



**Conference
on the
Remediation
of Sediments
ABSTRACTS
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P. O. Box 231, New Brunswick, New Jersey, 08903-0231
908/932-6555, FAX 908/932-8578

THE STATE UNIVERSITY OF NEW JERSEY
RUTGERS

Bob Gambrell
Wetland Biogeochemistry Inst.
LSU
Baton Rouge, LA 70803
504-388-6426
FAX 504-388-6423

**Environmental Chemistry of Metals in
Sediments and Dredged Materials**

The physical and chemical properties of sediments have an important influence on how strongly trace and toxic metals are immobilized. Depending on disposal methods selected for contaminated dredged sediments, metals may be effectively immobilized at a disposal site, or, changes in sediment chemistry may occur that will result in some release to more mobile and biologically available forms.

The chemistry of metals in sediments and dredged material will be discussed in terms of sediment properties, and examples given from various research projects demonstrating how selection of disposal methods and management of dredged sediments may be used to minimize contaminant release.

Ken Nealson
Center for Great Lakes Studies
600 East Greenfield Avenue
Milwaukee, WI 53204
414-649-3006
FAX 414-649-3005

**The coupling of anaerobic organic carbon oxidation
to the reduction of iron, manganese, or sulfur
by *Shewanella putrefaciens***

During our studies of the geochemistry of metals in freshwater and marine ecosystems during the past few years, we have isolated microbes in the group *Shewanella putrefaciens*, which have the ability to grow anaerobically using manganese or iron oxides as oxygen substitutes for respiration. These bacteria can utilize a wide variety of different electron acceptors in addition to Mn or Fe, including oxygen, sulfite, thiosulfate, elemental sulfur, nitrate, nitrite, trimethylamine-N-oxide, dimethyl sulfoxide, fumarate and glycine. Strains of *S. putrefaciens* have been isolated from a wide variety of marine and freshwater environments, suggesting that this group is ubiquitously distributed in environments containing oxic/anoxic interfaces. The reduction of iron, manganese or elemental sulfur, all of which are solid substrates, requires cell contact with the substrates, and during the reduction. This raises the possibility that such organisms can be isolated, or genetically engineered, which can be used to oxidize specific contaminating substrates, such as phenolics, naphthalenes, etc., using metal oxides or sulfur as electron acceptors. With regard to the latter possibility, we have now isolated key mutants with impaired abilities to anaerobically reduce iron, manganese and sulfur compounds, as well as regulatory mutants which are incapable of anaerobic respiration. These mutants have been complemented using a library prepared from wild type *S. putrefaciens*. Methods for introducing attached organisms to polluted environments are now under development in our laboratory and will be presented.

Louis J. De Filippi
Allied-Signal
50 East Algonquin Road
P.O. Box 5016
Des Plaines, IL 60017-5016

Bioremediation of Hexavalent Chromium Using Sulfate Reducing Bacteria

Chromates [hexavalent chromium, Cr(VI)] have found their way into the marine environment. This is of potential concern since Cr(VI) is a strong oxidant, is toxic, has a propensity to pass through mammalian cell membranes and, due to its high water solubility, has the potential for groundwater migration. Furthermore, Cr(VI)-contaminated groundwater or industrial effluent is often at an extreme pH, necessitating pH adjustment prior to treatment. In comparison to Cr(VI), the reduced form, Cr(III), possesses the properties of low toxicity, resistance to passage through membranes, low aqueous solubility (K_{sp} for $\text{Cr(OH)}_3 = 6.7 \times 10^{-31}$) and, consequently, low potential for groundwater migration. Therefore, reduction of Cr(VI) to Cr(III) is desirable from multiple standpoints. In a marine environment reduction of Cr(VI) by bacterially produced H_2S occurs naturally (1) as long as the appropriate nutrients are present. We have developed a bioremediation process for rendering Cr(VI) present in aqueous and solid wastes (especially those containing a high concentration of ionic species) insoluble by reductive precipitation as Cr(III). The method first involves adaptation of a marine-derived, anaerobic, sulfate reducing consortia for growth on an inexpensive carbon source, such as molasses or acetate, in a high ionic strength, sulfate-containing mineral salts medium. Bacterially produced H_2S yields Cr(VI) reduction to Cr(III) which precipitates as an extremely insoluble species within the pH range at which the fermentation is conducted. For Cr(VI)-containing waste of an alkaline nature we have adapted the consortia for growth at a high pH as follows. Mineral media supplemented with various nutrients and sodium thioglycollate was adjusted to pHs ranging from 6.75 to 9.52 placed under a nitrogen blanket in a sealed vessel and inoculated with a mixed sulfate reducing culture. At pHs up to about 9 sulfate reducing microorganisms grew readily. H_2S and black metal sulfides were produced. For a continuous process, standing water in contact with chromium ore tailings at a NJ site, possessing a Ph of 12 and containing 11 ppm Cr(VI), and supplemented with 38mM SO_4^{2-} , was passed into a mixing tank where the pH was adjusted to between 7 and 8 with 25% v/v acetic acid and then into a gravel and sulfate reducing sludge blanket-containing, 6 x 36 cm (ID x h) anaerobic column reactor in a upflow fashion. At dilution rates up to 0.05 h^{-1} the effluent contained $<0.21 \text{ ppm}$ Cr(VI). Inclusion of micronutrients allowed an increase to 0.1 h^{-1} with no detectable effluent Cr(VI).

Hans Stroo
Remediation Technologies
1011 Klickitat Way
Suite 207
Seattle, WA 98134
206-624-9349
FAX 206-624-2839

Slurry-Phase Bioremediation of Contaminated Sediments

Slurry-phase bioremediation is one option for biologically treating contaminated sediments. The paper will present the basic concepts of the technology, and experiences with slurry-phase treatment. The advantages of this system for contaminated sediments will be discussed, and laboratory-scale results will be presented for three sediments. The results will be compared to conventional bioremediation (land treatment), and thermal desorption treatment. The design and operation of a currently operating mobile pilot-scale slurry-phase bioreactor will be presented. Finally, a conceptual approach to full-scale treatment will be development, to allow an initial assessment of the feasibility of the approach from an engineering and economic perspective.

Philippe Corbisier
Department of Microbiology and
Immunology
University of Illinois at
Chicago
E-703 Medical Sciences Addition
835 South Wolcott Avenue
P.O. Box 4348
Chicago, IL 60612
312-996-8683
FAX 312-996-6415

Overview of Bacterial Heavy Metal Resistances

Bacterial plasmids determine resistance to a wide range of toxic metals including Hg^{2+} , Cd^{2+} , Zn^{2+} , AsO_2^- , AsO_4^{3-} , CrO_4^{2-} , Cu^{2+} , Pb^{2+} , SbO_2^- and Tl^+ . Our laboratories in Mol and Chicago are involved in studies of the basic mechanisms of bacterial resistance to heavy metals with the hope that someday the bacterial resistance systems may provide a basis for bioremediation of toxic waste or biosensing of environmental contamination. Different possible resistance mechanisms for mercury, cadmium, arsenic and copper will be presented. It will be shown how a reporter gene such as lux when fused to heavy metal operon can provide a rapid measurement of bioavailable toxic metals in aqueous environments, a mer-lux fusion, ars-lux fusion, cop-lux fusion and th-lux fusion will be presented. An experimental flat sheet reactor used to remove cadmium from polluted waste water using immobilized *Alcaligenes eutrophus* bacteria will be presented as an example of bacterial bioremediation.

Gene Madsen
WG21 Wing Hall
Dept. of Microbiology
Cornell University
Ithaca, NY 14853
607-255-2417

**A View Toward Bioremediation Based on Micro-biological Studies
of a Contaminated Aquifer**

Decades of research using laboratory flask assays have proven that microorganisms are capable of detoxifying and/or completely mineralizing many environmental pollutants. Bioremediation is a promising technology that would manage microbial processes in order to destroy contaminant compounds in field sites. Although microorganisms have successfully and spontaneously maintained the biosphere since its inception, industrialized societies now produce undesirable chemical compounds at rates that outpace naturally occurring microbial detoxification processes. An essential step toward attaining reliable bioremediation technologies is the development of criteria which prove that microorganisms in contaminated field sites are truly active in metabolizing contaminants of interest. These criteria rely upon genetic, biochemical, physiological, and ecological principles and apply to both *in situ* and *ex situ* bioremediation strategies. My research group has studied a field site whose ground water is contaminated with polycyclic aromatic hydrocarbons (PAHs) derived from coal-tar waste. We discovered that the naturally occurring ground-water microbial community was metabolically adapted to PAHs and contained enhanced numbers of protozoan predators inside but not outside the plume of contamination. *In situ* biodegradation was inferred because predators can only be supported by *in situ* growth of prey, which was linked to the contaminant PAHs (Sciences. 1991. 252:870). These results are an example of how the science of biodegradation can begin to deliver the technology of bioremediation.

Ronald Unterman
Envirogen, Inc.
4100 Quakerbridge Road
Lawrenceville, NJ 08648-4702
609-936-9300
FAX 609-936-9221

Biological Treatment of Chlorinated Organics

Over the past 20 years there has been extensive research into the microbiology, biochemistry, and most recently, process development for the biological destruction of chlorinated organics. This paper describes some of the alternative systems that are currently under development for bioremediation systems. These include the contrasting, yet complementary approaches of: 1) aerobic vs. anaerobic microbes, 2) indigenous vs. exogenous microbes, and 3) *in situ* vs. *ex situ* (reactor-based) treatment cells.

Naturally occurring bacteria are capable of degrading nearly all natural organic compounds. However, bacteria capable of degrading many synthetic chemical substances (such as PCBs and TCE) are much rarer. In addition, these synthetic chemical compounds tend to be more stable, which means that it may take years before such compounds are naturally degraded.

Bioremediation companies are now using isolated superior natural strains of bacteria that partially or completely degrade or accelerate the degradation of a number of complex hazardous wastes, including PCBs, TCE and other chlorinated solvents and PAH. These improved bacteria are generally isolated from contaminated sites using sophisticated enrichment techniques which allow scientists to select and isolate superior strains from the general population found at these sites. There is also activity in the design and testing of genetically modified bacteria for commercial use. These strains have many advantages over naturally occurring bacteria, including the ability to degrade wastes faster and to lower levels and at a lower overall cost. Also under development are pretreatment techniques for separating contaminants from the media in which they are found (such as soil or water) in order to enhance the effectiveness of the bacteria in degrading the hazardous waste.

Engineered systems using bioreactors or contained environments in which biodegradative microorganisms make contact with the contaminated air, water, sediment, or sludge are also under development. By use of a bioreactor, variables such as temperature and pH can be controlled and measured amounts of oxygen and nutrients can be added to the mixture of microorganisms and contaminated materials, thereby optimizing the degradation environment. *In situ* approaches are also under development for soil, sediment, and groundwater and ultimately these should be the most cost effective clean up technologies.

Carl E. Cerniglia
National Center for Toxicological
Research - Microbiology Div.
NCTR Drive
Jefferson, AR 72079-9502
501-543-7341
FAX 501-543-7576

**Biodegradation of polycyclic aromatic hydrocarbons by a
Mycobacterium sp.**

Bacteria capable of polycyclic aromatic hydrocarbon (PAH) degradation in pure culture and under simulated environmental conditions, including some that are able to oxidize or co-oxidize higher molecular weight PAHs, have been isolated in several laboratories. Research in my laboratory has documented the extensive mineralization of pyrene and fluoranthene by a *Mycobacterium* sp. originating from a chronically polluted location. Additionally, this *Mycobacterium* sp. has been shown to mineralize naphthalene, phenanthrene, 1-nitropyrene, anthracene, 6-nitrochrysene and 3-methylcholanthrene. Addition of the *Mycobacterium* sp. to non-sterile microcosms containing soil with no additional organic carbon source significantly enhanced pyrene and fluoranthene mineralization over that performed by the indigenous biota alone. The effect of one PAH on the mineralization rate of another PAH in multicomponent mixtures and the potential usefulness of the *Mycobacterium* sp. for the bioremediation of higher molecular weight PAHs in mixtures will be discussed.

Roger Prince
Exxon Research and Engineering
Clinton Township Route 22E
Annandale, NJ 08801
908-730-0100
FAX 908-730-3042

Assessing Bioremediation in Prince William Sound

Bioremediation played an important role in the clean-up of the oil spill in Prince William Sound, Alaska. Widespread application of fertilizers to oiled beaches was accompanied by a substantial monitoring program that focussed on assessing the safety and efficacy of the process. A variety of samples were collected and analyzed; interstitial water from within the oiled sediment was analyzed for nitrogenous nutrients (NH_4^+ , NO_3^- , soluble organic N), phosphate, dissolved oxygen, salinity, temperature and pH, sediment samples were analyzed for microbial populations and activities, and oil content and chemistry, and nearshore water was analyzed for acute toxicity, increased algal productivity, and hydrocarbons. Taken together, the data clearly indicated that bioremediation was both safe and effective in stimulating the natural process of biodegradation of the spilled oil.

Daniel Abramowicz
General Electric
Research and Development Center
Bldg. K-1 Room 3B19
P.O. Box 8
Schenectady, NY 12301-0008
518-387-7072
FAX 518-387-5604

PCB Biodegradation in the Laboratory and the Environment

Aerobic PCB biodegradation and anaerobic PCB dechlorination have been demonstrated in the laboratory with contaminated soils and sediments. In the summer of 1991, GE performed a large scale test of aerobic PCB biodegradation in the Upper Hudson River. The experiments involved six sealed caissons (six feet in diameter) lowered into the contaminated sediments. Results demonstrated good correlation between laboratory derived expectations and actual field performance. A large scale test of anaerobic PCB dechlorination in Housatonic River sediments is planned for the summer of 1992.

Jack Jones
Environmental Protection Agency
Environmental Research Lab
College Station Road
Room 151, Mail Code EPA8431
Athens, GA 30613
404-546-3468

Decontamination of PCB-Contaminated Sediments Through the Use of Bioremediation Technologies

Pollution control agencies have launched a concerted effort to reduce the environmental levels of accumulated polychlorinated biphenyls (PCBs) because of their persistence, toxicity and tendency to bioconcentrate (1-3). PCBs can be transformed into less harmful compounds by chemical, physical and biological methods; e.g., ultraviolet radiation, microwave treatment, and incineration (4). For contaminated sediments, microbial degradation offers a cost-efficient method of reducing PCB levels. Consequently, the coupling of anaerobic dechlorination to aerobic metabolism has been suggested as a possible cleanup method.

Results from bench scale studies suggest that PCBs can be biodegraded by both aerobic and anaerobic microbial processes (5,6). Aerobic bacteria usually degrade PCB congeners having one to five chlorine atoms, but not the higher-chlorine substituted congeners. The rate and extent of degradation, however, is influenced by the position of the chlorine atoms on the biphenyl ring (7, 8). For example, congeners with open 2, 3- or 3, 4-positions are degraded faster than congeners with chlorine atoms at these positions. These sites are necessary for the action of dioxygenase enzymes to effect the fission of the biphenyl ring. Several bacterial strains have been isolated from enrichments of contaminated soils and sediments that can biodegrade PCBs aerobically. These strains vary in their abilities to degrade PCBs; some degrade only mono-, di and trichlorobiphenyls, and others degrade tetra- and pentachlorobiphenyls (7). Anaerobic reductive dechlorination of PCBs was first proposed by Brown and coworkers (9). Later reports from laboratory studies provided unequivocal evidence that reductive dechlorination could occur in anaerobic sediments (10). Under anoxic conditions, the highly chlorinated PCB congeners are degraded readily, whereas the less chlorinated congeners and the biphenyl nucleus are more persistent (5). There is a preferential removal of chlorines from the para and meta positions. The products of anaerobic dechlorination, therefore, are mainly ortho-substituted congeners, which have fewer chlorine atoms on the biphenyl ring. Such congeners are less toxic and may serve as substrates for aerobic dechlorinating bacteria (10).

The objective of the current study is to determine the dechlorinating capacity of naturally occurring microbes in PCB-contaminated sediments from selected Great lakes sites. The dechlorination of indigenous PCB congeners in contaminated sediment

from the Saginaw River was determined under aerobic conditions. The initial chromatographic profile of Saginaw sediment indicated a preponderance of PCB congeners with one to four chlorine atoms, which suggested that the original Aroclor contaminant had been transformed from a mix of congeners of low and high chlorine content to primarily low-chlorine congeners. Aerobic incubation for 6 months resulted in a net loss of total congeners reported to be amenable to aerobic degradation. These congeners included 4-chlorobiphenyl, 2, 2'-trichlorobiphenyl (tri-CB), 2, 4', 5-tri-CB, 2, 4, 4'-tri-CB, 3, 4, 4'-tri-CB, 2, 3', 5', 6-tetrachlorobiphenyl (tet-CB) and 2, 2', 3, 4'-tet-CB.

Contaminated sediments collected from the Ashtabula River and Sheboygan Harbor were incubated under anaerobic conditions. These sediments also were depleted of highly chlorinated congeners. The Ashtabula sediment was amended with 2, 3, e', 4, 4'-pentachlorobiphenyl (penta-CB), 2, 3, 3', 4, 4', 5-hexachlorobiphenyl (hexa-CB), and 2, 2', 3, 4, 5, 6, 6'-heptachlorobiphenyl (hepta-CB) to determine the potential for dechlorination of highly chlorinated congeners. Each congener was added individually and in combination with the other two congeners. Reductive dechlorination was observed after lag periods of 5, 3, and 4 months for the penta-CB, hexa-CB and hepta-CB, respectively. The Sheboygan sediment was amended with three concentrations of 2, 2', 3, 3', 4, 5, 6, 6'-octachlorobiphenyl (octa-CB). After a lag phase of 5 months, a decrease in many of the higher chlorinated congeners was observed in all the samples amended with the octa-CB, but not in the unspiked and autoclaved sterile controls. These preliminary data suggest the presence of actively dechlorinating microorganisms in the sediments from these Great Lakes sites. Further research will include studies to determine the factors that will enhance the dechlorinating activity of these microorganisms.

A field demonstration is proposed for the Sheboygan River area using dredged sediment that has been placed in a Confined Treatment Facility. This study will examine the effects of alternating aerobic and anaerobic conditions, with and without the addition of inorganic nutrients, on the rate of extent of PCB biotransformation under simulated field conditions. Collecting biotransformation data under conditions that closely resemble a large scale treatment of PCBs will provide valuable information regarding the correlation of laboratory and field data and the development of scenarios for enhanced bioremediation.

Addition of inorganic nutrients and anaerobic-aerobic cycling are currently the most promising treatment options; both will be examined in the field scale study. During the aerobic phase, the bioavailabilities of PCBs may be enhanced through the degradation of naturally occurring organic compounds as well as contaminants such as oil and grease, all of which are known to bind PCBs. Further, biphenyl and lower chlorinated PCB congeners, primarily mono- and dichlorinated PCBs, are degraded preferentially under aerobic conditions (7, 8). Anaerobic conditions favor the biotransformation of the more highly chlorinated PCB congeners (6,

7, and 8 chlorines) through reductive dechlorination reactions (9, 10), producing lower chlorinated products amenable to aerobic attack. Taken together, the alternating aerobic/anaerobic scheme seems promising for more extensive bioremediation of PCBs (6). Concurrent lab experiments will verify and expand the field study.

Mark Brown
Blasland, Bouck & Lee
6732 Towpath Road
Box 66
Syracuse, NY 13214
315-446-9120
FAX 315-445-9161

**Sheboygan River Sediment Bioremediation Project -
Development of the Confined Treatment Facility**

A Confined Treatment Facility (CTF) was designed to demonstrate the bioremediation of PCBs in sediments removed from the Sheboygan River, Wisconsin. The pilot-scale project is being conducted by Blasland & Bouck on behalf of the Tecumseh Products Company and in cooperation with the USEPA. The CTF was designed and constructed to receive 2,500 cubic yards of sediment. The purpose of the CTF was to maintain an "aquatic" environment and enhance anaerobic dechlorination or aerobic biodegradation of PCBs. The design requirements included environmental controls such as water treatment and multiple barriers to chemical migration; ability to manage solutes within the interstitial water; and, the need to simultaneously evaluate different experimental treatments with appropriate controls.

The resulting CTF structure is an open vessel occupying 14,000 square feet of floodplain adjacent to the Sheboygan River. The CTF is constructed with interlocking sheetpiling walls and double high-density polyethylene (HDPE) liners along the walls and bottom. Other features include independent leachate collection and detection systems and a "passive" water-treatment system. The CTF was divided into four separate cells: two large cells for experimental treatments and two small cells to serve as corresponding controls. At the bottom of each cell is a separate amendment-delivery system consisting of perforated HDPE pipe. The cells were filled during 1989 and 1990 with sediment removed from selected deposits which contained the highest PCB concentrations in the Sheboygan River. The extent of PCB dechlorination evident in the baseline samples from the CTF is significant. The course of future work in the CTF, which is being undertaken in cooperation with the EPA's Assessment and Remediation of Contaminated Sediments program, and the parallel bench-scale treatability studies are described in following presentations.

Loring Nies
Environmental & Water Resources
Engineering
Engineering Bldg. 1A, Room 181
University of Michigan
Ann Arbor, MI 48109-2125
313-764-6488
FAX 313-763-2275

**Reductive Dechlorination of Polychlorinated
Biphenyls (PCBs) in Sediments**

Polychlorinated biphenyls (PCBs) have on occasion partitioned into sediments at concentrations sometimes greater than 50 ppm near sites of previous industrial discharges. Some locations with elevated PCB sediment concentrations are being considered for remediation. These PCBs are potentially available for microbially-mediated transformations, such as reductive dechlorination. Anaerobic reductive dechlorination of PCBs has been observed both in the environment and in laboratory studies. Dechlorination may reduce toxicity and increase the fraction of PCB congeners that are more readily biodegraded by aerobic microorganisms. Hence, the ability to enhance reductive dechlorination in natural sediments would be beneficial. Anaerobic batch incubations, using naturally occurring microorganisms from sediment-substrate enrichments, were tested to assess the potential for bioremediation of PCB dechlorination, or if dechlorination rates were limited by lack of microorganisms, or the bioavailability of the PCBs. The experiments show that in some cases the substrate (electron donor) can affect the dechlorination rate. The rate of reductive dechlorination of PCBs was proportional to the amount of organic substrate available in some sediments. However, the relative rates of reductive dechlorination of PCBs added to sediments in the laboratory were faster than the dechlorination rates of "weathered" PCBs that may have resided in sediments for years.

John F. Riddell
Dept. of Civil Engineering
University of Strathclyde
Glasgow, UK
011-44-41-552-4400
FAX 011-44-41-553-2066

Dredged Material to Soil - UK Examples of a Beneficial Use

Changing patterns of shipping and trade have resulted in many traditional dock facilities in cities worldwide becoming redundant. As the facilities are closed, redevelopment of the sites takes place, work which frequently involves the creation of large areas of amenity landscaping and environmental improvement. This creates a demand for soil able to support trees, shrubs, grass and flowers.

The closure of the shipping facilities may also mean a reduction in the need to undertake maintenance dredging of the docks and channels. Silt, much of which is derived from land erosion, starts to accumulate and may build-up to create environmentally unattractive mudbanks.

The recycling of the mud deposits to create a low cost source of amenity landscaping soil is a beneficial use of dredged material which has now been undertaken in a number of UK cities. The paper describes some of the difficulties and solutions appropriate to reuse, including contamination, drying, desalination and amelioration. Two case studies are examined in detail. In one the recovery and reuse is now undertaken on a fully commercial basis by a private contractor.

H.J. van Veen
Dept. of Environ. Technology
T.N.O.
P.O. Box 342
7300 AH Apeldoorn
Laan van Westenenk 501
The Netherlands
011-31-55-493-922
FAX 011-31-55-419-837

Remediation of Contaminated Sediments in the Netherlands

Contaminated sediment is a high-priority environmental problem in the Netherlands. It is mainly a dredged-material problem: many Dutch watercourses must be dredged for nautical reasons and for water management. Consequently, remediation of contaminated sediments in the Netherlands refers specifically to dredging and dredged-material treatment. Until a few years ago, all dredged material was removed to confined disposal facilities. Recently, treatment technology has been applied in sediment remediation actions. Jointly with these full-scale operations, a research programme has been executed: the first phase of this Development Programme Treatment Processes Programme was executed in 1989 through 1991, which the second phase is to start in 1992.

Tiedo Vellinga
Department of Public Works
P.O. Box 6633
Rotterdam 3002-AP
The Netherlands
011-31-10-489-4664
FAX 011-31-10-489-4547

Management of Dredged Material in the Port of Rotterdam

About 23 million m³ of dredged material must be removed from the port of Rotterdam yearly. Thirteen million m³ of this, mainly of marine origine, is relocated in the sea. The remaining 10 million m³, is contaminated to such a degree, that disposal into the sea or relocation on land sites is not permitted, unless special measures are taken.

The long term strategy of the port is that the dredged material should be clean. This will entail an improvement in quality of between 70% and 90%. In the meantime (until 2002 A.D.) the contaminated dredged material will be disposed into 2 specially constructed disposal sites. Strategy for these disposal sites is an efficient and sound use of the precious disposal capacity. Therefore research and experiments have been carried out to examine the possibilities for separation of fractions (contaminated versus non-contaminated) dewatering and re-use. Next to that the port of Rotterdam participates in the national "Development Program Treatment Processes". In this program all kinds of treatment and cleaning techniques are examined on different scales.

To implement the long term strategy, the Rhine Research Project (RRP) was initiated. This project aims to contribute to the ultimate goal through technical and legal investigation, public relations activities and a dialogue with the discharges of effluent. To date these efforts have resulted in agreements between the city of Rotterdam and effluent discharges in Switzerland, France and Germany.

The framework for all these activities is the management program for dredged material.

During the presentation the features and results of this management program will be highlighted.

