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Development of a Waterjet System for Direct Delivery of Granular Iron and Activated Carbon to Remediate Contaminated Aqueous Sediments

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While the techniques and technologies associated with contaminated sediment remediation are relatively mature, there are several issues associated with these practices that make them unattractive. The inability of currently used mechanical mixing implements to place amendments in aqueous environments and their intrusive behavior toward benthic communities are just two examples of a necessity for an improved delivery method. Waterjets may be a viable option for placement of particulate remediation amendments, such as activated carbon and granular iron, at depth. A custom waterjet nozzle and injection system has been fabricated by the authors to examine this delivery concept. The developed injection system's performance was tested by characterizing the waterjet-delivered amendment (activated carbon and granular iron) distributions in a surrogate sediment. The delivered amendment distributions followed similar patterns for a range of injection times and a variety of amendments. The injection depths, however, were dependent upon the type of amendment being injected. These findings have led to a better understanding of what occurs during an amendment injection, which can be used for a more controlled placement of remediation amendments using this technique in the future. The laboratory results indicate that the subject waterjet system may have the potential for field-scale applications, especially for granular iron delivery, as the authors were able to place between 60 and 70 wt percent into a surrogate sediment bed along the path of injection. © 2011 Wiley Periodicals, Inc.

INTRODUCTION

Contaminated sediments have become a widespread issue. The US Environmental Protection Agency (EPA) estimates that approximately 10 percent of the sediment underlying surface waters in the United States are sufficiently contaminated to cause health issues for benthic communities, their food chains, and human health (US EPA, 1998). Significant research has been performed and is ongoing in order to determine the possible means for mitigating the risks that contaminated sediments pose to human health and the environment.

The technique and technologies used for sediment remediation are relatively mature. There are several different treatment technologies that have been studied and that are

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considered for remediation of contaminated sediments. The foremost alternatives are capping, dredging, and chemical/physical treatment. Wang et al. (1991), Murphy et al. (2006), and Reible et al. (2003) researched *in situ* capping with various materials, such as sand and reactive core mats, which proved to reduce the flux of contaminants from the sediment to the water column. While the *in situ* capping technique reduces the flux of contaminants into the water column, the contaminants have the potential to be released back into the environment if the cap is disturbed (Reible et al., 2003). Capping has the potential to change the topography of the bottom of a water body, which could cause problems for navigable waterways.

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Dredging is the removal of contaminated sediment from the ecosystem. Sanchez et al. (2002) cited results of an extensive General Electric Company (GEC) study on dredged sites. GEC's study illustrated that many times dredging did not effectively reduce surface sediment contaminant concentrations and, in many instances, led to resuspension of contaminants into the water.

Contaminated sediment remediation performed *in situ* is typically attempted using the third alternative, chemical/physical treatment. A common chemical treatment technique, known as reductive dechlorination, uses zero-valent iron (ZVI) to reduce chlorinated compounds to an eventual less harmful product. The ZVI reductive dechlorination approach has been well recognized and established in groundwater remediation research by Reynolds et al. (1990), Gillham and O'Hannesin (1994), and others. It is important to note that the reductive decholorination process typically takes place through the use of granular iron rather than ZVI.

Granular iron is simply a ZVI core surrounded by oxide layers. For this study, granular iron was used, as the iron used in this study was exposed to both oxygen and water, creating the oxide outer layers.

A common physical means to perform contaminated sediment remediation is the use of powdered activated carbon (PAC) to adsorb the contaminants, thereby reducing the aqueous contaminant concentration and, in turn, its bioavailability. The attraction between contaminants and PAC particles is also a very well studied process by Jonker and Koelmans (2002), Zimmerman et al. (2004, 2005), and Ghosh et al. (2001), among others. The method has been proven to reduce the negative impacts of contaminants on benthic organisms by reducing the amount of contaminants they uptake. Zimmerman et al. (2004) demonstrated the effectiveness of this type of treatment by achieving 87 percent reductions in the aqueous equilibrium polychlorinated biphenyl (PCB) concentrations treated with 3.4 weight percent activated carbon (AC). McLeod et al. (2007) observed reductions in the uptake of PCBs by 84 percent in the *Macoma balthica* clam for AC-treated areas.

The previously described chemical/physical techniques are effective means to remediate certain contaminants, but the problem with these methods lies in the ability to place the necessary amendments within the targeted sediment zone. Currently, amendments are commonly delivered through mixing. Cho et al. (2007, 2009) both indicated that a rotovator direct mixing method can lead to reductions in the bioaccumulation of contaminants in benthic organisms and aqueous contaminant concentrations. The direct mixing technique is normally used in tidal mudflats at low tide conditions; this prevents the treatment of areas that are continuously submerged. Mixing implements, such as the frequently used rotovator, are certain to cause a high mortality

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rate in the benthic organisms inhabiting the treatment area as well as the resuspension of sediment into the water column.

The quantity of amendment that can be successfully added to the contaminated sediment by any delivery method is a variable that should be considered when choosing a treatment delivery system. Cho et al. (2009) successfully achieved 2.0 to 3.2 percent by weight concentrations of PAC in sediment through the use of direct mixing, which resulted in approximately 90 percent reductions in the aqueous equilibrium PCB concentrations 18 months after amendment delivery. The investigators targeted placement of the PAC in the uppermost 30 cm of sediment because they considered this to be the biologically active layer. Laboratory research by Rysavy et al. (2005) illustrated that at the proper ZVI dosage, the lag time for the dechlorination of some PCB congers can be reduced by up to 100 days in contaminated sediments. The amount of amendments delivered to the contaminated sediment is important, as the interaction between amendment and contaminant will control the remedial effectiveness (Zimmerman et al., 2004).

These and other works in the literature have demonstrated that PAC and granular iron or ZVI are effective at remediating contaminated subsurface media. However, the delivery of these amendments to contaminated sediments at significant depths and without having damaging impacts to the benthic communities remains the significant drawback of mechanically mixing the amendments into the sediment.

Waterjets have been used in a variety of industrial applications in a variety of mining, cleaning, and machining applications. Possibly one of the most innovative developments to waterjet usage is the addition of abrasives to the waterjet stream, which allows for a wider range of products to be cut with the jet as well as enhanced waterjet cleaning. These abrasive waterjet cutting/cleaning systems function by combining the force of a high-pressure water stream with the bombardment of the abrasive on a material to be cleaned or cut. These operations are typically performed at pressures from 2,000 to 58,000 psi (Summers, 1995). Waterjets have the potential to deliver amendments (in the place of abrasives) to contaminated sediments. Unlike rotovators, waterjets also have the potential to be used in both tidal mudflats and subaqueous environments at a lesser impact to the benthic community. This work describes laboratory experiments to apply PAC and granular iron to a saturated sediment using a waterjet. The work focuses on the quantification of the spatial distribution and concentration of the delivered amendments. Colorimetric and analytical techniques were used to quantify the carbon and iron distribution in the sediment.

Possibly one of the most innovative developments to waterjet usage is the addition of abrasives to the waterjet stream, which allows for a wider range of products to be cut with the jet as well as enhanced waterjet cleaning.

MATERIALS

Amendments

The type of granular iron used for the testing process was obtained from Quebec Metal Powders (QMP), Ltd. of Quebec, Canada. The variety of iron selected was QMP's ATOMET 86, which was fine enough to allow 73 percent of the powder to pass through a 325 mesh sieve. The relatively small grain size oxidizes rather rapidly and is readily accommodated by the waterjet system.

Calgon Carbon Corporation's (Pittsburgh, Pennsylvania, USA) PAC, known as WPH, was also used as an experimental amendment. The WPH PAC is an extremely fine carbon powder, and approximately 90 percent of the powder would pass through a 325 mesh sieve.

Surrogate Sediment

A surrogate sediment was used for the injection-system performance tests. The surrogate chosen was kaolin clay acquired from Unimin Corporation (New Canaan, Connecticut, USA). Kaolin was chosen for its cohesive nature, which is thought to be exhibited by many sediments dominated by a clay fraction, and for its uniform white color, which allowed for the quantification of the amendments that exhibit contrasting colors. The kaolin sediment was mixed using a concrete mixer using 45.4 kg of the kaolin powder with 31.2 liters of water. This ratio was based on a visual consistency comparison between a collected sample of lake sediment and the kaolin/water mixture.

By changing the diameter and/or shape of the collimating nozzle's exiting orifice, the flow's velocity and dispersion could also be changed, which would allow for more controlled placements.

EXPERIMENTAL SECTION

Nozzle Development

The notion of injecting remediation amendments into contaminated sediment has been previously researched by Cantrell et al. (1997) and Cable et al. (2005). Both of these research groups attempted to develop a delivery method that would inject particulate amendments as a slurry into the sediment. The slurries were composed of a type of polymer (for example, guar gum), water, and the amendment. The polymer's purpose was to retain the amendments in suspension, allowing them to be pumped into the exiting high-pressure water stream.

It was discovered during this research process that the ability to keep dense particles like granular iron in suspension was difficult. Continuous mixing of the slurry was required, and even then pump clogging and pulsating flow was exhibited during testing. The inability to provide a uniform discharge led to the development/consideration of a new delivery system that would avoid these issues.

An amendment delivery system that used a pressurized air and water stream was then investigated. The system would function through the development of a specialized nozzle. The type of nozzle being considered has been previously used in abrasive waterjet cleaning and cutting (Summers, 1995), and the design of these nozzles was used as a departure point for this design process. The nozzle was machined from a solid piece of aluminum and incorporated a fully adjustable mixing chamber to reduce amendment-plugging problems. An interchangeable collimating nozzle was integrated into the design so that different collimating nozzles could be created and used for injecting a wide range of particle sizes. By changing the diameter and/or shape of the collimating nozzle's exiting orifice, the flow's velocity and dispersion could also be changed, which would allow for more controlled placements. There were two different types of water-injection nozzles fabricated for testing. A concave, cone-shaped nozzle and a cylindrical-shaped nozzle were both tested. The concave nozzle led to plugging issues within the mixing chamber; therefore, the cylindrical nozzle was deemed the superior option and was used throughout the testing process. Exhibit 1 illustrates the configuration of the nozzle's different components.

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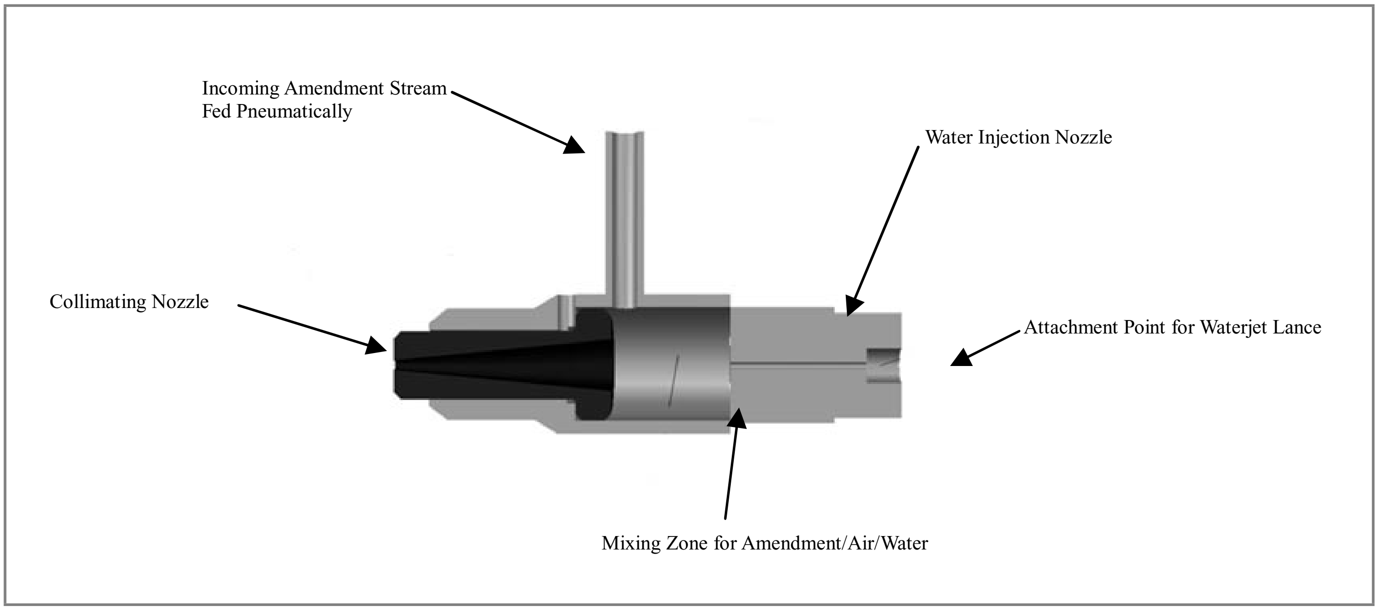

Exhibit 1. Schematic of amendment injection nozzle

Injection System

Exhibit 2 shows the four main components of the experimental waterjet system. First, commercially available pressure washers were used as the waterjet platform because these units were capable of injections at lower pressures and flow rates. A gasoline-powered unit (Troy-Bilt [Cleveland, Ohio, USA)]; Model#020344) was used for tests at pressures from 700 to 1500 pounds per square inch (psi), while an electric unit (Task Force [Wilkesboro, North Carolina, USA]; Model#TF1600) was used for pressures below 500 psi. Traditional abrasive waterjet cleaning and cutting methods are performed at higher pressures ranging from 2,000 to 58,000 psi, while minimizing the solids used. For this research, the low pressures and flow rates are targeted to prevent unnecessary water input, while maximizing the amendment delivery. A commercial pneumatic sandblasting tank was used for this amendment delivery to the mixing chamber. Third, a standard

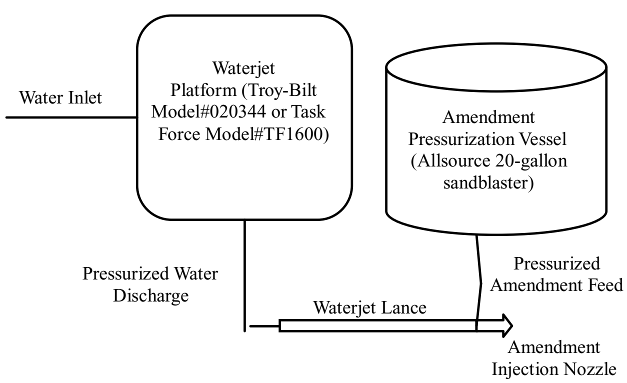

Exhibit 2. Schematic of the developed injection system

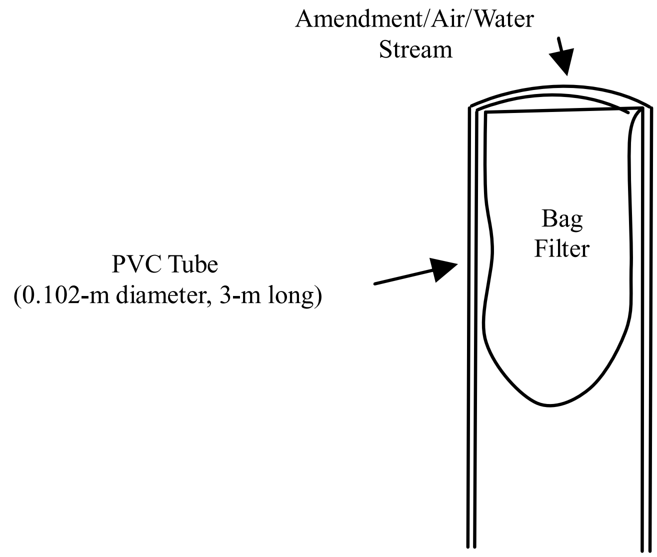

Exhibit 3. Setup for concentration tests

pressure washer lance and trigger assembly were used, with the addition of a pressure gauge. Finally, the fourth component of the system was the nozzle itself.

Performance Testing

Concentration tests were run to characterize the water and air pressure settings that would allow the highest percentage of amendment (both iron and PAC) to be mixed into the collimating stream of amendment/air/water. The test objective was to determine the optimal pressure to prevent the injection of excess water into the contaminated environment and to predict the length of injection time to reach the target amendment concentrations. The test nozzle was calibrated so that the maximum amount of amendment flow in the discharge could be achieved. The chamber volume was set with 8.89 cm of setback distance from the collimating nozzle exit, allowing the maximum flow to be achieved by the incoming amendment feed line. The incoming pneumatic amendment pressure was set to approximately 60 psi, and once set this pressure was maintained for the duration of the tests. The pressure of the waterjet entering the mixing chamber was chosen to be the variable in these tests, as this parameter was thought to be the controlling factor for the depth of injection, the concentration of amendment found in the exiting stream, and the effects on the benthic communities.

The testing apparatus (Exhibit 3) was created from a 3 m long, 10.2-cm diameter polyvinyl chloride (PVC) pipe that was capped on one end. The uncapped end of the PVC pipe was then fitted with a 15-cm diameter, one-micron bag filter (Midstates [Pacific, Missouri, USA] high-strength, one-micron, polyester,double-chain-stitched bag filter) so that the bag filter was held open and in place at the open end of the pipe. The pressurized amendment/air/water stream was then injected through the bag filter and down into the PVC pipe. The amendment was captured in the filter, and the water was collected within the PVC pipe. The mass of both the amendment and water leaving the collimating nozzle

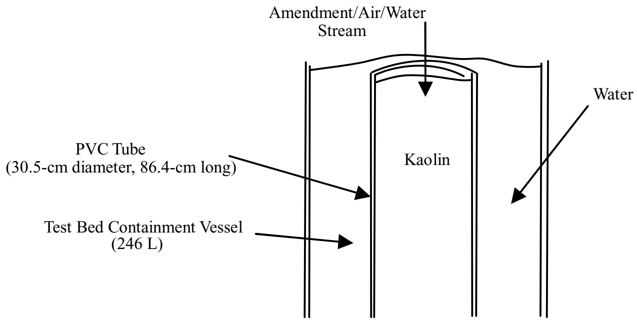

Exhibit 4. Setup for test-bed injections

could then be determined. The process was timed so that a flow rate could be calculated. These tests provided a departure point for the next series of injection tests.

Test-Bed Injections

The injection system was next tested on the surrogate sediment to characterize its ability to deliver amendments to a target depth and concentration. Targets were set for both the depth and concentration reached during each injection. The targets were based on results published by Zimmerman et al. (2004) and Cho et al. (2007, 2009). The depth target was 30 cm, and the concentration target was 3.4 dry weight percent of PAC throughout the test bed of surrogate sediment. Granular iron is typically used in permeable reactive barriers for groundwater remediation; therefore, it was difficult to attain proven target values for sediment remediation. For this reason, the targets for the granular iron injections were set at the same values as the PAC injections. The targets were used as a departure point for the laboratory testing, as in the field conditions would typically dictate the amount of necessary amendment to be delivered and the location of delivery.

The test beds (Exhibit 4) were 30.5-cm diameter PVC tubes, 86.4 cm long, which were filled with kaolin and capped with a vented 30.5-cm PVC cap. Once filled and capped, the PVC tube was then submerged in a 246-liter water vessel to simulate working in a subaqueous environment. The depth from the water surface to the kaolin surface was measured both before and following each injection to determine if material loss had occurred. Once the system was calibrated, a stream of amendment/air/water was injected into the column. The injection was performed with the discharging nozzle at the kaolin surface, and each injection was timed. Following the injection, the PVC tube containing the kaolin column was removed from the water-filled vessel and then dried using electro osmosis. Once dried, the kaolin column was removed from the PVC tube and then sliced horizontally at 2.54-cm intervals down the column. Five samples were taken from each of these slices, so that each sample could be analyzed for the concentration of amendment delivered. The samples were taken from each slice with the assumption that the concentration would be uniformly distributed around the point of injection. Therefore, samples were taken from the point of injection along the radius to the outer edge of each slice. The symmetrical distribution assumption was checked periodically by taking samples on the radius directly opposite. A blank sample was also collected from each of the kaolin-filled tubes to develop a baseline. Pictures were taken of each slice immediately before removing the samples.

Powdered Activated Carbon Concentration Distribution Analysis

PAC concentrations were measured using a FieldSpec Pro model spectroradiometer from Analytical Spectral Devices, Inc. (Boulder, Colorado, USA). The spectroradiometer measures the reflectance of light from a sample. The reflectance is measured by the spectroradiometer in terms of a reflectance factor and is plotted against the wavelength emitted from the light source. The spectroradiometer was calibrated and set up according to the user manual. The outputted reflectance factor versus wavelength plots were compared to prepared standards of known dry weight percents of PAC. The spectroradiometer was able to differentiate between PAC concentrations differences as small as 0.1 percent by dry weight.

Granular Iron Concentration Distribution Analysis

The granular iron used in the testing exhibited a light gray color that did not contrast enough with the white kaolin sediment to allow the spectroradiometer to accurately characterize the concentration of granular iron delivered. Therefore, the iron-containing samples were analyzed using two different methods. First, each slice was photographed immediately after being removed from the testing column. These photographs were visually compared to a color chart to characterize the relative difference in iron concentrations, as shown in Exhibit 5.

Samples were also collected from the slices at various intervals and sent to ACME Analytical Laboratories (ACME Labs) in Vancouver, Canada, where the samples were dried, digested, and run through ICP emission spectrometry to determine the percent

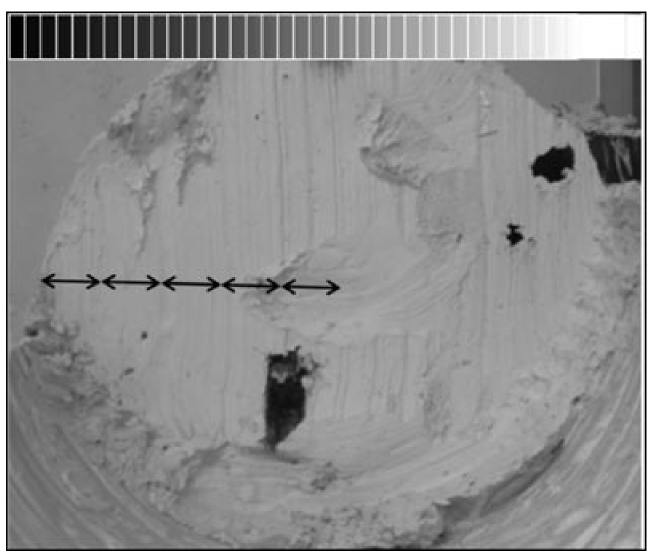

Exhibit 5. Visual comparison using developed color chart to predict distributions

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Exhibit 6. Discharge concentration results (tests performed at pressure vessel at 60–70 psi)

Amendment	Pressure of Waterjet (psi)	% by Weight of Amendment in Discharge	Volume of Amendment in Discharge (mL)
PAC	200	0.500	48.1
PAC	300	0.200	12.5
PAC	700	0.200	15.9
PAC	1,000	0.100	8.9
Granular Iron	700	33.0	162
Granular Iron	1,000	54.8	397
Granular Iron	1,500	46.8	290
Granular Iron	1,500	46.5	311

weight of iron delivered to each column sample. The laboratory data were used to quantify the relative values used for the colorimetric analysis.

RESULTS AND DISCUSSION

Discharge Concentration Testing

The injection system was initially tested to determine the maximum concentration of amendment that could be mixed into the stream exiting the nozzle. These tests were performed for both the PAC and the granular iron, and the results are displayed in Exhibit 6.

Differences were observed between the data collected for the two different amendments because a significantly higher volume of granular iron could be mixed into the waterjet stream. The difference appeared to be due to a drawback of the pressure vessel. The vessel was designed to deliver abrasives such as sand, which had a density closer to granular iron than to the lighter, less dense PAC particles. The data gathered during this experiment did demonstrate the effectiveness of the system to mix granular iron into the discharge stream, as concentrations from 33–54 percent by weight of granular iron in the discharge were achieved. The collected data also provided a departure point for the injection tests, as the time required to achieve the target amendment concentration within a sediment test bed could be calculated based on the flow rate of amendment leaving the nozzle.

PAC Sediment Injections

The durations of PAC injections required to attain the target concentration were calculated using the amendment flow rates found in the discharge concentration testing and the volume of the kaolin within each column. The calculated durations were on the order of hours because of the low weight delivery of PAC in the waterjet stream. It was not realistic to perform injections over a single location for extended time periods, so the mass delivery calculations were confirmed by conducting PAC injections over time

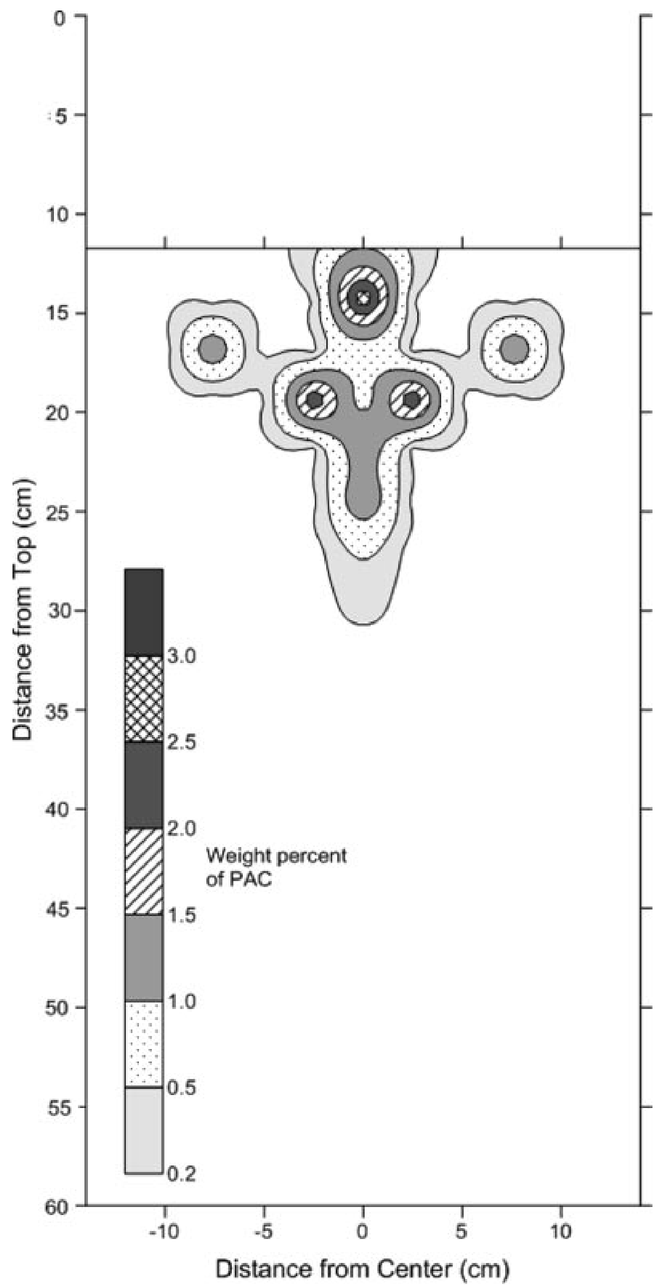

Exhibit 7. PAC percent by weight distribution for a 5-minute injection

periods of 5 and 10 minutes at injection pressures of 1,500 psi. The results from the 5-minute injection are shown in Exhibit 7. These relatively long injection durations resulted in the liquefaction and excavation of approximately 7.6 cm of clay during the 5-minute injection and 15.2 cm of clay during the 10-minute injection. The results of spectroradiometry analysis for the PAC injections illustrated the effect that this excavation had on the system's ability to place amendments. The 5-minute injected column contained a vein of PAC ranging in concentration from approximately 0.5 to 3.0 percent dry weight down through the first 20 cm of kaolin, as shown in Exhibit 7. The concentration of PAC below 20 cm was insignificant. The 10-minute injected column only contained two sample locations with PAC concentrations exceeding 0.1 percent throughout its depth. Significant excavation of sediment was observed after the 6-minute point during the injection. The removal of this uppermost 15 cm of sediment would most likely have caused the removal of the amendment delivery zone.

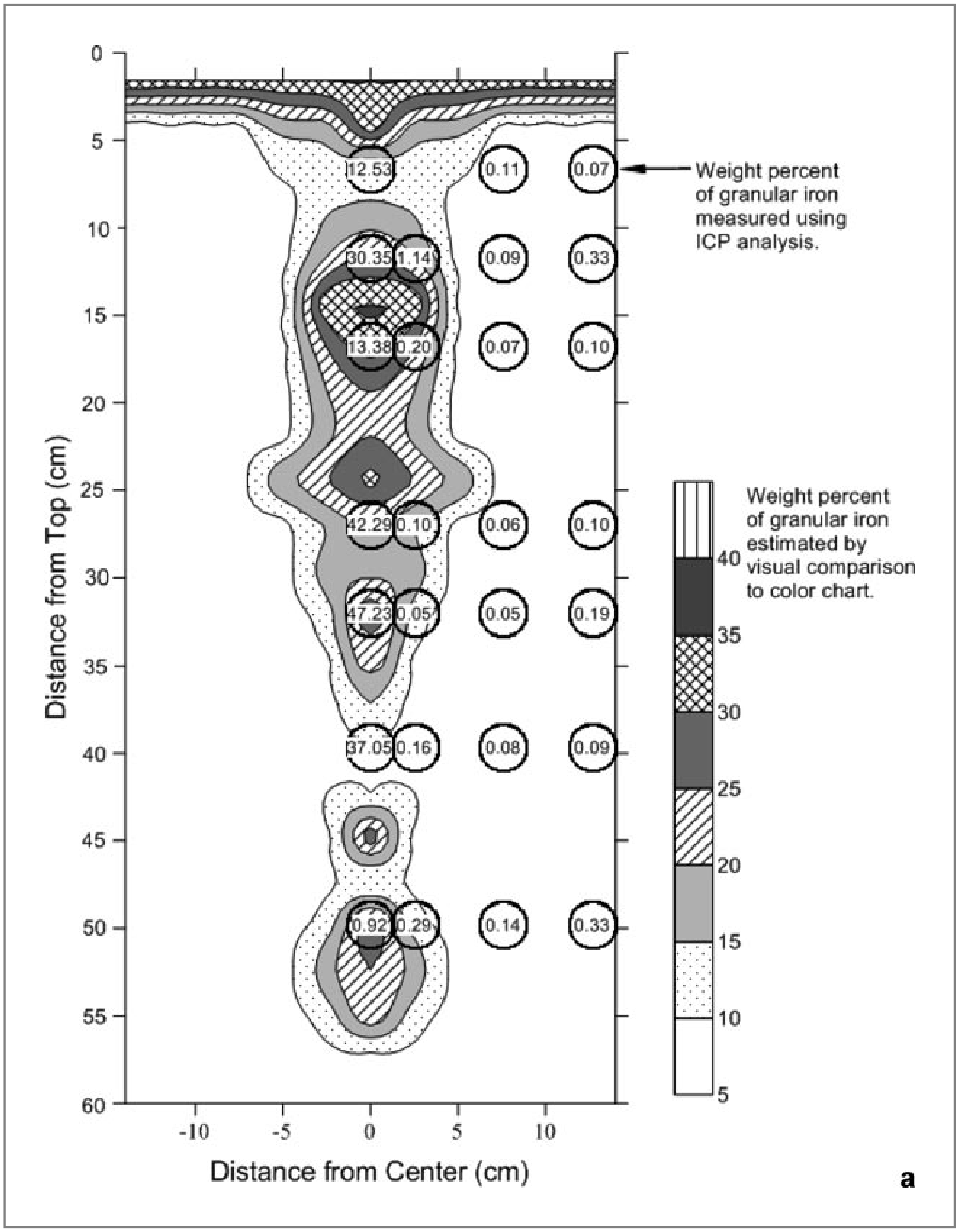

Exhibit 8. (a) Granular iron distribution for a 0.5-minute injection. (b) Granular iron distribution for a 1-minute injection. (c) Granular iron distribution for a 3-minute injection

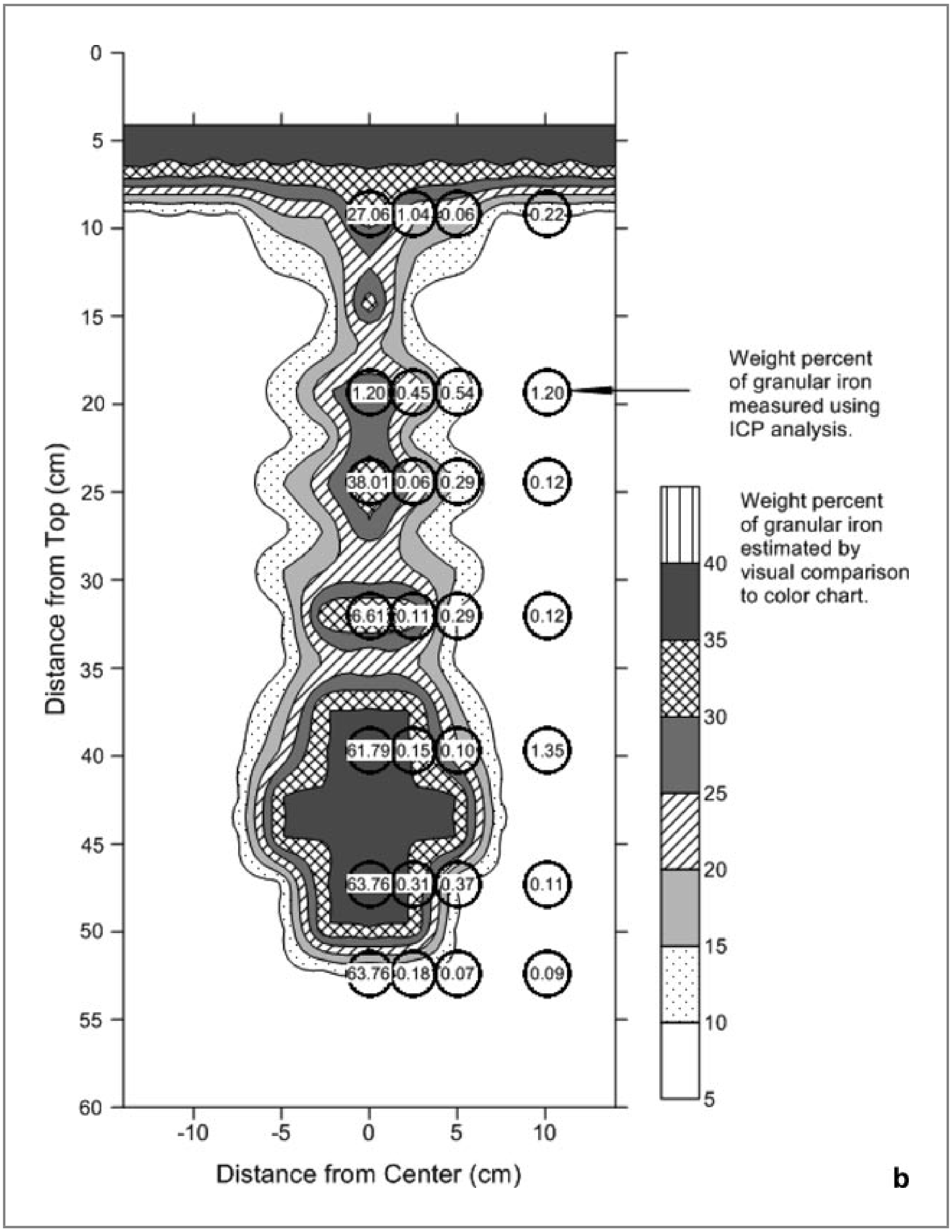

Exhibit 8. Continued

Granular Iron Injections

The distribution patterns for the three granular iron injections are depicted in Exhibits 8a through 8c. The time of injection to reach the targets was calculated using the discharge concentration testing results provided in Exhibit 6 and proven to be 0.5 to 1 minute. Therefore, tests were performed at 0.5 minute, 1 minute, and 3 minutes at an injection pressure of 1,500 psi. The visual comparison distribution is presented by different

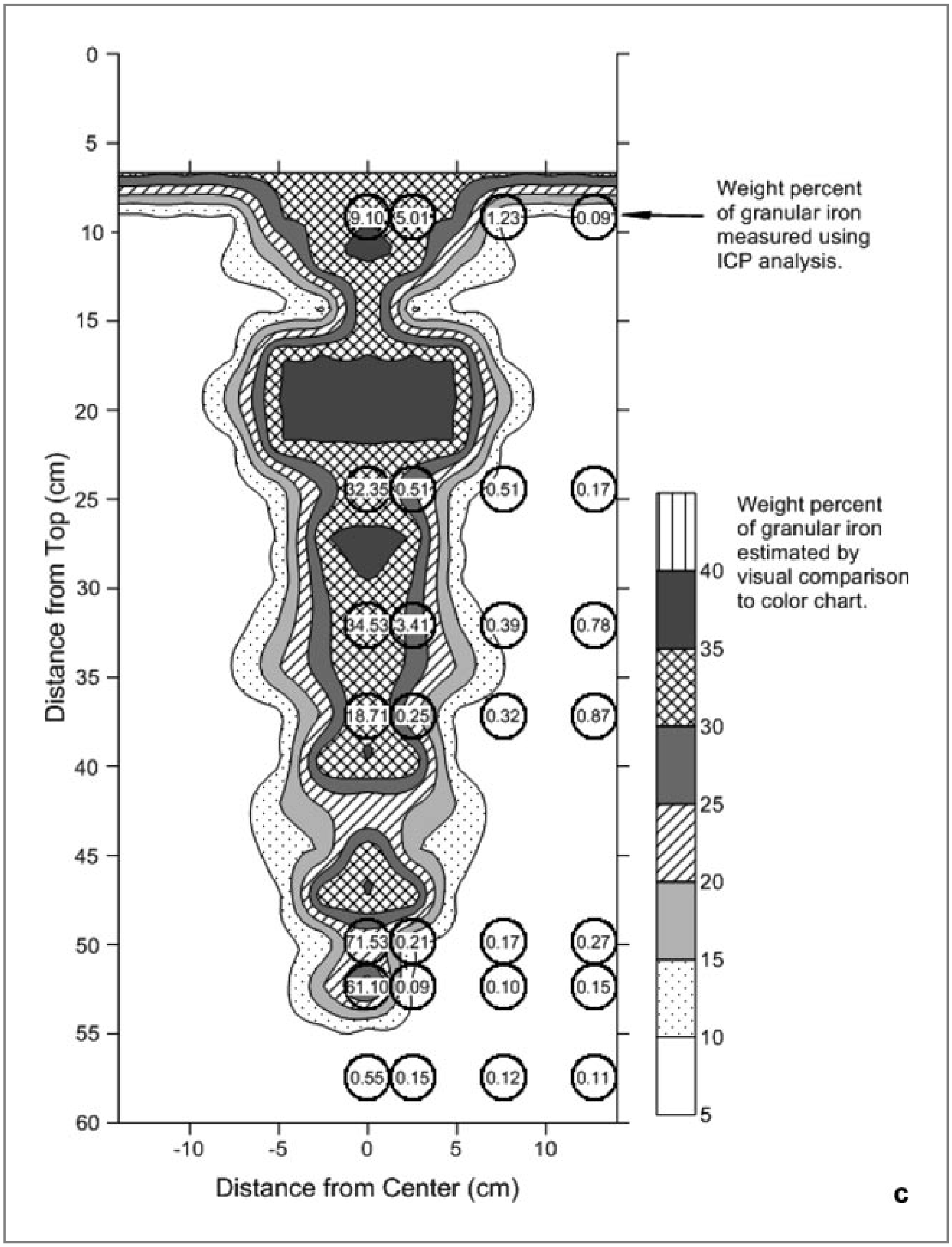

Exhibit 8. Continued

patterns, and the scale presents the different concentrations present in each column. The circles indicate the sample locations that were analyzed by inductive coupled plasma (ICP) analysis and the results for each of these locations are provided within each circle. There are similarities between the semiquantitative and visual comparison analysis, which allows for a comparison between the two as a means to estimate the granular iron concentrations placed within the kaolin column. The depth of amendment placed for these injections was

2.5 times deeper than those experienced during the PAC injections. The granular iron was able to reach depths of approximately 51 cm in each of the injections. The concentration of delivered granular iron exceeded the target of 3.4 percent by dry weight. Concentrations were as high as 60 to 70 percent by weight in some samples taken along the path of injection. However, the distribution was not uniform through the length of the column, as the vein of injected iron took on more of an hourglass shape. The symmetrical assumption samples that were analyzed indicated that in many instances the distribution was not uniform around the line of injection.

The results of the laboratory testing showed that the subject waterjet system could deliver amendments to saturated sediments to varying degrees of success depending on the density of the particular amendment.

Data Modeling

Groundwater model calibration equations were used to compare the ICP data to the visual comparison data. The coefficient of residual mass (CRM) equation is given by Spitz and Moreno (1996) as:

$$\mathrm{CRM} = \frac{\sum_{i=1}^{n} O\,i - \sum_{i=1}^{n} P\,i}{\sum_{i=1}^{n} O\,i}$$

where O is the observed value (ICP data), P is the predicted value (visual comparison data), and n is the number of data sets compared. Spitz and Moreno (1996) state that the closer the CRM value is to zero, the more accurate the predicted values are to the observed. The CRM was 0.44, using the visual comparison concentration range shown in Exhibit 5.

CONCLUSIONS

The results of the laboratory testing showed that the subject waterjet system could deliver amendments to saturated sediments to varying degrees of success depending on the density of the particular amendment. Target depths and amendment concentrations could be achieved with the granular iron, but the experimental system did not deliver a sufficient volume of PAC necessary to achieve the corresponding target concentrations.

Second, and possibly the most significant finding from this study, the distribution of the injections seemed to follow a distinctive pattern. The pattern could be characterized as being larger-diameter, high-concentration amendment pockets connected by smaller-diameter, lower-concentration veins of amendment. This distribution pattern was witnessed for both the granular iron and PAC injections. The pattern indicates the repetition of a cycle of energy buildup and dissipation. The jet of amendment/water/air initially contains a significant amount of energy in the form of momentum, but as it travels down into the sediment bed, the energy begins to dissipate until it reaches a point where downward motion ceases. At this point, lateral motion begins and a pocket of amendment begins to form. As the pocket grows, so does the pressure, which eventually leads to enough energy buildup, and downward motion resumes. The process appears to occur downward through the kaolin column until eventually either there is no longer a sufficient amount of energy for propagating the injection or the time for energy buildup was not attained.

Third, it was discovered that the granular iron was capable of reaching greater depths within the sediment bed. This most likely occurred because of the differences in mass and

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resulting momentum between the two types of amendment. Momentum is described as the product of the mass and the velocity, and the system was set up so that the velocity during each of the injections would be nearly equal. Therefore, differences in amendment momentum would be solely dependent upon the mass/amount of the amendment present in the discharging stream. As a significantly higher volume of the higher-density granular iron was placed in the injection stream, it can be inferred that these injections would have a much higher momentum than that created by a PAC injection. This would account for the deeper penetration of the granular iron injections.

Finally, there were some less positive observations made during the course of the project. Limitations were discovered for the volume of PAC that could be delivered from the system. A significant volume of granular iron could be mixed into the stream exiting the injection nozzle (up to 54 weight percent), but only very small volumes of PAC could be achieved (0.5 weight percent). This is due to differences in the density of these amendments because the granular iron is nearly eight times denser than the PAC. Increasing the duration of the injection to increase the mass of PAC delivered to the sediment resulted in the unwanted excavation of the sediment.

The overall conclusion to this work is that it was demonstrated that a waterjet system could deliver granular iron at depth in a saturated surrogate sediment. The results of this work imply that a series of pulsed injections could potentially deliver a prescribed concentration of granular iron over a defined area of contaminated sediment.

ACKNOWLEDGMENTS

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