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Leaching of Mercury from Carbonated and Non-Carbonated Cement-Solidified Dredged Sediments

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This study investigated traditional cement-based and non-conventional (using accelerated carbonation) solidification/stabilization to treat 2 dredged sediments contaminated with mercury from two different locations in UK. Canal and estuarine-derived sediments were mixed with blended binders and powdered activated carbon. Fresh mixtures of sediment and cement were exposed to gaseous carbon dioxide and were allowed to carbonate for fixed time periods, after which they were cured for 28 days. Following curing, samples were leach tested to evaluate the fixation of mercury in the treated products. The results obtained indicated that both conventional and accelerated carbonated treatments were capable of reducing the concentration of mercury in the eluates to acceptable limits.

Keywords Sediment, mercury, solidification/stabilization, carbonation

Introduction

Aquatic sediments have been described as the "ultimate sink" or storage place for pollutants (Renholds, 1998). However, re-suspended contaminated sediments are a source for contamination in the aquatic environment. Mercury (Hg) is one of the most hazardous inorganic contaminants often associated with dredged sediment. The primary sources of Hg are various industrial and agricultural processes and it may be found either bound to the organic carbon fraction of the sediment or as a metal sulfide. However, Hg undergoes methylation to form the highly toxic mono- and di-methyl Hg complexes, which can bio-accumulate in organisms, such as fish, and may enter the food chain, causing detrimental effects on human health.

The most common way to manage Hg contaminated sediment is by dredging, dewatering and disposal to landfill. However, dredging encourages re-suspension of fine particles and this enhances methylation (Bloom and Lasorsa, 1999). Thus, *in-situ* remedial technologies such as solidification/stabilization (S/S), which involves encapsulation and/or chemical stabilization, present an alternative approach to contaminated sediment management. The success of S/S, however, depends greatly on the water content of the waste, which

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has a significant effect on the porosity and permeability of the treated product (Glasser, 1997).

A variation of S/S under development involves the accelerated carbonation of materials in the presence of a suitable binder, such as Portland cement or other CO_2 reactive materials. In this application, the cement is freshly applied and the resultant mixture is immediately (and thoroughly) exposed to carbon dioxide gas in a closed environment.

This results in the formation of calcium carbonate and decalcified siliceous reaction products (Lange *et al.*, 1996). The control of moisture in the sample is important as the carbonation reaction is hindered when CO_2 has to diffuse through water-filled pores or when the pores are completely dry (Gervais *et al.*, 2004). As a consequence of this, the high water content of some dredged sediments may require adjustment prior to the application of an accelerated carbonation treatment step.

In the present study, S/S was used to remediate two contrasting Hg-contaminated sediment types obtained from a freshwater canal in Scotland and an estuarine-sediment in S.E. England. The treated products were evaluated by comparing the leach test results to different available guidelines for Hg at the time experiments were conducted.

The first sample was taken from the Union Canal in Scotland (Figure 1a). A 9 km stretch of this canal near Falkirk was heavily contaminated with mercury and was undergoing dredging and pre-treatment, prior to disposal involving S/S by Land and Water Services Ltd. The source of the Hg contamination had been the former Nobel-ICI factory at Reddingmuirhead, which had produced some 73 million detonators containing mercury fulminate.

The second location was the Medway estuary located to the north of Kent (Figure 1b). The contaminated sediments were collected from two different locations in an area known as Bloor Quay. A study of the history of this site revealed that it was used extensively for naval shipbuilding, which has contributed to various heavy metal contamination in the estuary and surrounding area, including Cu, Ni, Hg, Zn and Pb, and which may impact on the general health of biota within the estuary. Where contamination is high (as in several specific areas), this may preclude the beneficial re-use of sediments (Spencer *et al.*, in press). There is also a sewage treatment works located nearby, which until 2001 discharged primary sewage effluent (where only the large particles were removed) into the estuary. The sediment in Bloor Quay is microbiologically impacted by the raw sewage (*Pers. com.* Medway Unitary Authority, Public Health Department) and by former shipbuilding activities and discharges from the coal-fired power plants.

Materials and Methods

Materials

A number of cements were used that were supplied by Lafarge Cements, UK, namely Rockfast A (RF-A), a sulfoaluminate cement, Portland Cement (PC), Sulfate Resisting Portland Cement (SRPC), and a finely ground SRPC (FSRPC). Powdered activated carbon (Fisher Chemicals), pulverized fuel ash, also known as fly ash, (Class-1/ Class-F PFA; ex-British Nuclear Fuel Limited) and quick lime (Buxton Lime Industries Limited) were also used in the study. The analytical reagents employed were Nitric acid (HNO_3)-Aristar grade, Hydrochloric acid (HCl), Stannous (II) Chloride-Aristar grade, Potassium permanganate ($KMnO_4$), Sulfuric acid (H_2SO_4) and Hydroxy ammonium chloride, which were obtained from Fisher Chemicals.

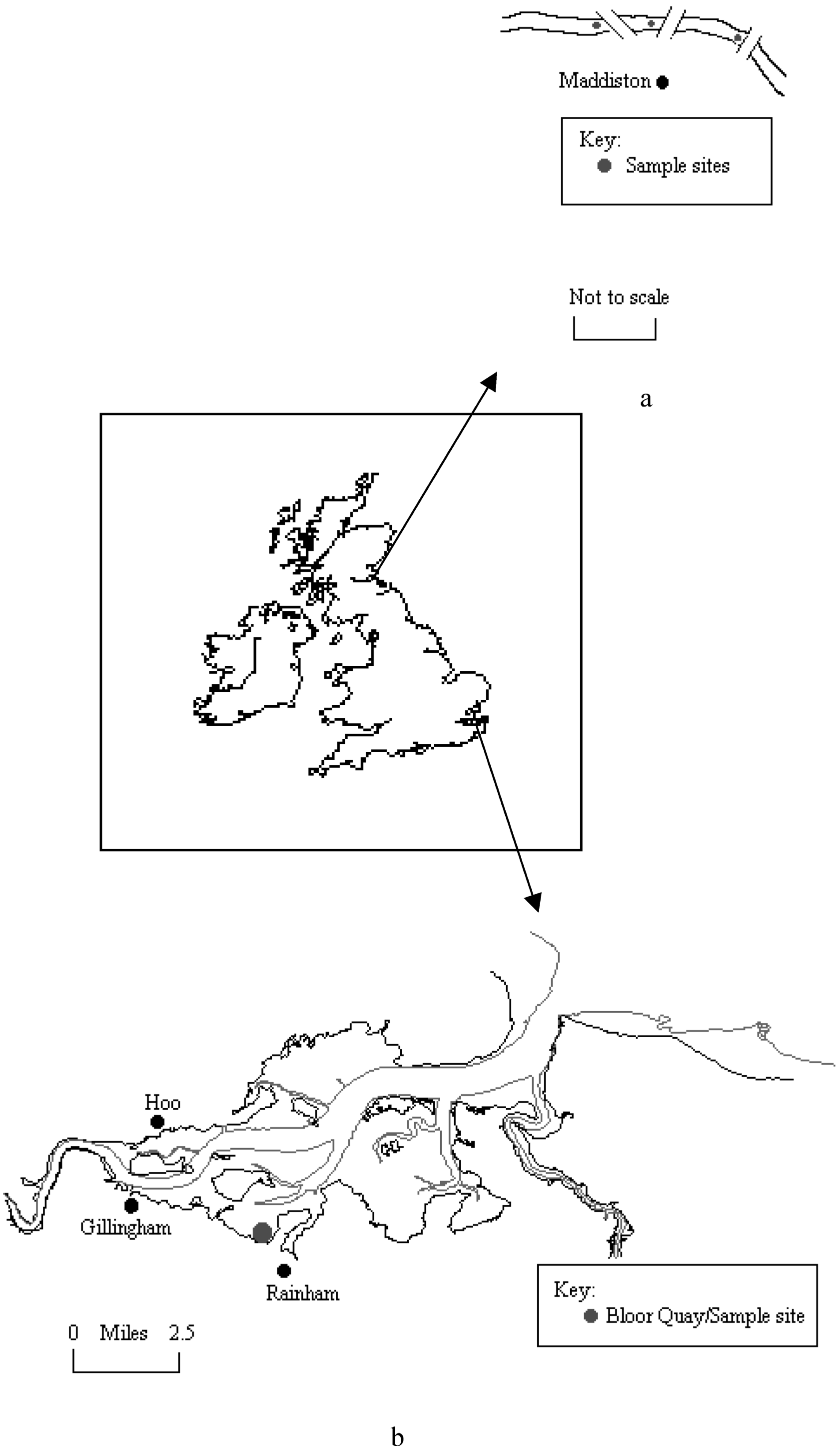


Figure 1. Sample sites: (a) Union canal (b) Medway estuary.

Composite Sample Preparation

Land and Water Services Ltd. supplied samples collected from three different locations along the contaminated stretch of the Union canal. These samples, which were devoid of stone particles or vegetable matter, were thoroughly mixed together to form a composite sample, which was stored in an airtight plastic container.

Two samples were collected from Bloor Quay but were sieved through a 10 mm sieve to remove stones, and mixed together to form a composite sample. Both were stored in a refrigerator at 4 degrees centigrade.

Sediment Digestion

Hatch and Ott (1968) describe the digestion procedure adopted in this study, where inorganic- or organic-Hg in sediment is broken down to free $Hg^{2+}_{(aq.)}$. Five replicate samples were digested along with two certified reference materials (digested in triplicate). Three blank samples were also included for digestion. The procedure used involved adding 5 ml concentrated H_2SO_4, 2 ml concentrated HNO_3 and 5 ml saturated $KMNO_4$ solution to approximately 0.2 g of a sample placed in a BOD bottle. The bottle was then covered with thick aluminum foil to prevent Hg leakage during digestion and was heated on a hot plate for 3 hours at 90 degree centigrade. Upon cooling to room temperature, 100 ml deionized distilled water was added followed by 12 ml of 10% hydroxylamine hydrochloride to reduce any excess $KMNO_4$.

Moisture Content Estimation

The moisture content of the composite and treated samples was analyzed using the procedure adopted in British Standard 1924: Part 2 (1990) to determine the water: solid (w/s) and water: binder (w/b) ratios, on a wet weight basis. Moisture contents were determined in triplicate for the treated samples and in five replications for the composite samples.

Sample Pre-Treatment

The sediment samples from Union canal and Medway estuary were pre-treated to reduce the moisture content prior to the addition of cement. Excess water from the Union canal sediment was first decanted and then removed by centrifugation at 2000 revolutions per minute for a period of 10 minutes.

The Medway estuary sediment, which was of a different consistency from the Union canal sediment, was treated by the addition of lime at 5% and 10% dosage and by a combination of 5% lime and 5% PFA, and then mixed thoroughly for 10 minutes in a 5 L Hobart mixer and allowed to stand.

Trial Mixes

The optimum mix designs for both the wastes were chosen from different trial mixes. However, the different consistency of the two wastes prompted an alternative remedial approach: the high moisture content of the canal sediment means that treatment by accelerated carbonation could not be performed and as such, activated carbon was used as an additive for the sorption of Hg. A blend of different hydraulic binders was identified as suitable for use with this sediment, as described below.

The lower moisture content of the estuarine sediment, however, allowed an accelerated carbonation step to be carried out. In this case, PC, SRPC and FSRPC performed best in the trial mixes.

Mix Designs

Figure 2 illustrates the treatment process of the two sediments. The Union canal sediment was mixed with different dosages (10, 20 and 30% weight/weight of entire waste) of PFA to

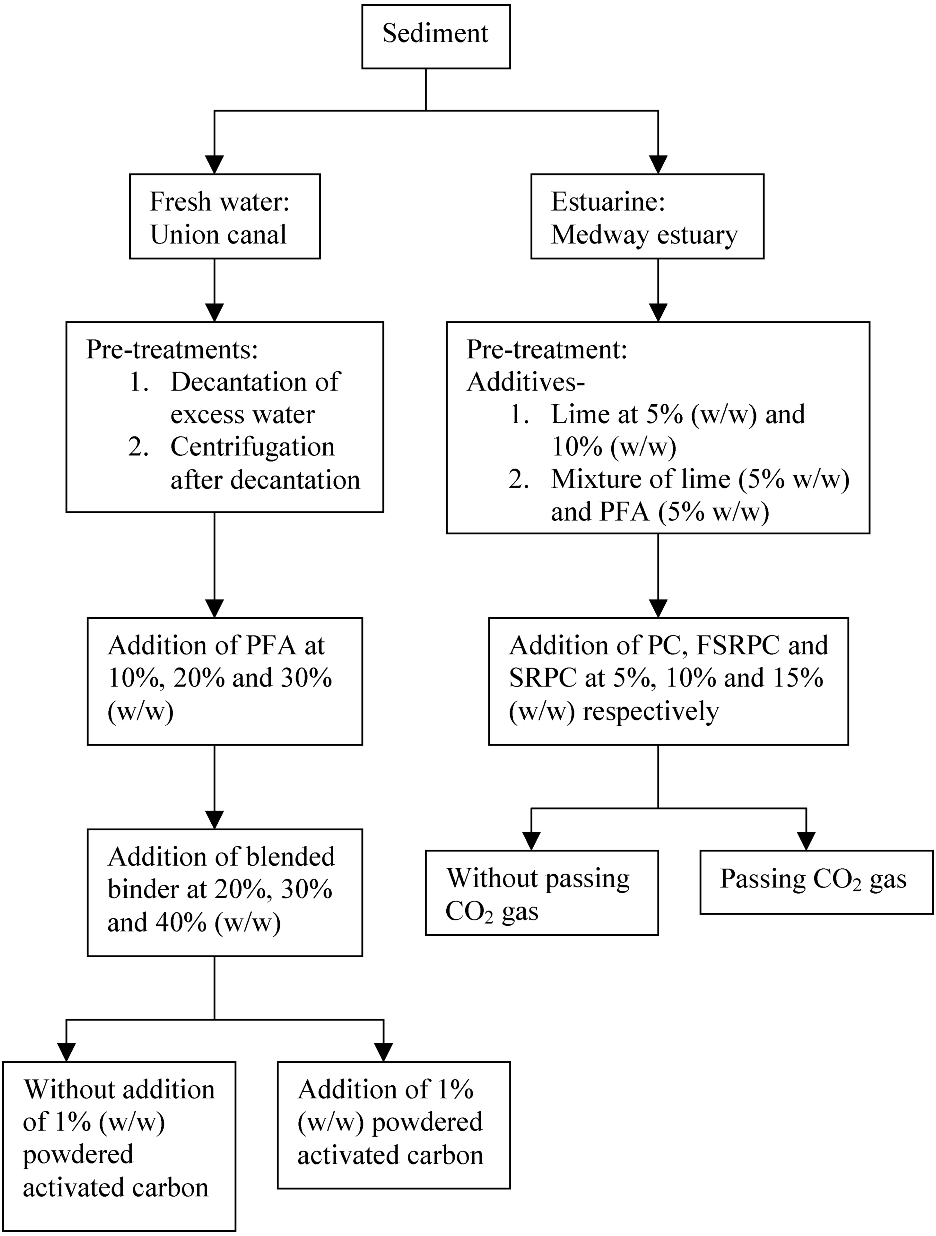


Figure 2. Flow diagram depicting sample treatment process.

Table 1
Optimum mixes for Union canal sediment

Additive % (w/w)					
PFA*	Activated carbon	Binder % (w/w)	Moisture %	W/B	W/S
10	—	20	49	1.64	0.96
20	—	20	41	1.03	0.70
30	—	20	41	0.82	0.70
10	—	30	46	1.15	0.85
20	—	30	40	0.80	0.67
30	—	30	39	0.65	0.64
10	—	40	45	0.90	0.82
20	—	40	38	0.64	0.61
30	—	40	37	0.53	0.59
10	1	20	45	1.45	0.82
20	1	20	40	0.98	0.67
30	1	20	29	0.57	0.41
10	1	30	44	1.07	0.79
20	1	30	38	0.75	0.61
30	1	30	29	0.48	0.41
10	1	40	43	0.84	0.75
20	1	40	36	0.59	0.56
30	1	40	28	0.39	0.39

W/B, water/binder ratio where binder is the sum total of cement and additive (s).
W/S, water/solids ratio.
*PFA % was calculated based on the moisture % after pre-treatment, i.e. 70%.

adjust the water/solid and then mixed with different dosages (20, 30 and 40% weight/weight of entire waste) of a blended binder comprising RF-A, PC and PFA (in the ratio's 1: 1: 0.25), together with powdered activated carbon at 1% w/w, based on wet weight. Samples without addition of activated carbon at 1% w/w were also prepared in similar manner. Samples were mixed in a 5 L Hobart mixer. A sample comprising untreated sediment was also prepared for investigation. The treated mixes of the Union canal sediment were hard and monolithic at 28th day. Table 1 records the optimum mixes used for this sample.

Following pre-treatment of the Medway estuary sediment, samples were prepared from SRPC, FSRPC and PC at 5%, 10% and 15% additions (weight/weight of entire waste), respectively, and then mixed for 40 minutes. To achieve accelerated carbonation, CO_2 gas was constantly passed through the sample during mixing, which was achieved by adapting the 5 L mixer to create in a closed mixing environment. Non-carbonated samples were prepared in a similar manner but without introducing CO_2 gas into the samples. Non-carbonated and carbonated control samples without the addition of additives and cements were also prepared. All the treated mixes of the estuarine sediment were hard and monolithic at the 28th day. The optimum mixes for this sample are given in Table 2.

Leaching Procedure

The leaching procedure adopted, DIN 38414-S4 (German standard, 1984), was carried out on samples of 28 days of age. Deionized, distilled water was used at a solid/liquid ratio

Table 2
Optimum mixes for Medway estuary sediment

Additive % (w/w)		Cement % (w/w)					
Lime	PFA	PC	FSRPC	SRPC	Moisture %	W/B	W/S
5	—	—	—	5	45	4.50	0.83
5	—	—	—	10	42	2.80	0.71
5	—	—	—	15	40	2.00	0.68
10	—	—	—	5	41	2.74	0.70
10	—	—	—	10	40	2.00	0.66
10	—	—	—	15	37	1.48	0.58
5	—	—	5	—	45	4.50	0.83
5	—	—	10	—	41	2.74	0.70
5	—	—	15	—	41	2.05	0.69
10	—	—	5	—	41	2.74	0.69
10	—	—	10	—	39	1.95	0.63
10	—	—	15	—	34	1.36	0.51
5	—	5	—	—	41	4.10	0.70
5	—	10	—	—	38	2.54	0.61
5	—	15	—	—	36	1.80	0.56
10	—	5	—	—	38	2.54	0.60
10	—	10	—	—	35	1.75	0.54
10	—	15	—	—	31	1.24	0.46
5	5	—	—	5	41	2.74	0.69
5	5	—	—	10	39	1.95	0.63
5	5	—	—	15	37	1.48	0.59
5	5	—	5	—	41	2.74	0.68
5	5	—	10	—	38	1.90	0.62
5	5	—	15	—	35	1.40	0.55
5	5	5	—	—	37	2.47	0.59
5	5	10	—	—	34	1.70	0.51
5	5	15	—	—	33	1.50	0.49

W/B, water/binder ratio where binder is the sum total of cement and additive (s).
W/S, water/solids ratio.

of 1:10. Samples were constantly rotated for 24 hours, and allowed to stand undisturbed, for 30 minutes (to allow settlement of suspended particles), before the sample was filtered using 0.45 μm cellulose acetate filter paper. The leachate was collected and stored in 1% (v/v) HNO_3 for analysis. Each mix was leached in triplicate. One blank and one reference PC were also leached per batch.

Analysis of Hg

The total Hg concentration in the digested sediment and the leachates obtained was analyzed by a dedicated mercury analyzer, CETAC M 6000A, based on the principle of Cold Vapour Atomic Absorption Spectrometry (Hatch and Ott, 1968).

CO_2 Uptake Analysis by Thermo-Gravimetric Method

The CO_2 uptake of the carbonated sediments was compared with the non-carbonated mixes using a thermo-gravimetric method. The amount of carbonation was measured and compared by calculating an index value *(the carbonation index value)*, from the formula given below, to calculate the degree of carbonation achieved during treatment:

$$\text{Carbonation index} = \%\ CaCO_{3[\text{carbonatedmixes}]} \text{ - } \%\ CaCO_{3[\text{non-carbonatedmixes}]}$$

Quality Guidelines for Mercury

In the UK at present, no sediment quality guidelines are available. As a result of this, we referred to the guidelines developed by the US National Oceanic and Atmospheric Administration (NOAA) (Buchman, 1999). The fresh water and estuarine sediment have been compared with their respective standards (Threshold Effects Level, TEL). TEL is equal to or below the value that has no toxic effect on the biota. The TEL for mercury in fresh water and marine sediment is 0.174 and 0.130 mg/Kg dry weight, respectively.

The different Environmental Quality Standards (EQS) for mercury, as mentioned in EC Dangerous Substances Directive, 76/464/EEC, are 1 μg/L (total Hg) for fresh water and 0.5 μg/L (dissolved Hg) for estuarine water (Environment Agency, 2005). The EQS's are concentration limits which, when exceeded, are believed to be detrimental to aquatic organisms. The leaching results from treated mixes are compared with the EQS's for fresh water and estuarine EQS's.

Results

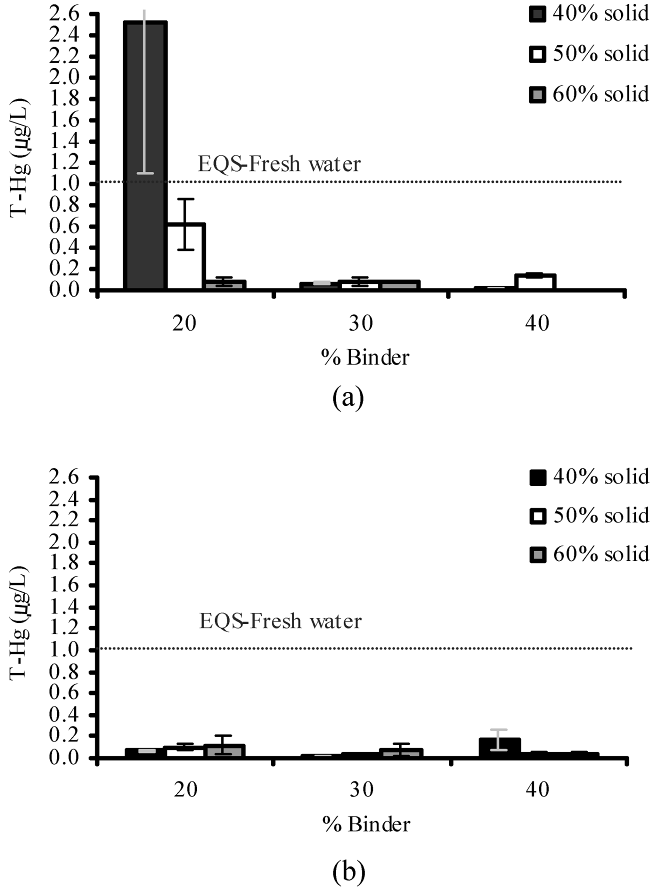
Sediment Characterisation

The total Hg concentration in the sediment from Union canal was 728 mg/Kg, which was significantly higher than TEL of fresh water sediment (0.174 mg/Kg dry weight). The sediment pH was 7.3. The sediment also had 81% water content, which after pre-treatment was reduced to 70%. Pore water analysis (total-Hg – 0.15 μg/L) indicated that Hg was tightly bound to the canal sediment.

The concentration of total Hg measured at 0.2 mg/Kg in the Medway sediment is higher than TEL for marine sediment (0.130 mg/ Kg dry weight). Moreover, Hg when ingested by aquatic organisms is bio-accumulated (Stein *et al.*, 1996), ultimately resulting in higher recorded Hg concentration in later years. The sediment was very wet with moisture content of 54% and the pH 7.6.

Conventional S/S Leaching Results

The leaching from the control (un-treated) sample is 0.001 μg/L, which may be due to Hg being tightly bound to the organic matter in the sediment. The pH of the leachates ranged from 10–11 for treatments with and without activated carbon addition, indicating that with respect to the latter complete carbonation might not be carried out. Mercury leaching was below the fresh water EQS (1 μg/L) for all the mixes without activated carbon addition, except 40% solid/20% binder mix (Figure 3a). Results indicated that an increase in the solid content of the cement-bound waste resulted in lower Hg leaching. Activated carbon addition during the solidification process reduced Hg leaching to below EQS (1 μg/L & Figure 3b).



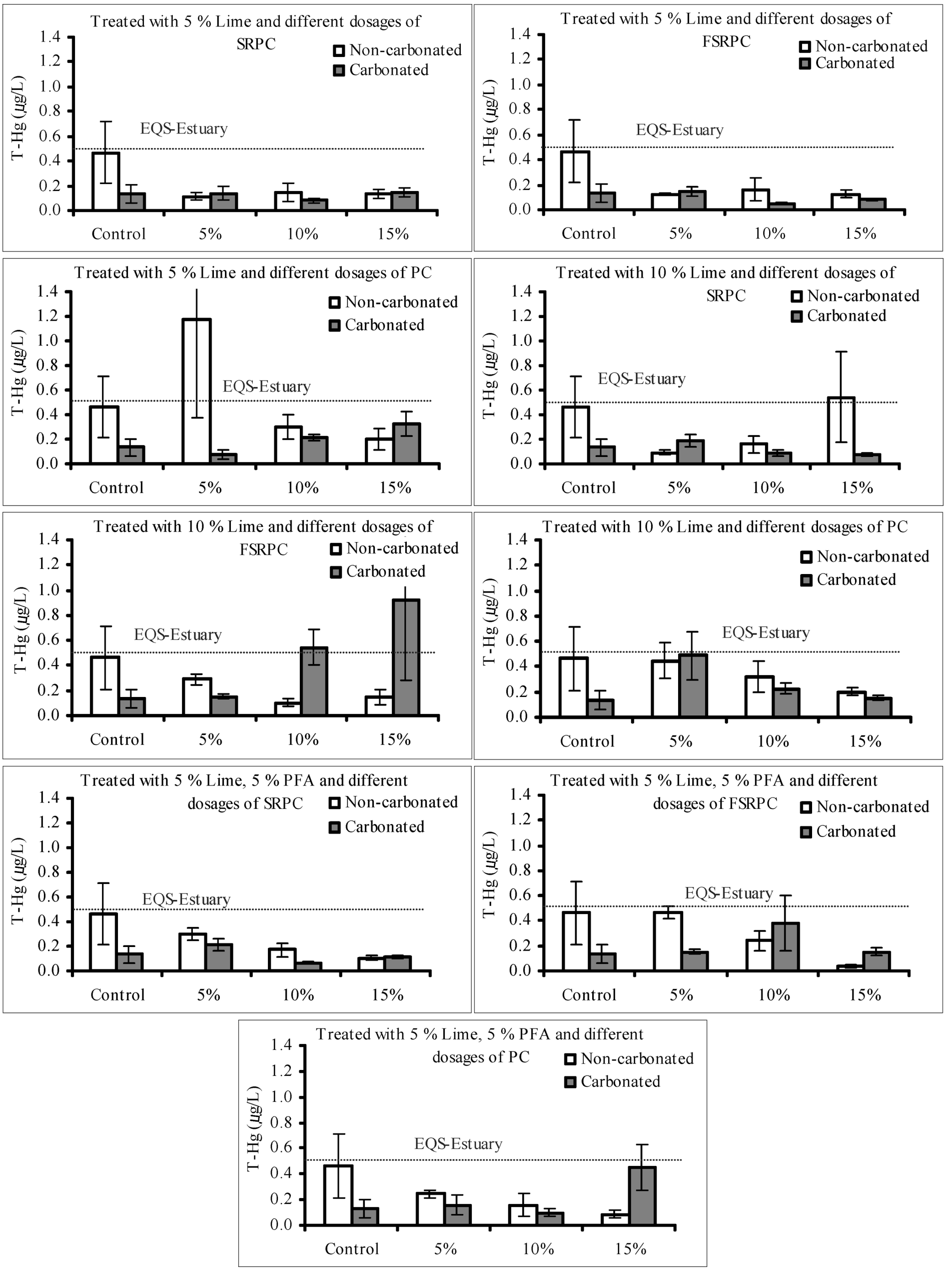
Error bar = ± Confidence interval based upon Estimated Standard Deviation at 95% confidence level

Figure 3. Leaching results for treatments (a) without activated carbon addition and (b) with activated carbon addition.

Leaching of Accelerated Carbonated Samples

Figure 4 gives the leaching results for the different additive(s) and cement dosages. It is evident that almost all the carbonated mixes have met the estuarine EQS (0.5 μg/L). When compared to their non-carbonated counterparts, the leaching of Hg was reduced by carbonation with few exceptions. These may be due to insufficient carbonation, as discussed later.

Carbonation mixes of Figure 5 give the carbonation indices and Hg leaching from different carbonated mixes, and show that irrespective of additive type there is a slight increase when the PC dosage is raise to 10%. But, when this binder dose is increased to 15%, the index either remains the same or decreases in value. A similar trend was observed for FSRPC with 10% lime or 10% lime/PFA. For SRPC, a similar trend was observed when the additive was 5% lime. However, when 10% lime and lime/PFA combinations were used, carbonation increased with the increasing cement dose. When different cement types are compared, the degree of carbonation mostly follows the order PC > FSRPC > SRPC. When

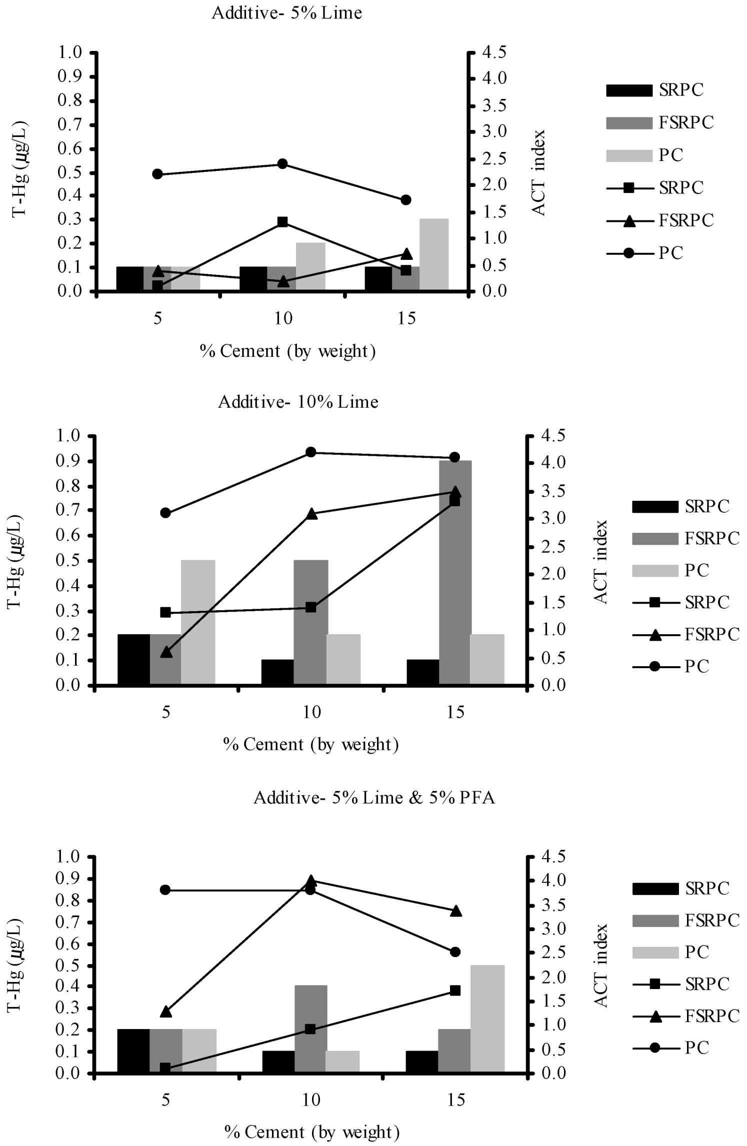


Error bar = ± Confidence interval based upon Estimated Standard Deviation at 95% confidence level

Figure 4. Leaching results for non-carbonated and accelerated carbonated samples.

lime (10% w/w) was partially replaced with PFA (5%), the carbonation index values were slightly lower than for lime only.

As indicated by Figure 5, Hg fixation may be related to the degree of carbonation for PC and SRPC. A higher carbonation index correlated with greater Hg fixation for these two cements, however, for FSRPC, the reverse was apparent.


Additive- 5% Lime

Additive- 10% Lime

Additive- 5% Lime & 5% PFA

Figure 5. Hg leaching versus carbonation index (columns indicating leaching data and lines indicating carbonation index).

Discussion

Conventional S/S

The immobilization of Hg is believed to be precipitation controlled and in a cement-based matrix Hg may be present as highly dispersed insoluble HgO (Cocke and Mollah, 1993). In this study the solids content of the cementitious matrix is seen to influence Hg immobilization in certain mixes (Figure 3), indicating that the porosity of the matrix is important to leaching. This observation is further supported by reduced Hg leaching when PFA is added to the mixture, increasing the total % solid content. Thus fixation of Hg may result from:

a) Minimization of water, as a reduced w/s ratio is exponentially related to the decrease in porosity (Roy and Scheetz, 1993), which helps in retaining Hg within the matrix.
b) As the Ca:Si ratio of the gel phase decreases from 1.7 to approximately 1.2 with the addition of PFA the sorption of cations onto C-S-H increases (Glasser, 1997), further binding the Hg tightly within the cement matrix during leaching.

As shown above (Figure 3), activated carbon addition during s/s favors Hg immobilization due to its adsorption (100% Hg adsorption in current study) on the carbon particles and subsequent encapsulation of Hg-rich carbon particles in the cement matrix. Zang and Bishop (2002) found 98.5% Hg adsorption for 20% (w/w) dose of reactivated powder activated carbon with their surrogate waste contaminated with 500 mg/Kg Mercury (II). The higher Hg adsorption % obtained in this study, higher than Zang and Bishop (2002), may be due to the difference in the adsorption capacity of the activated carbons used. Zang and Bishop (2002) used thermally reactivated previously used activated carbon, whereas the current study uses virgin activated carbon and this difference in their origins may cause a difference in their adsorption capacities. Again, adsorption capacity of activated carbon is inversely proportional to pH (Arafat *et al.*, 1999) and contradicts the higher immobilization shown by activated carbon-binder mixes in this study, where the pH is high due to cement addition. The contradiction supports Hebatpuria *et al.* (1999)'s explanation that adding cement and activated carbon to the waste as during this study does not affect the adsorption capacity of the carbon particles since adsorption reaction proceeds at a much faster rate than hydration reaction.

The cement-carbon-sediment interaction is explained by Figure 6, which illustrates that when cement and activated carbon are added to the sediment the carbon particles rapidly adsorb mercury onto its surface. In the meantime, the pore-water in the sediment initiates hydration and calcium ions are leached from the cement grains to produce mainly $Ca(OH)_2$, leaving behind silica-rich hydration products (Arafat *et al.*, 1999). This $Ca(OH)_2$ again reacts with silicic acid on the cement grains to produce semi-permeable Calcium-Silicate-Hydrate (C-S-H) gel around the cement granules, which gradually extends to connect other granules entrapping Hg-rich carbon particles. Part of the pore-water interacts with activated carbon itself in a mechanism described by Wassay *et al.* (1995) when investigating granular activated carbon with acidic potassium iodide to remediate Hg-polluted soil. According to

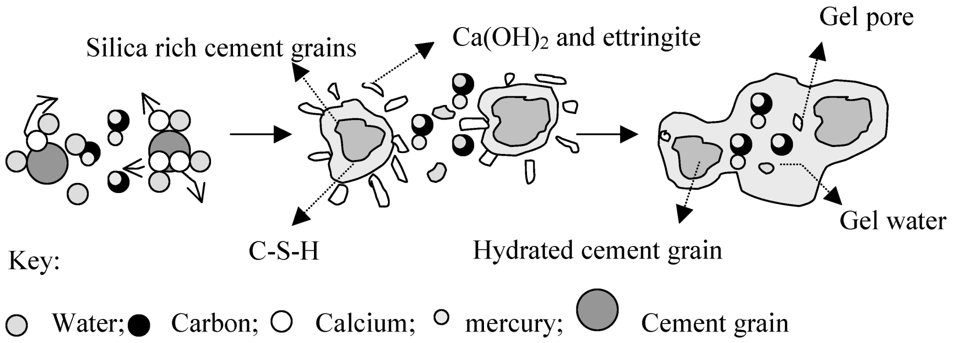


Figure 6. Schematic representation of cement-activated carbon-sediment interaction.

their explanation at the activated carbon-water interface the following reactions occur:

$$C_xO + H_2O = C_x^{2+} + 2OH^-.$$
$$C_xO_2 + H_2O = C_xO^{2+} + 2OH^-$$

Sediment pore-water reacts with activated carbon to produce C_x^{2+} and C_xO^{2+}. These ions in turn react with the Hg-complexes of the sediment, making them immobile.

Accelerated Carbonation

The amount of cement used seems to influence the degree of carbonation for PC and FSRPC by impeding carbonation when the dose was increased from 10% to 15%. This effect may be the consequence of a reduction in CO_2 permeability. An experimental study by Lange *et al.* (1996) found that higher waste contents resulted in increased CO_2 permeability. However, in contrast to the other 2 binders, increased SRPC addition resulted in increased CO_2 uptake.

The difference in the relative amount of carbonation of the three cement types may be related to composition and fineness and the following order was established (PC > FSRPC > SRPC). In this context, Lange *et al.* (1996) suggested that there was a relationship between the ferrite content of cement and the amount of $CaCO_3$formed. However, in this work the major difference between FSRPC and SRPC was likely to be the surface area of the cement as FSRPC was the most finely ground of the three cements.

From Figure 5 it is evident that higher lime dosage has an increased effect on the degree of carbonation. This may be the result of higher amounts of portlandite [$Ca(OH)_2$] formation as CaO hydrates on exposure to the water present in the sediment. The solubility of CO_2 in water at a particular temperature and partial pressure is approximately proportional to $[OH^-]^2$ present in solution (Dow and Glasser, 2003). The following equations explain the chemical interactions of CO_2 with pore water:

$$CO_2(g) \rightarrow CO_2(aq.) + H_2CO_3 \tag{1}$$

$$H_2CO_3 \text{ (weak acid)} \rightarrow HCO_3^- + H^+ \tag{2}$$

$$HCO_3^- + OH^- \rightarrow CO_3^{2-} + H_2O \text{ (Dow and Glasser, 2003)} \tag{3}$$

Thus, an increased lime dose increases the portlandite concentration, which in turn increases the OH^- concentration of the solution to such an extent that rapid dissociation of bicarbonate ions occurs in accordance with equation (3). This relationship between CO_2 solubility and OH^- concentration magnifies the CO_2 absorption capacity of pore water in excess alkalinity, increasing the carbonation reaction rate of according to the stoichiometry given below:

Carbonation of portlandite:

$$Ca(OH)_2 + CO_2 \text{ (aq.)} \longrightarrow CaCO_3 + H_2O \tag{4}$$

The results suggest that a partial replacement of lime with PFA would reduce the amount of $Ca(OH)_2$ present.

The relationship between Hg fixation and degree of carbonation clearly indicates that, in these experiments, the choice of binder influences the degree of carbonation effected, and that those binders that carbonated to a greater degree were accompanied with greater level of Hg fixation. Lange *et al.* (1996) discussed the carbonation of PC and SRPC and showed that the mass transport properties of carbonated cement-based waste forms were significantly affected. This resulted from product densification, reduced pore diameter/surface area, and total pore volume as a result of $CaCO_3$ precipitation. With respect to Hg leaching, a modified pore structure would result in enhanced Hg retention in the solid phase. It is interesting to note, however, that in hydrated systems, carbonation may be associated with an increase in mean pore diameter (in micro-porous regions), making the resultant matrix more porous and permeable (Gervais *et al.*, 2004).

Conclusions

Several conclusions can be drawn from this study-

- Both hydrated and carbonated mixes were capable of meeting the threshold values chosen for Hg leaching.
- For conventional s/s, an increase in solid % (with PFA) favors Hg immobilization, and this probably results from pore refinement and formation of a Si-rich C-S-H gel.
- Addition of activated carbon during s/s significantly reduces Hg leaching.
- Accelerated carbonated mixtures containing 15% w/w PC and FSRPC were carbonated less than 10% cement containing mixtures.
- The degree of carbonation effected increased with the addition of Lime (10%)-SRPC and Lime-PFA-SRPC combinations.
- The relative degree of carbonation for the different cement types was in the order PC > FSRPC > SRPC, and this may be due to composition/surface area.
- Increased doses of lime resulted in higher CO_2 uptake and this was probably due to increased availability of portlandite for carbonation.
- The partial replacement of lime with PFA resulted in lower CO_2 reactivity.
- Mixtures that were carbonated to a higher degree were found to have lower levels of Hg leaching,
- The reason why mercury leaching in carbonated finely ground SRPC was increased is not known.

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