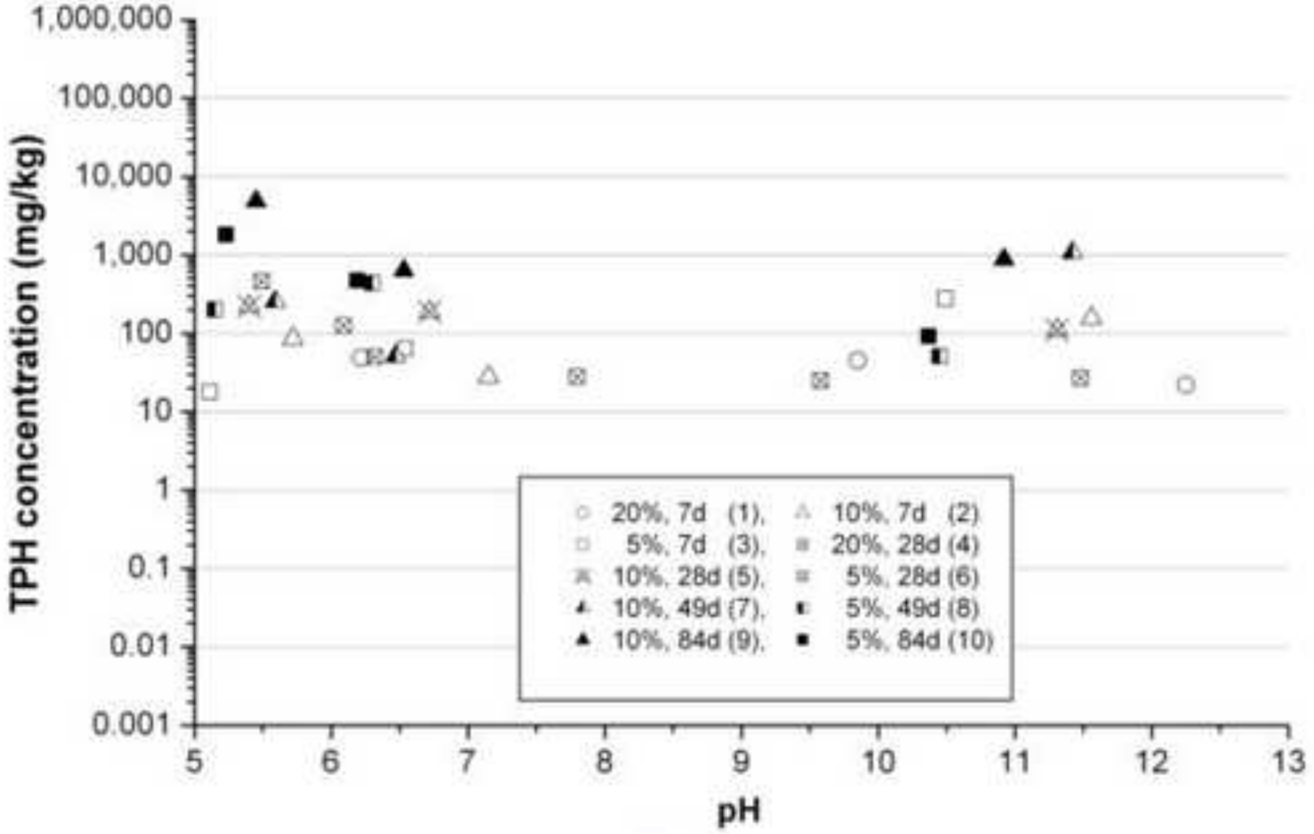
(a)


Nickel concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
pH
5
6
7
8
9
10
11
12
13
20%, 7d (1),
10%, 7d (2)
5%, 7d (3),
20%, 28d (4)
10%, 28d (5),
5%, 28d (6)
10%, 49d (7),
5%, 49d (8)
10%, 84d (9),
5%, 84d (10)
Stegemann (2005)
6
6
6
5

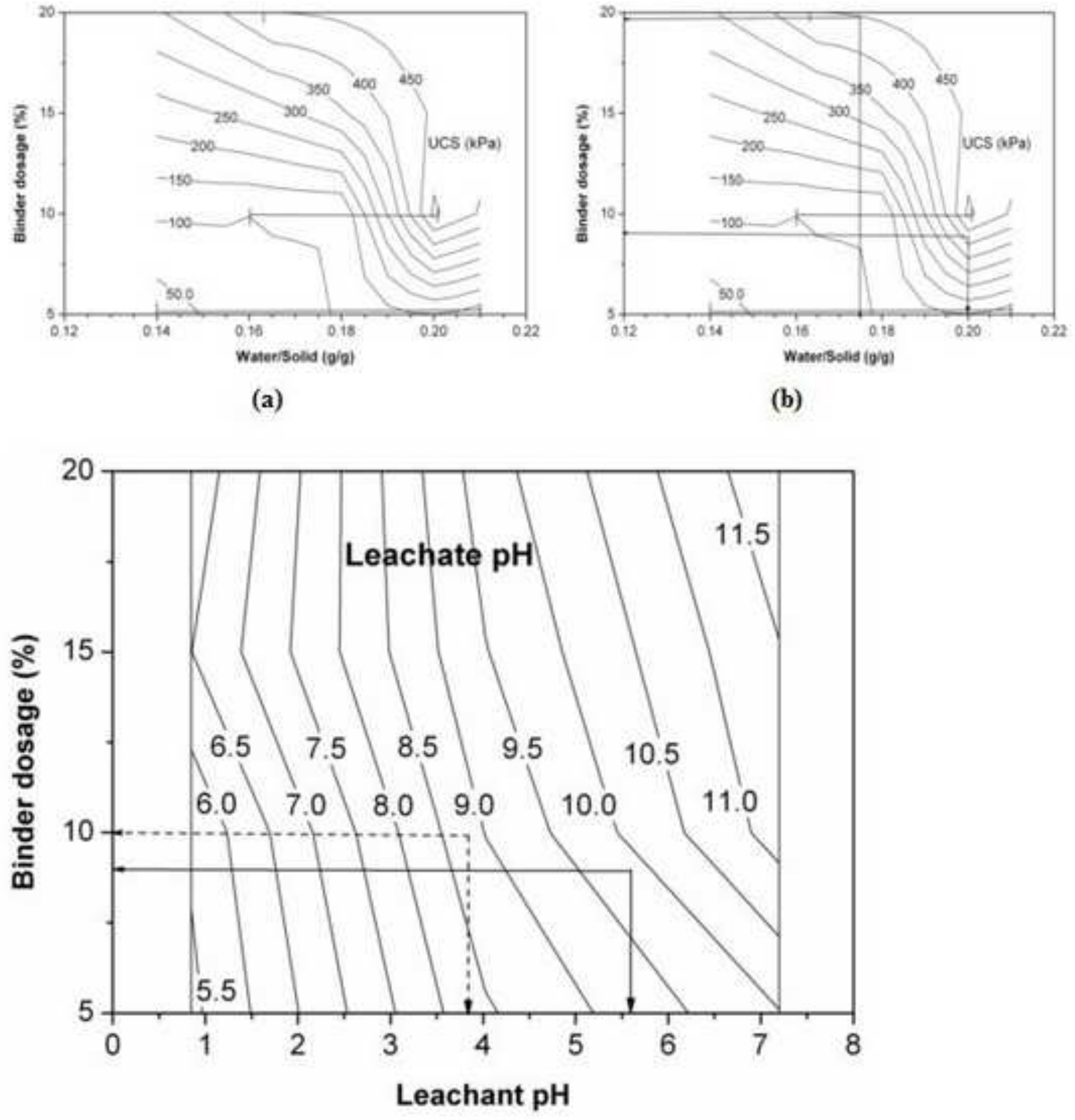
(b)


Zinc concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
pH
5
6
7
8
9
10
11
12
13
20%, 7d (1),
10%, 7d (2)
5%, 7d (3),
20%, 28d (4)
10%, 28d (5),
5%, 28d (6)
10%, 49d (7),
5%, 49d (8)
10%, 84d (9),
5%, 84d (10)
Stegemann (2005)
3,8
5,6
1
3,9

(a)


TPH concentration (mg/kg)
1,000,000
100,000
10,000
1,000
100
10
1
0.1
0.01
0.001
pH
5
6
7
8
9
10
11
12
13
20%, 7d (1),
10%, 7d (2)
5%, 7d (3),
20%, 28d (4)
10%, 28d (5),
5%, 28d (6)
10%, 49d (7),
5%, 49d (8)
10%, 84d (9),
5%, 84d (10)

(b)

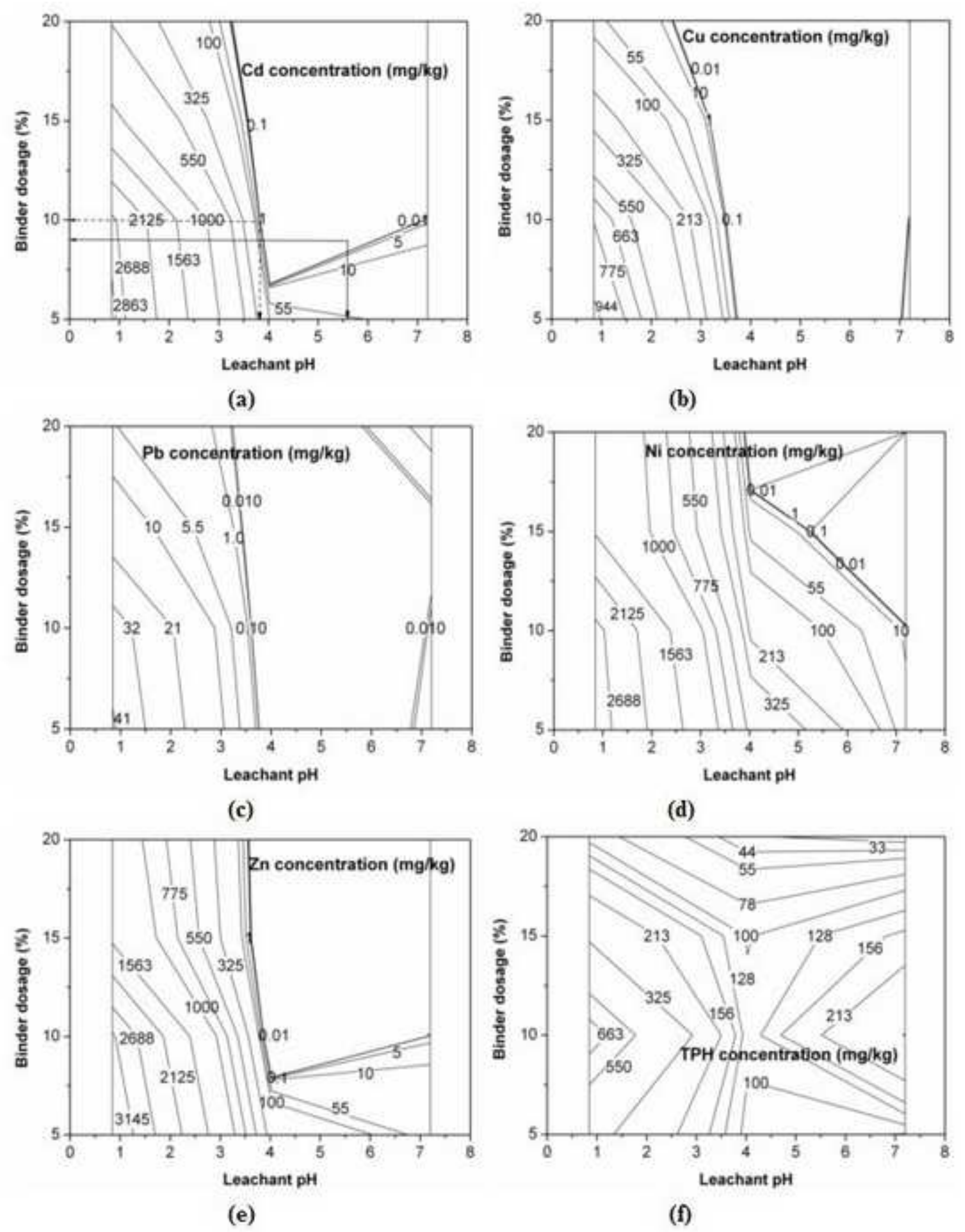

Binder dosage (%)
Water/Solid (g/g)
0.12
0.14
0.16
0.18
0.20
0.22
5
10
15
20
UCS (kPa)
50.0
100
150
200
250
300
350
400
450
Leachate pH
Leachant pH
0
1
2
3
4
5
6
7
8
5.5
6.0
6.5
7.0
7.5
8.0
8.5
9.0
9.5
10.0
10.5
11.0
11.5

(a)

(b)

(c)

Figure



(a)

(b)

(c)

(d)

(e)

(f)