





University of Mississippi

FLOOD MITIGATION ENGINEERING RESOURCE CENTER (FMERC) - PROJECT EC14-005

<u>Final Report Appendix D- Pollution Prevention Analysis during Sandy's</u> <u>Hurricane and Flooding</u>

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SUMMARY

Flooding or flooding surge after extreme hurricanes often brings heavy land contamination near coastal regions. Typical contaminants include spilled oil, persistent organic pollutants (POPs), pesticides that contain endocrine disrupting compounds (EDCs), heavy metals, microbial pathogens or other invasive and infectious disease-causing species. To rapidly eliminate these contaminants from soil or ground water (even sediment), one needs to develop and utilize in situ remediation technologies. One of the existing and also popular technologies is nanoremediation (or nano-enhanced remediation), which relies on the use of state-of-art nanotechnology. Specifically, highly reactive metallic nanomaterials are applied to the contaminated areas (topsoils, subsurface soils, groundwater, or sediments) to provide strong reducing or oxidizing power, high available surface area for adsorption, or antimicrobial activity that increase the removal efficiency of organic compounds, immobilize heavy metals, and deactivate pathogenic microorganisms. For this 6-month feasibility study, NJIT will evaluate the potential of active metallic nanomaterials toward the removal of spilled oil, POPs, heavy metals, microorganisms. pathogenic Based on the research. NJIT will and suggest environmentally benign (not harming ecosystems themselves), low cost, and high efficient nanomaterials for remediation purposes. Ultimately, NJIT will present a comprehensive feasibility report on in situ nanoremediation to address the multiple contamination issues for the coastal regions that are impacted by the aftermath of hurricane. Moreover, this project will also establish a unique connection between academic groups with public, industrial partners, and government agencies.

BACKGROUND

Extreme disasters such as hurricanes result in massive coastal flooding and other serious coastal impacts such as shoreline erosion and water pollution that affect man-made infrastructure and coastal ecosystems.(Titus & Anderson, 2009) One of the important subsequent ecological and economic risks for low-lying coastal zones is the contamination of coastal waters by industrial and agricultural chemicals released by accidental spills during severe storm events. Moreover, with a malfunctioning wastewater drainage system during flooding events, sewer waste from coastal residential areas may also release considerable amounts of pollutants and increase the spread of microbial pathogens and the chances of infectious disease. As recently as 2012,

Hurricane Sandy devastated the New Jersey shore, resulting in around 7,700 gallons (29,146 liters) of fuel spilled from Phillips 66's Bayway Refinery in Linden, New Jersey and 300,000 gallons (1,150,000 liters) of diesel fuel spill from a northern New Jersey oil facility. Furthermore, fertilizer runoff, dumping of hazardous materials into the oceans, waste discharge, fishing, mining, and drilling were all shown to negatively affect the coastal regions. As shown in Fig. 1, coastal areas are vulnerable to increases in the intensity and frequency of storm surge and heavy pollution. The predicted increase in frequency and severity of coastal flooding makes the



Fig. 1. Coastal flooding provides a mechanism for potentially harmful chemicals on impacted land.

potential for severe contamination events very high unless proper precautions are implemented.

Along with oil, other chemicals also have the capacity to damage complex costal ecosystems. For instance, POPs are far more persistent than those found in oil and accumulate in the aquatic food chain, causing long-term effects.(Huggett & Bender, 1980) POPs include several classes of pesticides and industrial chemicals (e.g., PCBs, chlordanes, DDTs) that can bioaccumulate to relatively high concentrations in top-level predators (e.g., fish and marine mammals) through trophic transfer. POPs may enter, deposit and accumulate in the coastal region through the flooding surge that brings POPs from EPA Superfund clean-up sites, industrial discharge sites, and agricultural runoff. Other environmental hazards such as cleaning solutions, paints and pesticides may also have been released from the destruction of residences and commercial facilities. The large quantity of construction and demolition waste generated from destroyed residences is also of concern. This debris includes asbestos-containing materials such as roof shingles, siding and flooring, electronic waste such as computers and televisions that contain heavy metals. The disposal of storm debris created by Sandy's destructive force and by the resulting flooding and cleanup presents environmental challenges.

Microbial pathogens, invasive species, infectious bacteria are the causative agents of critical human health. Hospital-acquired infections caused by microbial contamination are not only the leading cause of increased mortality and morbidity among patients from hurricane-impacted areas, but are also contributing to the long term sanitation issues. Elevated concentrations of fecal coliforms typically follow periods of heavy rainfall in urbanized areas along the land–sea interface, which have been documented in estuarine systems following hurricane events. (Mallin et al., 2000; Paerl et al., 1998) Bacterial inputs to coastal waters can originate from the increased use of septic tanks in the surrounding urban watersheds. It is also found that 10⁶ fecal coliforms/g can be expected from animal feces; thus, animal manure represents a sizable potential fecal bacterial load to receiving waters. Suburban streets, driveways, and lawns have been shown to be major source areas of fecal coliform bacteria in stormwater runoff. (Young & Thackston, 1999) It is important to characterize these microbial contaminations along the coastal region and evaluate the alternative technologies for the *in situ* removal to alleviate the health risks.

WHY NANOREMEDIATION?

Nanotechnology promises a potential revolution in approaches to soil and groundwater remediation.(Müller & Nowack, 2010) The nanotechnology market for soil and groundwater remediation is expected to grow to around \$23.6 billion worldwide.(Mueller & Nowack, 2010) Nanomaterials are significantly more reactive than larger particles because of their much greater surface area per unit of mass, and this effect is made use of in environmental remediation. Zero valent iron (ZVI) has been used in reactive barriers at numerous sites all over the world for the removal of organic and inorganic contaminants.(Bernd, 2008; Karn et al., 2009) The reaction rates of nanoscale zero-valent iron (nZVI) are 25–30 times faster than the reaction rates of granular iron in the micrometre to millimetre range, and the sorption capacity is also much higher.(Li et al., 2006) Table 1 gives examples of the current use of nanoparticles in remediation.

Process mechanisms	Process mechanisms Nanomaterials		
Photocatalysis	TiO ₂ Organic pollutants		
Adsorption	Iron oxides, dendrimers	Metals, organic compounds, arsenic	
Redox reactions	nZVI, nanoscale calcium peroxide	Halogenated organic compounds, metals, nitrate, arsenate, oil	
	Silver	Bacteria, pathogens	

Table 1. Examples of the use of nanoparticles in remediation

nZVI remediation traditionally is applied via a permeable reactive barrier (PRB), which is effective in the reductive degradation of a wide range of halogenated solvents, such as chlorinated methanes, brominated methanes, trihalomethanes, chlorinated ethenes, chlorinated benzenes, other polychlorinated hydrocarbons, and pesticides and dyes.(Chang et al., 2007; Chang et al., 2005; Zhang, 2003) This PRB technology involves placement of reactive media in one or more soil trenches located directly in the path of contaminated ground-water flow, thereby providing direct contact between contaminants and the reducing agent. An alternate ZVI-based remediation technology employs reactive media in the form of slurry. The slurry is injected directly into the subsurface through one or more injection wells strategically placed within the target treatment zone. In the United States, the US Navy and NASA have used nZVI in remediation for a number of years and have had positive experiences.(Mueller & Nowack, 2010; Müller & Nowack, 2010)

Adding nanoscale metal oxides as nano-adsorbents to polluted soils will effectively immobilize toxic heavy metals.(Schorr, 2007) A mixture of iron and iron oxide has also been shown to be even more effective than higher-cost products such as activated alumina – while being active for even longer periods.(Schorr, 2007) Green rust – a very reactive iron oxide – can be used to reduce Cr(VI) to Cr(III), which is not soluble and much less toxic than the mutagenic Cr(VI). Iron oxide minerals can be used to adsorb heavy metals. Carbon-based nanomaterials, such as dendrimers and polymers, are also currently being explored for the removal of metals and organics from soils and groundwaters.(Mueller & Nowack, 2010)

Nanoscale calcium peroxide (nCaO₂) has recently been used for the clean-up of oil spills.(Karn et al., 2009) Several projects have been conducted in New Jersey, USA. Two American companies are using nano-sized calcium peroxide as an oxidant in the remediation of soils containing various organic contaminants, such as gasoline, heating oil, methyl tertiary butyl ether (MTBE), ethylene glycol and solvents. Despite the advances of nanotechnology in site remediation, little work is being carried out on topsoil remediation and coastal hazard mitigation.

Metal oxide nanoparticles (NPs) are known to possess strong antimicrobial properties. Inorganic metal oxides are being increasingly used for antimicrobial applications. The main advantages of using inorganic oxides when compared with organic antimicrobial agents are their stability, robustness, and long shelf life. The considerable antimicrobial activities of inorganic metal oxide NPs such as ZnO, MgO, TiO₂, SiO₂ and their selective toxicity to biological systems suggest their potential application in therapeutics, diagnostics, surgical devices and nanomedicine based

antimicrobial agents.(Adams et al., 2006; Reddy et al., 2007; Sawai, 2003) The advantages of using these inorganic oxides nanoparticles as antimicrobial agents are their greater effectiveness on resistant strains of microbial pathogens. A higher concentration of smaller particles with a higher surface area gives a better antibacterial behavior,(Sawai et al., 1996) whereas crystalline structure and particle shape also influences the antimicrobial activity. Fig. 2 demonstrated the broad-spectrum antimicrobial efficiency of CaO NPs and highlighted the potential of CaO NPs as promising antibacterial agents at cheap costs.



PROJECT DELIVERABLES

In this feasibility study, it aims at answering a series of questions on whether applying active nanomaterials at appropriate doses on contaminated topsoils after the impact of hurricanes would potentially elevate the degradation of POPs and oil-spill-related organic pollutants, immobilization and detoxification of heavy metals, and reduce the spread of microbial pathogen. In line with this aim, the following the research tasks are proposed:

- (1) Investigate topsoil contamination and characteristics (e.g., concentration, types, and distribution) in the Hackensack areas/Meadowlands.
- (2) Evaluate the efficacy of pollutant removal by using different types of nZVI, nCaO₂, nCaO, or α -Fe₂O₃ on typical contaminants of topsoils.
- (3) Conduct comprehensive feasibility research on the removal efficiency of organic compounds, free soluble metals or indicator microorganisms (e.g., fecal *E. coli*) by a number of typical commercialized nanomaterials through the three major process mechanisms as summarized in Table 1; the report will also discuss the influencing factors such as the mixing of topsoils and nanomaterials, soil moisture, pH and salinity on the organic pollution degradation and removal.

POTENTIAL SITES FOR NANO-REMEDIATION

Superstorm Sandy inundated hundreds of homes in Moonachie and Little Ferry with dirty floodwater and caused one of the largest sewage spills in North Jersey history. Fig. 3 shows the geological distribution of potential toxic sites near the Hackensack, Little Ferry, and Moonachie areas. In a heavily developed region dotted with toxic sites–2,835 in Bergen County and 1,394 in Passaic County –flooding tends to bring a barrage of pollution within close proximity to residents. Superfund sites–the worst toxic sites in the United States present the biggest because they contain deadlier and more concentrated pollution.

Specifically, the following contamination sites are highlighted here as examples that are worth further investigation and protective measures for future possible pollution spread during flooding.

Site 1: Superfund site in Kearny (Diamond Head Oil, New Jersey, EPA ID#: NJD092226000), which is in the New Jersey Meadowlands and is next to the Hackensack River, is contaminated with a number of hazardous chemicals including polychlorinated biphenyls (PCBs), dioxin, and asbestos.

Site 2: The Standard Chlorine Chemical Company Superfund Site (Standard Chlorine Chem Co. Inc. New Jersey EPA ID#: NJD002175057) is a 25-acre site located in the town of Kearny, New Jersey, on an industrialized peninsula along the Hackensack River releases dioxins, benzenes, naphthalene, PCBs and other semi-volatile or volatile compounds into the Hackensack River and adjacent wetlands. (http://www.epa.gov/superfund/sites/npl/nar1672.htm)



Superfund sites

The federal Superfund program was launched 30 years ago as a way to accelerate the cleanup of the nation's worst toxic sites, but only three of the sites in Bergen and Passaic counties have been fully cleaned. Eight have been on the list for more than 25 years.

🛡 Bergen and Passaic county sites

New Jersey DEP sites

The state Department of Environmental Protection oversees about 2,800 contaminated sites in Bergen and Passaic counties. They range from leaking residential oil tanks to old industrial complexes where factories dumped pollution unchecked for decades.

More than 100 sites

Less than 50 sites

Special sites

Other projects or locations in North Jersey that have an impact on the environment

Other toxic sites

Fig. 3. Distribution of superfund sites and other potential toxic sites near or around Hackensack areas.

Site 3: Kearny freshwater marsh site (Fig. 4): Elevated concentrations of arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), and mercury (Hg) that exceed the Ontario Aquatic Sediment Quality Lower Effect Levels (a commonly used Sediment Quality Guidelines for the Protection of Aquatic Life established by the Ministry of Ontario, Canada) were found in the sediment at this site.



Fig. 4 Map of the sites in the Meadowlands District.(Tsipoura et al., 2011)

Site 4: Trace level organic chemical contaminants in the sediment of Hackensack River. For instance, organic contaminants such as TCDDs and total DDTs may reach up to 0.02 μ g/g.(Onwueme & Feng, 2006) Perturbation of sediment during a hurricane and flooding may facilitate the resuspension and spread of these toxic chemicals onto the inland areas of Hackensack.

SIMULATION OF CONTAMINANT MIGRATION DURING SANDY'S FLOODING

This study applied CCHE2D-Chem, an integrated model developed in the NCCHE, to simulate pollutant migration profiles under two hypothetical scenarios. The contaminants are assumed to be released in the site of the Kearny freshwater marsh (Fig. 4 and Fig. 5) when the storm surge waters of Hurricane Sandy inundate this area. In the two cases, one is assumed with a high flux rate and the other with a slow flux rate. The flow fields computed by CCHE2D-Coast are used as input flow conditions for simulation of contaminant migration. The simulation conditions and results are briefly discussed below.



Fig. 5. The contaminant source location in the numerical mesh.

To demonstrate the potential heavy metal migration from marshland to flood water, the pollutant source location in the numerical grid is shown in Fig. 5. The mobility of heavy metals are affected by many factors including the chemical forms and properties, soil organic content, the quantity and type of soil binding sites, soil ion strength, pH, temperature, the concentration of complexing anions (organic and inorganic), and competing cations in soil solution. All these factors are site specific and need extensive investigations to find out their respective impacts. An approximation was used to estimate the range of the migration rate of arsenic (As) from the marshland to the floodwater. There were studies (Solomon et al. 1990, Smedley and Kinniburgh 2002, USEPA 2005, Dubey et al. 2007, Zhu et al. 2011) showing Arsenic leaching from CCAtreated woody debris to the floodwater and sediments after Hurricane Katrina. Measured As concentration in sediment right after Hurricane Katrina ranged from 1-100 ppm (µg/kg). For that specific case, the leaching rate was estimated at 0.1 mg/s from soil to the overlying floodwater during the inundation period. In our case, we assume that the As contaminant is assumed to leach through flood waters due to rising of surge tide in Hurricane Sandy in 2012. Two scenarios of the leaching were simulated: one with high leaching rate of 0.1 mg/s (Scenario 1) and the other with slow leaching rate of 0.001 mg/s (Scenario 2). For this short period of inundation, chemical decay is assumed negligible.



Fig. 6. Arsenic concentration distribution in water at Day 17 of migration.

The simulations were calculated using CCHE2D-Chem model (Zhu, et al. 2012, Jia et al. 2013). It is a two-dimensional depth-averaged model to simulate chemical fate and transport with the input of simulated flow fields. Based on mass balance and fate processes, the model solves the transport equations of contaminant in water, within bed sediments and the interaction between water and sediments through the adsorption partition. Users can specify single or multiple pollution sources/sinks. The general fate processes include volatilization, photolysis, hydrolysis, and biodegradation. For the simulation of contaminant release, the flow fields of the surge tide were already computed and stored into a dataset. The computation of the leaching started at 0000 UTC, 10/29/2012. It continued for three days, till 0000 UTC, 11/1/2012. Time step for simulation of leaching was 10 sec. After three-day leaching during the period of inundation by

Sandy, the As concentration distributions in water at 0000 UTC 11/1/2012 are shown in Fig. 6 (Scenario 1) and (Scenario 2). It can be seen that the As distribution patterns are similar for both scenarios. Scenario 1 with the high leaching rate resulted in a peak concentration level of 0.22 ppb compared with 0.0022 ppb in Scenario 2. There is no violation of the drinking water quality standard for As of 10 ppb (10 μ g/L), which is based on assumed leaching rates. The distribution patterns are determined by the hydraulic conditions. The area near the marsh was flooded and then quite isolated from the other flooded area. As a result, the leached As remained in the isolated area after inundation for both scenarios.

CASE STUDIES OF NANOMATERIALS FOR SITE REMEDIATION

This section briefly reviews some of the local cases that have previously employed commercial nanomaterials for pollution mitigation and site remediation. These examples give us a general sense of the technical feasibility, running cost, and treatment capability, which are important for developing feasible engineering solutions for this NJDEP project.

(1) Klockner Road Site, Hamilton Township, NJ

A patented BNP-based product known as NanoFe PlusTM, developed by PARS Environmental, Inc., is used for remediation at the Klockner Road Site. PARS Environmental, Inc., reports that the reactive agent is administered in a water-based slurry containing one pound of reactive material in each 4 gallons of solution. The nanomaterial manufacturing plant is located in Trenton, NJ.

(2) PermeOx® Plus Used for Remediation of petroleum contaminated soil Successful treatment of petroleum contaminated soil using PermeOx[®] (containing nano-sized calcium peroxide or CaO₂) was also demonstrated elsewhere. A spill of diesel fuel occurred in a high traffic area in the winter. Because the spill occurred on a busy highway interchange, cleanup efforts had to be quick and non-invasive, with minimal mobilizations to the site. PermeOx® Plus was tilled into the top 3 feet of contaminated soil to enhance the microbial degradation of the remaining fuel contamination. Injection applications of a 25%-65% slurry containing calcium peroxide are usually performed at a typical rate of 0.1%-1.0% by weight on soil (approx. 2-6 pounds/cubic yard of soil). After only 85 days, the total Petroleum Hydrocarbon (TPH) concentrations in soils decreased from 1,500 mg/Kg to the cleanup goal of 100 mg/Kg. It is noteworthy that the actual TPH degradation rate from the addition of PermeOx Plus far exceeded the anticipated rate with ambient temperatures in the 20°F range as illustrated in Fig. 7.



Data provided by FMC Corporation

Fig. 7. Degradation of TPH in the presence of PermeOx Plus containing nano-sized calcium peroxide or (CaO₂).

(3) PermeOx® Plus Used for Remediation of underground water

A large volume of gasoline and related petroleum products had leaked from a major petroleum storage terminal over a period of time causing soil and groundwater contamination with benzene, ethylbenzene, toluene, and xylenes (BTEX) over an area approximately three acres in size. Concentrations of up to 3,700 µg/l BTEX were detected in source area groundwater on-site over the last 10 years. After several other technologies and products failed to lower site-wide BTEX concentrations, a slurry of PermeOx® Plus was injected into groundwater using a grid injection strategy within the contaminant plume. The dissolved oxygen (DO) levels in groundwater rose immediately after injection and remained elevated after 150 days. After three months, the average BTEX concentration in groundwater across the site dropped 77%, to an average concentration of 258 µg/l as shown in Fig. 8. The most dramatic decrease in concentrations occurred in two wells, where concentrations dropped 100%, from roughly 450 µg/l to non-detectable levels. The application of PermeOx® Plus as a slurry had increased the DO in groundwater to over 20 mg/l (supersaturated) in many areas across the site. This was found to be sufficient to enhance the natural degradation process, which had stalled due to lack of oxygen.



Fig. 8. Degradation of BTEX and increase of DO after addition of PermeOx chemicals.

ENGINEERING APPROACHES AND COST ANALYSIS

A majority of earlier work focused on specific contamination sites. Our proposed work will potentially apply a large quantity of nanomaterials in an aqueous suspension form onto the contaminated areas. The dose (concentration multiplied by volume) will be important for both contaminant removal and the operational cost. Both preventive and protective purposes should be fulfilled through the application of nanomaterials. In the event of flooding, the spread of contamination plume should be effectively stopped or directed to specific detention areas for further remediation through constructing small ditches or canals. In order to be effective, application techniques must be customized to meet the demands of a particular site, but typically include the following as illustrated in Fig. 9:

Option 1. Direct application (spray or mechanical mixing) in an excavation

Option 2. Subsurface injection in a grid within the contaminant plume

Option 3. An injected leading edge barrier

Option 4. Injection into an existing well



Fig. 9. (a) Modes of application of nanomaterials on contaminated topsoils. (b) Liquid spray. (c) Mixing nanomaterials with contaminated topsoils.

The price of nanomaterials varies significantly depending on a number of factors including raw material cost, manufacturing cost, licensing fees, and other economic factors (such as supply and demand). As a result of significant variability in the type of nanoscale metals and

catalyst/support selected, these nanomaterial products can vary significantly in physical-chemical characteristics and performance. Table 2 summarizes the potential costs for the proposed nanomaterials for consideration and their suppliers.

Process mechanisms	Nanomaterials	Target contaminants	Provider(s)	Cost
Adsorption	Iron oxides (α- Fe ₂ O ₃)	Heavy metals, organic compounds, arsenic	Nanostructured&Amorphous Materials, Inc.	\$0.16/g~\$0.8/g (30-50 nm)
Redox	nZVI (nanoscale zero valence iron)	 Halogenated aliphatics (PCE, TCE, 1,1,1-TCA, 1,1,2,2- TeCA) Halogenated aromatics PCBs Halogenated herbicides and pesticides Nitroaromatics Metals (e.g., Cr-, As) 	PARS Environmental Inc. Nanitech, LLC OnMaterials, Inc.	\$0.06g~0.12/g \$0.17/g \$0.05/g
reactions	Calcium peroxide	 Chlorinated Solvents BTEX PAHs Oxeygenates Petroleum hydrocarbons Phenols Chlorobenzenes 	FMC Corporation Solvay Chemicals	\$0.01/g
-	Calcium oxide (CaO)(Roy et al., 2013)	Microbial pathogen including bacteria, fungal, virus, parasite	Strem Chemicals, Inc Sigma-Aldrich	\$40/g (<20 nm) \$3.3/g (<160 nm)

Table 2. The proposed use of nanomaterials for remediation and the cost ranges.

At the treatment sites mentioned above, the total NZVI injection implementation costs were in the range of \$250,000 to 300,000. The desired levels of performance (treatment), the types of nanomaterials and different injection techniques could all vary the operational cost eventually. For instance, the cost of the ZVI material itself also was similar (\$32,500-\$37,500); however, the price (unit cost) of the micron-sized iron used can be much lower than the price of the NZVI used. If the primary objective of the treatment is to deliver sufficient iron mass into the aquifer, then the microscale iron appears to be more cost-effective than NZVI. The microscale iron also appears to sustain reductive conditions longer, enabling continued treatment of residuals. Therefore, the recommendation is that depending on specific contamination sites, characteristics, and desirable levels of remediation, using nanomaterials are highly appealing due to the demonstrated success in local areas of NJ in removal of various contaminants (mostly organic chemicals). Furthermore, pathogen removal is suggested although there is no documented evidence on the relationship between flooding events and pathogenic disease outbreaks. Finally,

heavy metals are largely ignored, which similar to the trace levels of carcinogenic compounds such as dioxin could contribute to the chronic diseases and health issues upon exposure. Thus, a combination use of different nanomaterials could be attempted to achieve removal of multiple contamination. Depending on funding availability, further studies may consider performing lab experiments to simulate contamination and decontamination processes in water and soils using the representative contaminants and the corresponding nanomaterials.

EXPECTED BENEFITS

This project will catalyze the nanotechnology applications and business opportunities while pursuing the goal of hazard mitigation after extreme flooding events. The rapid hazard removal enabled by various available nanomaterials will facilitate the environmental quality restoration and ensure economic and social development. This project will also lead to broader impacts by linking industrial manufacturers, regulatory authorities, researchers and the local public in developing rapid hazard mitigation and management during the aftermath of extreme hurricane and flooding impacts.

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