PROPOSAL

EMISSIONS AND ATMOSPHERIC TRANSPORT OF PCBs AND Hg FROM STABILIZED HARBOR SEDIMENTS

SUBMITTED TO:

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1.0 INTRODUCTION

The disposal of sediments removed from navigation channels by maintenance dredging has become a significant problem for many ports and harbors. The annual maintenance dredging at all ports in the United States yields over 3×10^8 m^3 of dredged material. In New York Harbor alone, maintenance dredging produces an annual volume of about 3×10^6 m^3 of dredged material (Weinstein and Douglas, 2000). Much of this material is contaminated and has in recent years been ruled unsuitable for ocean disposal. One alternative to ocean disposal is to use dredged materials, stabilized with cement or lime, as fill at construction sites. The NJ Marine Sciences Consortium (NJMSC) and the NJ Commerce and Economic Growth Commission, Office of Maritime Resources (NJEGC/NJMR) recently provided funding to Rutgers University and Stevens Institute of Technology for a study entitled: *Monitoring of PCB and Hg Air Emissions in Sites Receiving Stabilized Harbor Sediment.* This project was designed to assess the land-air emission rates of polychlorinated biphenyls (PCBs) and mercury (Hg) at an 80 ha site in Bayonne, New Jersey (Fig. 1) where up to 3×10^6 m^3 of stabilized dredged material (SDM) from the NY/NJ Harbor will be used as landfill. The primary driver behind this research was to assess the impact of land-applied harbor sediments on ambient air concentrations of PCBs and Hg in the city of Bayonne.
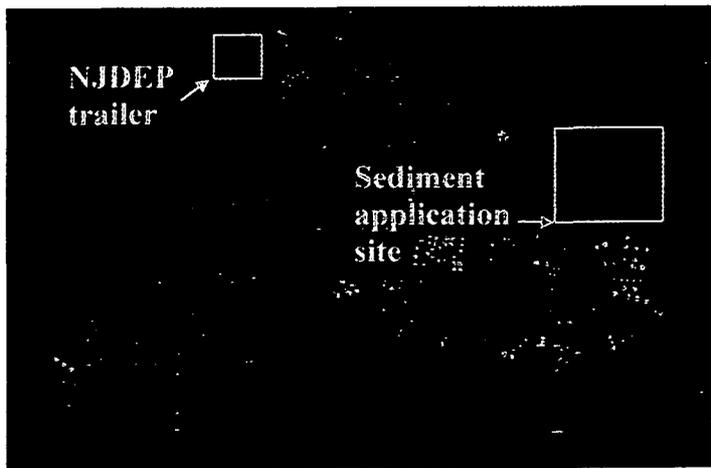
Figure 1. Aerial view of the Bayonne peninsula showing the sediment application site and the NJDEP air monitoring trailer where background concentrations of PCBs and Hg were measured.

The SDM at the Bayonne site comes from maintenance dredging operations in New York Harbor, primarily from the channels and berthing areas for containerships such as those in Newark Bay. These sediments contain many types of contaminants including heavy metals, polyaromatic hydrocarbons, and polychlorinated biphenyls (PCBs). According to Ho et al. (2000), the average PCB concentration in New York Harbor exceeds 400 μg/kg of dry sediment. This average value for PCBs exceeds the concentration for toxic effects in marine organisms. Mercury, Hg, is also a major contaminant of concern. Among large East Coast urban estuaries, the lower NY/NJ Harbor Estuary has some of the highest concentrations of Hg in surficial sediments (EPA, 1997). The primary form of Hg in estuarine sediments is oxidized Hg(II)

bound in sulfide solids or adsorbed to organic-rich mineral phases, with monomethylmercury (CH_3Hg) accounting for < 2% of the total (Mason and Lawrence, 1999).

Emissions of PCBs and Hg from SDM

Prior to placement, dredged sediments are mixed with cement and the resulting SDM contains 12% cement by weight. This processing of harbor sediments and their subsequent placement on land could lead to the volatilization of gas phase PCBs and Hg. Enhanced gaseous PCB and Hg release from dredged sediments may occur during stabilization, drying, and compaction due to heat build up, or the chemical production of volatile forms of inorganic mercury including Hg° and $HgCl_{2(g)}$. Sediment-bound Hg may become volatile upon exposure to sunlight as a result of the photochemical reduction of Hg(II) to Hg°. Emissions of PCBs and Hg from native, processed, or land-applied sediments could be important in evaluating the environmental impacts of various uses of dredge materials and in furthering the understanding of PCB and Hg cycles in urban harbors.

The extent and rate of drying of the SDM will have a large effect on the magnitude of PCB and Hg fluxes from this material. A laboratory study conducted with contaminated St. Lawrence River sediment found a very high correlation between the evaporation rate of water and the amount of PCBs volatilized in the first 24 hours of drying (Chiarenzelli et al., 1996). As time passes however, and the SDM dries, fluxes will reduce consistently until a point when the sediment is effectively dry. At this point the fluxes will approach zero. In another study, sediment from a Federal Superfund Site was monitored for PCB losses over a period of two days. This study revealed that 21.3% of the PCBs were lost in the first 24 hours, but this loss stopped when the soil dried out. After the same sediment was rewetted an additional 7.5% of total PCBs were lost over a similar 24-hour span. During both of these time periods, PCB loss was positively correlated to loss of moisture in the soil (Chiarenzelli et al., 1996).

Preliminary results from the Rutgers-Stevens study demonstrate that significant volatilization of PCBs and Hg from the stabilized sediment is occurring. Average PCB concentrations at the sediment application site were found to be 1.5 to 2-fold higher than those measured at a background site (NJDEP air monitoring trailer) in Bayonne (Fig. 2). The high ambient PCB levels measured at the sediment application site relative to local background suggest that volatilization from the sediment is a significant source of local atmospheric PCBs. In addition, small-scale spatial gradients in PCB concentrations demonstrate that PCBs are enhanced downwind of the applied sediment relative to levels measured at upwind samplers. In the worst case, gas-phase PCB concentrations downwind were 4 times upwind levels. Total gaseous Hg concentrations were also found to be significantly higher (p < 0.01) at the sediment application site than at either the Bayonne or New Brunswick background sites (Table 1).

We have also estimated vertical gradients of PCBs and Hg at the sediment disposal site and estimated land-air vertical fluxes using a micrometeorological approach. The results indicate net land-air fluxes of PCBs and Hg during the day, but there is substantial uncertainty in these flux estimates due to difficulties in measuring the vertical concentration gradient of PCBs and atmospheric stability factors. Thus, it would be advantageous to estimate sediment-air PCB and Hg fluxes in controlled flux chamber studies to validate our current findings.
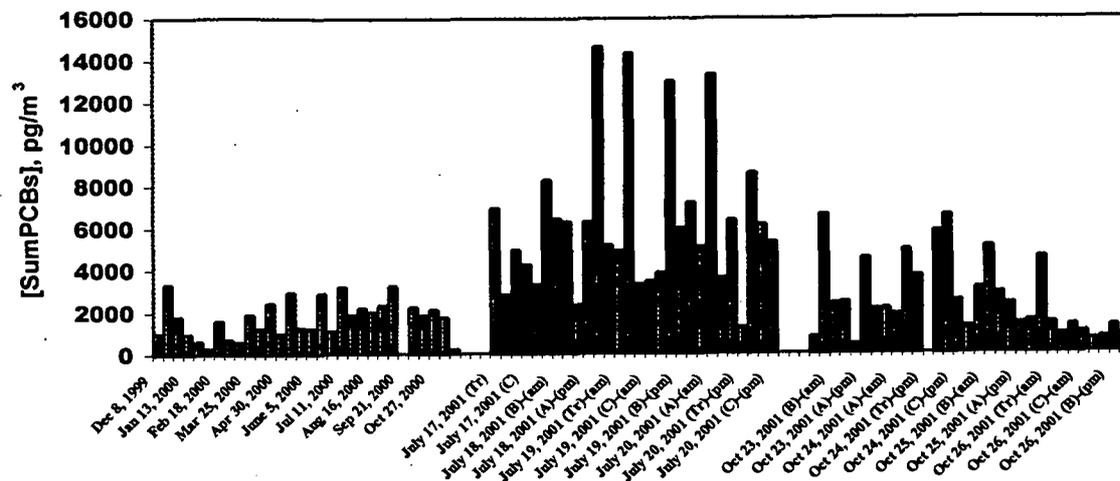
Figure 2. Concentrations of gas phase PCBs measured at the trailer site (blue) and at the sediment application site (red).

Table 1. Total gas phase Hg concentrations at the Bayonne landfill site (red) and at background sites (black) in Bayonne and New Brunswick, NJ.

Sampling location	Date	Hg (ng m^{-3})	
		Ave	Std
On-site	Aug 30, 2001	3.29	0.72
Background (Bayonne trailer)	Sept 17, 2001	2.06	0.20
Background (New Brunswick)	Oct 11, 2001	1.77	0.49
On-site	Oct 23, 2001	2.75	0.31
	Oct 24, 2001	3.54	0.81
	Oct 25, 2001	4.98	1.94
Background (Bayonne trailer)	May 6-7, 2002	2.53	0.33
On-site	May 7, 2002	3.23	0.44
	May 8, 2002	3.07	0.18

Local Atmospheric Transport of PCBs and Hg from SDM in Bayonne, NJ

Despite evidence that significant volatilization of PCBs and Hg from stabilized sediments is occurring, the extent to which these emissions negatively impact the air in Bayonne or in other populated areas of New Jersey is not clear. During three sampling intervals of the initial Rutgers-Stevens study, the background NJDEP trailer site was downwind of the Bayonne landfill under east/southeast winds. On July 18, 2001 in the afternoon, the concentration of gas phase ΣPCBs was 8700 pg/m^3 at the sediment application site, but just 2300 pg/m^3 downwind at the trailer site (Figure 2). Similarly, on July 20, 2001, the ΣPCBs concentration was 6700 pg/m^3 at the application site and 1300 pg/m^3 at the trailer. East/southeast winds also occurred on October 23, 2001 in the morning. On this day, the ΣPCB concentration at the sediment application site was 6619 pg/m^3 downwind of the applied sediment while across town at the NJDEP trailer it was

791 pg/m^3, almost a factor of eight lower. Thus high concentrations at the sediment application site do not necessarily lead to elevated PCB concentrations in the city of Bayonne.

We hypothesize that the atmospheric transport of PCBs and Hg in the Bayonne area is influenced by gas phase-aerosol exchange and the complicated local meteorology of the Bayonne peninsula. Gas-particle partitioning may remove much of the gas-phase PCBs and some of the gaseous Hg during transport across the city of Bayonne. PCBs and Hg are volatilized from the stabilized sediment into the gas phase. This volatilization is likely enhanced by the elevated temperature of the sediments, which are heated by the exothermic curing of the cement additives and by solar radiation. The air directly above the sediments is also heated, but as the air masses rise, they cool, potentially inducing the PCBs in the air to condense onto airborne particles. Under light winds, air movement across the peninsula will take an hour or more, allowing time for gas-particle exchange. Similar processes may also lead to the removal of gaseous Hg by aerosols particularly if Hg is emitted from the SDM as a reactive gaseous form (see below).

The concentrations of atmospheric PCBs and Hg experienced in the city of Bayonne may also be heavily influenced by fine-scale meteorology. Bayonne is located on a peninsula surrounded by the New York Harbor and Hudson River. The geography of the region lends itself to complicated local meteorology, in particular to sea breeze effects. In the summer, the air above the disposal site is heated by the disposed sediments, and rises. Cooler air from over the Hudson River may move in to replace it. As the rising air cools, it may move eastward over the Hudson River in the classic sea-breeze circulation pattern. In this scenario, the wind direction observed at the meteorological tower at the sediment disposal site could be southeasterly, but the trajectory of the PCB- and Hg-laden air mass might not take it toward the city of Bayonne.

In order to assess the impact of PCB and Hg emissions from SDM on local communities, it is necessary to understand how sediment properties and environmental factors affect sediment-air fluxes of PCBs and Hg and how gas phase-aerosol exchange and local meteorology influence their transport. We therefore propose to determine gas phase PCB and Hg fluxes from SDM in controlled, flux-chamber experiments. Using this information, we will attempt to model the volatilization of PCBs and Hg from SDM. We further propose to quantify particulate PCB (and possibly Hg) concentrations at the sediment disposal and trailer sites using filters collected during the initial Rutgers-Stevens project, and to combine our measurements of atmospheric PCB and Hg concentrations with regional atmospheric (RAMS) and chemical transport (HYPACT) models. The RAMS/HYPACT models will also be used to generate fluxes of PCBs and Hg from stabilized sediments for comparison with measured fluxes.

2.0 OBJECTIVES
The objectives of the proposed study are to:

1. Quantify through laboratory studies the magnitude of PCB and Hg volatilization from SDM in order to determine the sediment-side chemo-dynamic properties of SDM as it cures.
2. Quantify the fraction of PCBs (and possibly Hg) adsorbed to airborne particles in samples previously collected at the sediment disposal site and at the NJDEP trailer site in order to determine if the gas-particle system is at equilibrium and assess the importance of aerosol sorption of gas-phase PCBs and Hg emitted from the stabilized sediment.
3. Utilize a RAMS/HYPACT model with measured PCB and Hg concentrations to characterize air circulation patterns and estimate the fluxes of PCBs and Hg from the SDM during our previous field sampling campaigns.

3.0 APPROACH TO ADDRESS EMISSIONS OF PCBS AND HG FROM SDM
Sediment-air fluxes of PCBs and Hg will be measured using a chamber in which sediment and atmospheric characteristics such as moisture content, humidity, and temperature can be controlled. Using the "vignette" model to predict the flux of contaminants from sediments[7] will enable clarification of the soil and air side effects that control the magnitude of the contaminant fluxes. In this model the overall mass transfer resistance, R, which is inversely proportional to the contaminant flux N_a is assumed to depend on the apparent "resistance" on the air side of the interface, R_a, plus the "resistance" on the sediment side, R_s. The resistance on the air-side is equal to the reciprocal of the mass transfer coefficient, k_a on the air-side and the "resistance" on the sediment side is the reciprocal of k_s, the equivalent mass transfer of the soil side. Thus K_T, the overall mass transfer coefficient can be determined using the formula:

$$\frac{1}{K_T} = \frac{1}{k_s} + \frac{1}{k_a}$$

In our field experiments in Bayonne we have been concentrating our efforts on characterizing the highly variable air-side mass transfer while ignoring to some extent the processes on the sediment side. In the laboratory experiments the air-side mass transfer coefficient will be held constant and its value will be calculated using laminar boundary layer theory. The emphasis will shift to a determination of the sediment side mass transfer as the stabilization becomes effective through the curing of the concrete. In effect, there will be no attempt to replicate the airside field conditions; instead we are investigating the control imposed on the fluxes of PCB's and Hg through the changing resistance on the sediment side of the interface.

3.1 Experimental Set-up
The experiment will occur in a volatilization flux chamber as shown schematically in Fig. 3. It is planned to build the volatilization flux chamber with an exposure area of one square meter. Recent studies conducted by the Center for Environmental Engineering (Stevens Institute of Technology, Hoboken, NJ) at the OENJ-Bayonne site to determine PCB flux rates, have shown that that this area will provide sufficient PCB mass for analysis. The dimensions of the

volatilization chamber enclosure will be 2 meters long and 0.50 meter wide. The chamber height will be 0.60 meter. The depth of the deposited SDM will be 0.10 meter.

The average airflow rate will be 0.5 m³/min, which translates into a cross-sectionally averaged air velocity of 0.03 m/sec. Air flow rate will be varied within the operating range of the air blower to confirm the effect of Reynolds number on the mass transfer coefficient of PCB's and Hg between the SDM and air. The volatilization chamber will be equipped with a diffuser to ensure uniform airflow throughout the chamber. The inlet air moisture content will be controlled via a Drierite desiccant and a bubbler column, as shown in Fig. 3. The volatilization chamber will be insulated to minimize heat losses through the enclosure walls.
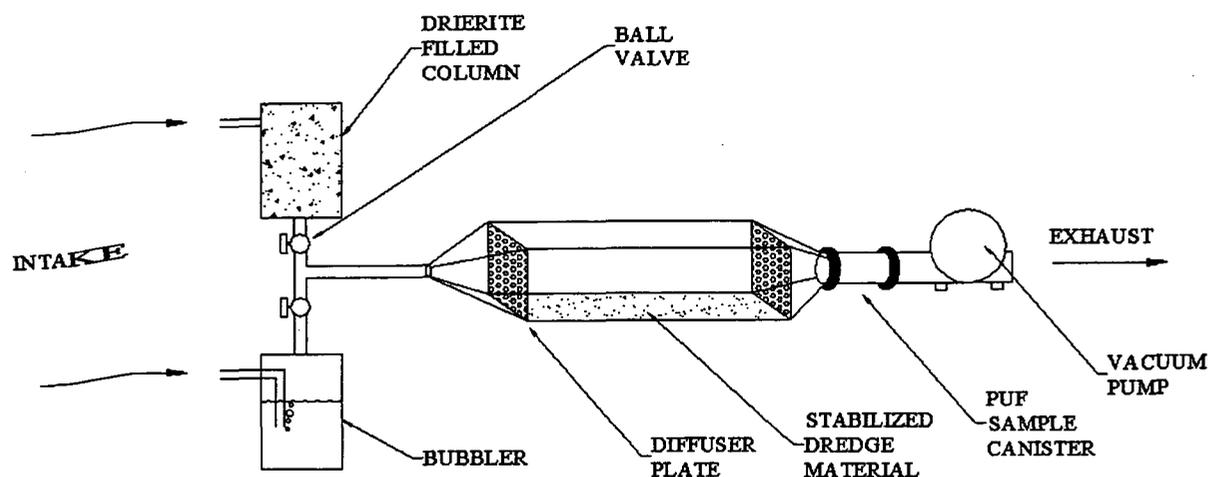
Figure 3. Schematic diagram of the PCBs volatilization flux chamber.

The effluent air from the chamber will be pulled through a modified Tisch Hi-Vol Poly-Urethane Filter (PUF) sampler, intended to adsorb PCB's contained in the exit air stream. Concentrations of total gaseous Hg (TGM, Hg° and reactive gaseous Hg species) will be measured in the influent and effluent air using the Tekran Model 2537A Continuous Ultra-Trace Mercury Vapour Analyzer (Tekran). This Hg monitoring system is capable of measuring TGM with a detection limit of < 0.1 ng m⁻³ at a rate of 1 measurement every 5 min. thereby providing a continuous record of Hg concentrations under all relevant air flow and temperature conditions. The Tekran continuous Hg analyzer features dual gold amalgam Hg traps for uninterrupted sample collection, high precision mass flow controllers, an internal Hg standard source, and cold vapor atomic fluorescence spectrometry (CVAFS) analysis. The Tekran dual sampling port solenoid will be used to alternate sample collection between inflow and outflow air of the flux chamber.

3.2 Experimental Methodology

Dredged sediment collected from the Bayonne OENJ site, will be transported to the Center for Environmental Engineering at Stevens Institute of Technology. The sediment will be taken, raw, from the site in 55-gallon drums. The sediment will then be weighed and added to a cement mixer. The appropriate amount of dry cement (6% or 12% cement by weight) will be weighed out and added to the cement mixer. The sediment has high water content, and as a result no water will be added to the mixture. The sediment and cement will be mixed for approximately 5 to 10 minutes. The mixture will then be poured into the flux chamber, it will be rolled out flat, and the chamber will be closed. Measurement of fluxes will commence immediately. The stabilized sediment will still have high moisture content; as a result it has a consistency that allows it to be poured into place. From our experience at the OENJ Bayonne site the SDM requires three or four days after placement to form a semi-rigid crust.

After SDM placement in the volatilization flux chamber, the enclosure will be properly sealed to ensure an airtight fit. Air will then be passed over the SDM surface at a controlled flow rate. Simultaneously, a lab blank will be collected to determine the PCB concentration in the ambient air. The influent air humidity will be adjusted by using an in-line bubbler to add moisture vapor as required by the experimental protocol. For dry-inlet air runs, the bubbler will be by-passed, and the air will be passed through a drierite-filled column. The air leaving the volatilization flux chamber will be directed through a PUF sampler to capture the PCBs present in the exiting air stream. The PCB volatilization flux [$N_{PCB}(t)$] through the chamber will be calculated by determining the total mass of PCB's captured by the PUF sampler in a given time interval using the equation:

$$N_{PCB}(t) \quad \frac{\Delta m}{A \cdot \Delta t} \tag{1}$$

where:

Δm = mass (ng) of compound collected in the PUF sampler over time Δt
Δt = time interval (hr)
A = area of the SDM-air interface (m^2)

The initial sampling interval for freshly placed sediment will be one hour. As the SDM ages the sediment side resistance will be increasing and the mass of PCBs volatilized in a specified time interval will decrease. Consequently, the sampling time interval might have to be increased to up to 12 hours as the SDM cures. Each run will comprise 7 to 8 sampling time intervals in series. The run will be interrupted a few minutes between two sampling time intervals for removal and insertion of the PUF into the air sampler. During the course of each run, the background PCB's and Hg concentration inside the laboratory will be determined.

The Hg vapor flux [$N_{Hg}(t)$] will be determined in much the same method, however since the Tekran will produce concentration values the flux will be determined using the equation:

$$N_{Hg}(t) = \frac{Q}{A}\int_0^T C(t)dt \qquad (2)$$

where

C(t)	=	concentration of Hg at time t (ng/m^3)
T	=	time interval (hr)
A	=	area of the SDM-air interface (m^2)
Q	=	flow rate through chamber (m^3/min)

During the course of the experimental runs, additional relevant environmental parameters affecting the rate of PCB and Hg volatilization will be continuously measured. These parameters include inlet and exit air temperatures and moisture content, and SDM temperature and moisture content. The moisture content of the air will be measured with a LI-COR 6262 CO$_2$/H$_2$O Gas Analyzer (LI-COR, Lincoln, Nebraska), and the SDM moisture content will be measured with a soil moisture probe. The spatially averaged SDM temperature will be measured with a TCAV (thermocouple averaging) probe. The gathered data will be stored in a Campbell 23X Data logger (Campbell Scientific Inc. Logan, UT). To validate the SDM moisture probe data, SDM samples (one at the beginning of the test run, and one at the end of the test run) will be analyzed for moisture content using ASTM protocol.

SDM samples will be taken at the beginning and the end of each test run for the determination of the PCB and Hg concentrations. SDM samples for PCB and total Hg analysis will be stored frozen in polyethylene bags until analysis. The SDM samples will also be analyzed for fractions of silt, sand, and clay by sieve analysis, bulk soil density, particle density, total porosity, air filled porosity, total organic carbon, water content, and temperature. These properties will be determined prior to and after stabilization.

The PCB's adsorbed onto the PUFs adsorption media or in the SDM samples will be extracted at 100°C and 1500 psi (US EPA Method 3545) with dichloromethane using a Dionex ASE 300 Accelerated Solvent Extractor (Dionex Corporation, Marlton, NJ). The resulting extract will be concentrated using a Rotary Evaporator, and then filtered via Supelclean LC-Florisil SPE Tubes. At the end of the filtration process, the filtration equipment will be rinsed with hexane to ensure that none of the extract is retained in the Florisil tubes. Finally, the filtered extract will be concentrated using a nitrogen evaporator (Organomation Associates, Inc. Berlin, MA).

The concentrated extract will be analyzed for PCB's, using a Dual Column Gas Chromatograph equipped with an Electron Capture Detector (GC/ECD), at the Center for Environmental Engineering. The list of PCB congers to be analyzed can be found in Appendix 1. The quality control procedures for PCB analysis include matrix spike, blank spike, field blank, and extraction blank preparation and analysis. The operating procedures were developed in the Center for Environmental Engineering (CEE) and based on EPA Method 8082. Prior to initiation of the experiments, an equipment blank will be collected in order to determine adsorption to the walls of the enclosure. This will be completed by taking advantage of the fact that there are ample PCBs in the ambient air in our region. Two air samples will be taken simultaneously, the first, an ambient air sample, the second, will be ambient air pulled through the empty flux chamber.

The two PCB sample masses collected on the PUFs will then be compared to determine if there is a sink of PCBs in the chamber.

Total Hg in SDM samples will be measured by cold vapor atomic fluorescence spectrometry (CVAFS, Bloom and Fitzgerald, 1988; U.S. EPA, 1995). Prior to analysis, SDM samples will be digested in concentrated $HNO_3:H_2SO_4$ (2:1) at 95°C in acid-cleaned Teflon vials followed by oxidation with 5% $KMnO_4$ (Balogh et al., 1999) in accordance with USEPA Method 7470A. Excess oxidant will be neutralized by addition of hydroxylamine hydrochloride and Hg^{2+} will be reduced to Hg° with $SnCl_2$ in gas scrubbing bottles. Hg° will be collected on gold traps and analyzed using a Tekran 2500 CVAFS mercury detector.

Recent studies of atmospheric Hg (Lindberg and Stratton, 1998; Stratton and Lindberg, 2000; Sheu and Mason, 2001) have identified gaseous forms of Hg(II), which may include HgO, $HgCl_2$, $HgBr_2$, and $Hg(NO_3)_2 \cdot H_2O$ and are collectively referred to as reactive gaseous mercury (RGM). RGM typically accounts for 2% to 5% of the total gaseous Hg, but can reach as high as 30% under certain circumstances. RGM is thought to be formed in the atmosphere as a result of reactions with photochemical oxidants (Lindberg and Stratton, 1998; Sheu and Mason, 2001) and is also emitted from combustion point sources. As reactive gases, RGM species are expected to have relatively high deposition velocities and short atmospheric residence times.

In order to evaluate the contribution of RGM to the total Hg vapor emissions from stabilized dredge material, we will quantify RGM in the outflow of the flux chamber. RGM will be collected over two hours using an annular KCl denuder (Xiao et al., 1997; Sheu and Mason, 2001) placed in-line between the flux chamber and the Tekran continuous Hg analyzer. Gas phase elemental Hg in the effluent of the denuder will be monitored continuously during RGM collection for mass balance assessment. RGM in influent chamber air, which is expected to be negligible, will also be determined. After sampling, the denuder will be heated at 500°C for 45 min to reduce all collected RGM to Hg° which will be collected on gold-coated sand under Ar gas. The trapped Hg will then be analyzed by CVAFS.

Eight sampling runs are proposed consisting of 7-8 sampling intervals. Environmental factors will be varied over the different runs. The schedule will be as follows:

Run #	Influent Humidity (%)	% Cement (by weight)	Airflow (m^3/min)
1	100	12	0.5
2	100	6	0.5
3	0	12	0.5
4	0	6	0.5
5	50	6	0.5
6	50	12	0.5
7	50	12	<0.5
8	50	12	>0.5

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3.3 Determination of the sediment-side mass transfer coefficient

The objective of the present study is to determine the sediment-side mass transfer coefficient for volatilization of PCBs and Hg from SDM exposed to the atmosphere. Using the "Vingnette" model presented earlier we will break down the factors that control the flux. Since our previous study investigated mainly the air-side mass transfer, this study will hold the air-side mass transfer constant using boundary layer theory for laminar flow. The air-side mass transfer coefficient will be function of the flow rate, which is known, and the diffusivity, which can be estimated from the literature. Using this value we will determine the k_s and the characteristics of the sediment and atmosphere that affect it.

The mass transfer phenomenon takes place within a hypothetical boundary layer that exists at the SDM-Atmospheric interface as illustrated in Fig. 4. Experimentally, it is not possible to measure the species concentration at the SDM-atmosphere interface and therefore, it is convenient to define the overall mass transfer coefficient between the bulk SDM phase and the bulk air phase for which the concentrations can be readily measured. Transport will occur down a gradient that exists from high sediment concentrations to relatively low air concentrations. The following is a derivation of the air-side mass transfer coefficient for the laminar flow regime that will be present in our flux chamber.
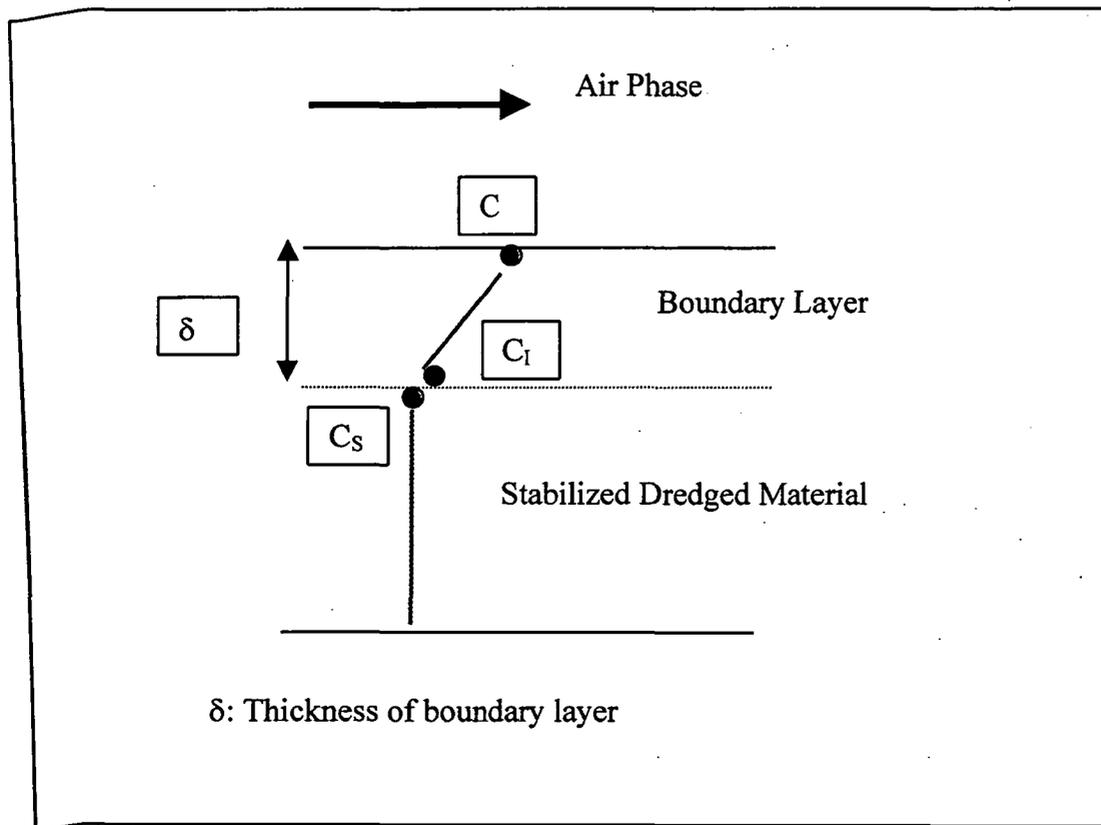

Air Phase

C

δ

Boundary Layer

C_I

C_S

Stabilized Dredged Material

δ: Thickness of boundary layer

Figure 4. Schematic of the stagnant boundary layer film model at location x along the volatilization test chamber.

The overall mass transfer flux, perpendicular to the interface, is given by:

$$N_i \quad \cdot \quad K_{Ti} \, (C_s - C_b)$$

(3)

where N_i is the flux of the species i, K_{Ti} is the overall mass transfer coefficient for the species i across the boundary layer and C_s and C_b are the concentrations of the species i in the sediment and in the bulk air phase respectively. The reciprocal of the overall mass transfer coefficient is the sum of the reciprocals of the mass transfer coefficient for the sediment side and the air-side, i.e.,

$$\frac{1}{K_T} = \frac{1}{k_s} + \frac{1}{k_a}$$

(4)

Determination of k_a is possible using boundary layer theory. Since the PCBs and Hg are composed of sparingly soluble species, the SDM system can be simulated as a sparingly dissolving flat slab over which air flows. The length of the volatilization chamber and the velocity of the flowing air are such that the flow system can be considered laminar. When dealing with flow over a flat plate, the flow regime is considered to be laminar (Schichting, H., 1968) as long as the Reynolds number is less than 300,000. The Reynolds number for this case is defined as:

$$R_e \quad = \quad \frac{\rho \, v^0 \, x}{\mu}$$

(5)

The following theoretical relationship for the mass transfer coefficient (k_a) can be derived for laminar flow of a fluid over a sparingly soluble flat plate (Welty, et al., 1976).

$$\frac{k_a \, x}{D} \quad = \quad 0.323 \left(\frac{\rho \, v^0 \, x}{\mu} \right)^{\frac{1}{2}} \left(\frac{\mu}{\rho \, D} \right)^{\frac{1}{3}}$$

(6)

where:

k_a local mass transfer coefficient at position x
x   position along x-axis
D   Molecular diffusion coefficient of species being transferred
ρ Flowing fluid density
μ Flowing fluid dynamic viscosity
v^0 Flowing fluid average velocity

$\dfrac{\mu}{\rho \, D}$ = Schmidt number

The mass transfer coefficient (k_a) given by Equation 6 is a local mass transfer coefficient since it is a function of the x position along the x-axis. This x dependence is expected because the aerodynamic and concentration boundary layers thicknesses vary with x. Since our material

balance is applied to the total mass of the SDM inside the volatilization test chamber (Equation 1), the mass transfer coefficient k_{ma} is different from the local mass transfer coefficient k_a. Here, k_{ma} is length-averaged mass transfer coefficient defined as:

$$k_{ma} = \frac{1}{L} \int_0^L k_i \, dx \qquad (7)$$

where L is the length of the SDM pool in the volatilization chamber.

Substitution of Equation (6) into Equation (7) yields the following expression for k_{ma}

$$\frac{k_{ma} L}{D} = 0.646 \left(\frac{\rho \, v^0 \, L}{\mu} \right)^{\frac{1}{2}} \left(\frac{\mu}{\rho D} \right)^{\frac{1}{3}} \qquad (8)$$

Equation (8) indicates that the length-averaged mass transfer coefficient k_{ma} is proportional to $Re^{1/2}$ and $Sc^{1/3}$.

Since the flux values and the concentration values in the air and sediment will be directly measured, then, by using equation (3), we can calculate the value for K_t, the overall transfer coefficient. Since k_{ma} will be calculated from equation (8) and will be the air-side mass transfer coefficient, then the sediment side mass transfer coefficient, k_s, will be determined from equation (4). For each experimental run there will be a determination of the time history of k_s. The dependence of the sediment side mass transfer coefficient on sediment properties, most importantly the soil moisture content, will be evaluated with the aim of establishing a predictive model.

3.4 Flux Chamber Sampling Schedule

Eight experimental runs will be carried out. During these runs, the effect of air humidity and Reynolds number on the volatilization rate and mass transfer coefficient will be assessed. The number of samples to be analyzed for PCBs will be as follows:

Sample ID	Number of runs	Samples per run	Total Samples
PUF (from experimental set-up)	8	10	80
PUF (lab blanks)	8	3	24
PUF (laboratory extraction equipment blanks)	8	1	8
Sediment	8	2	16
TOTAL			128

4.0 APPROACH TO ADDRESS GAS-PARTICLE EXCHANGE

4.1 Gas/particle partitioning

The distribution of semi-volatile compounds between the gas and particle-bound phases is the most important factor determining their removal mechanisms and residence times in the atmosphere (Junge, 1977; Eisenreich et al., 1981; Pankow, 1987; Pankow and Bidleman, 1992; Cousins and Mackay, 2001). Compounds which partition to particles are rapidly removed from the atmosphere via dry and wet deposition, while gas-phase compounds will remain in the atmosphere much longer and be transported over greater distances. In addition, small particles are more efficiently entrained in the human lung, although the extent to which PCBs sorbed to such particles may affect human health is unknown.

The theory regarding the adsorption and absorption of semivolatile organic compounds (SOCs) onto and into the atmospheric particle matrix has been reported (Junge, 1977; Pankow, 1987, 1991, 1992, and 1994; Pankow and Bidleman, 1992; Finizio et al., 1997; Jang and Kamens, 1998; Cousins and Mackay, 2001; Dachs and Eisenreich, 2001) and has been examined in natural and urban environments for a wide range of SOCs (e.g., Simcik et al., 1998; Lohmann et al., 2000; Harner and Bidleman, 1998). In gas-particle partitioning, the equilibrium partitioning coefficient (K_P), describes the ratio between the particle phase concentration (C_P, pg/m^3) and the gas phase concentration (C_G, pg/m^3) of the SOC, normalized by the TSP concentration (μg/m^3) (Pankow, 1987 and 1994; Harner and Bidleman 1998):

$$K_P \cdot \frac{C_P}{C_G(TSP)} \tag{9}$$

Higher molecular weight compounds having lower vapor pressures will sorb to a greater extent to particles than low molecular weight compounds. Thus values of K_P (m^3/μg) calculated for individual SOCs may be regressed against a chemical-physical property of the SOC such as vapor pressure. Slopes of the regressions should approach -1 at equilibrium but can be affected by large changes in environmental conditions, particle characteristics, or SOC concentrations during sampling (Pankow, 1991 and 1992; Pankow and Bidleman 1992; Finizio et al., 1997; Dachs and Eisenreich, 2001). Finizio et al. (1997) supports applying the log of the octanol-air partitioning coefficient (K_{OA}) as a correlating variable with log K_P:

$$\log K_P \cdot m_r \log K_{OA} \cdot b_r \tag{10}$$

where m_r and b_r are the slope and intercept of least squares regression, respectively. In this case, the slope should approach $+1$ for the equilibrium situation. Recent measurements of K_{OA} and its dependence on temperature for PCB congeners have made an investigation of gas-particle partitioning by this method possible. Harner and Bidleman (1996) have reported direct measurements of K_{OA} for 19 PCB congeners. Zhang et al. (1999) expanded the list of available K_{OA} values to include over 100 congeners using a thermodynamic gas chromatographic approach.

Gas-particle partitioning is most easily studied in environments where PCB concentrations in the particle phase are sufficiently high. Bayonne is thus ideally suited to such a study, as NJADN data has indicated that concentrations of PCBs in both the aerosol and gas phase are elevated in this region, even prior to the land application of Hudson River sediment. We hypothesize that gas/particle partition of PCBs will be far from equilibrium (i.e. slopes much less than 1) in

samples collected at the sediment disposal site due to gas-phase emissions of PCBs. Partitioning may approach equilibrium (with slopes approaching 1) at the trailer site, as air masses generated at the sediment disposal site will undergo partitioning during their travel time. Particle-phase PCBs frequently comprise 10% or more of the total atmospheric burden, which may account for much of the decrease in gas-phase concentrations of PCBs observed at the trailer site.

Because the particle phase was routinely collected on quartz fiber filters during air sampling, no additional fieldwork will be required. Money is requested for laboratory supplies needed to conduct the extraction and analysis of the filters (most of which will occur in year 1), and for salary for the technician (0.3 FTE) to conduct the analysis and manage the data.

4.2 Analysis of particulate PCBs

Methods for the quantitation of particle-phase PCBs are well established in the PI's laboratory. Surrogate standards (10 or 25 ng) containing 3,5-dichlorobiphenyl (IUPAC # 14), 2,3,5,6-tetrachlorobiphenyl (IUPAC # 65) and 2,3,4,4',5,6-hexachlorobiphenyl (IUPAC # 166) will be added to the QFFs, which will then be extracted in a Soxhlet apparatus with 4:1 (v/v) mixture of petroleum ether and dichloromethane for 24 hours. The extracts will be concentrated in a Buchi rotavap apparatus where solvent will be exchanged to hexane and cleaned up and fractionated on a 4 g 3 % H_2O-deactivated) alumina column. The PCB fraction (1st fraction – hexane as solvent) will be eluted and then concentrated to about 1mL. Details of a similar procedure may be found in Simcik et al. (1997) and Brunciak et al. (2001). The internal standards used in this study will be 2,3,5,6-tetrachlorobiphenyl (IUPAC # 30) and 2,2',3,4,4',5,6,6'-octachlorobiphenyl (IUPAC # 204). The final extract will be concentrated to 0.1 to 0.2 ml under N_2 and analyzed on a HP 5890 gas chromatograph (GC) equipped with a ^{63}Ni electron capture detector (ECD), a HP autosampler, and a HP 3365 ChemStation on a 60 m 5% DB-5 capillary column (0.32 mm id.) with 0.25 μm film thickness (J & W Scientific). Concentrations of the individual PCB congeners were calculated using the internal standard method.

4.3 Analysis of particulate Hg

An attempt will be made to determine total particulate Hg concentrations using portions of the quartz fiber filters that have been collected for particulate PCB analysis. Filters will be divided using a stainless steel scalpel to give two halves of equal aerosol collection area. The filter half for Hg analysis will be further divided into equal sections and digested with 2 ml concentrated nitric and hydrofluoric acids (95:5, HNO_3:HF) in Teflon vials for 6 h at 100°C. Digested particles will be diluted with 150 ml ultrapure water and total Hg concentrations measured by $SnCl_2$ reduction and cold vapor atomic fluorescence spectrometry (CVAFS; Bloom and Fitzgerald, 1988). Analytical accuracy will be established with measurements of gaseous Hg° injected directly into the analytical gas stream and trapped on the analytical gold column to yield standard curves with r^2 values of ≥ 0.999. The efficiency of aqueous Hg reduction and trapping (100%) will be checked through the analysis of aqueous $HgCl_2$ solutions.

5.0 APPROACH TO TRANSPORT MODELING
5.1 RAMS/HYPACT Modeling of PCB transport
Fine-scale transport and deposition of atmospheric tracers is highly dependent on the local circulation, terrain, and precipitation distribution. Coarse input meteorological information often causes a major uncertainty in calculations of fine-scale distribution of atmospheric pollutants especially in the vicinity of the sources like one discussed in this study. Unfortunately fine-scale

meteorological fields (with spatial resolution of hundreds of meters) are not available either from the observations, or from the operational forecast models. Here we will employ the Regional Atmospheric Modeling System (RAMS), to downscale available meteorological information and to produce fine-scale local circulation and precipitation over the Bayonne peninsula for the periods of observations. These meteorological fields will be used for fine grid transport and deposition calculations using the of-line Hybrid Particle And Concentration Transport model (HYPACT). This approach will provide a better estimate of local transport and deposition of PCBs from the sediment application site at the Bayonne peninsula.

5.2 Calculation of meteorological fields

RAMS is a state-of-the-art nonhydrostatic regional model with well developed cloud microphysics and surface interaction parameterizations (Pielke et al., 1992). It predicts the three velocity fields, temperature, water vapor mixing ratios, pressure, subgrid-scale turbulent kinetic energy, and several types of cloud hydrometeors including cloud water, ice, graupel, and snow.

Grid nesting is used in RAMS to provide high spatial resolution in selected locations, while covering a large domain at lower resolution. Therefore, effects of a large-scale circulation patterns can be transferred to an internal fine resolution region. A nested grid occupies a region within the computational domain of its coarser parent grid. Any number of nested grids may be used. The two-way interactions between nested grids are calculated following Clark and Farley (1984), Clark and Hall (1991), and Walko et al. (1995).

Various plug-compatible parameterization modules are available for most physical processes, including radiation, turbulence, and land system. Vertical and horizontal turbulent eddy mixing is parameterized using the 2.5 level scheme of Mellor and Yamada (1974, 1982) when resolution is relatively coarse and parameterized convection performs most of the vertical transport. The 1.5-order-of-closure scheme of Deardorff (1980) is used when convection is explicitly resolved. RAMS accounts for 12 soil textural classes and 18 vegetation types. The patchy land-atmosphere interactive dynamics scheme developed by Avissar and Mahrer (1988) and Avissar and Pielke (1989) was adopted to evaluate the land surface energy, mass, and momentum fluxes in the atmospheric surface layer as well as heat and water flux in the ground. The radiative schemes of Chen and Cotton (1987) or Mahrer and Pielke (1977) can be used for radiative transport.

Modified versions of Kuo (1974) and Fritsch and Chappell (1980) convective parameterizations are implemented into RAMS (Tremback, 1990). For cloud-resolving calculations, RAMS does not require any convective parameterization.

The cloud microphysics scheme is based on Tripoli and Cotton (1980, 1982) and Cotton et al. (1982). This scheme consists of a set of conservation equations for water vapor and six hydrometeors: cloud droplets, raindrops, pristine ice, snow, graupel, and aggregates. Their tendencies are affected by advection, turbulence, and microphysical transformations in size distribution and from one class to another.

RAMS has the ability to combine and blend several input data sets in the data analysis as well as allowing a 4-D data assimilation regime of calculations. In this study we plan to initiate RAMS calculations using "coarse" resolution fields from the operational analysis produced by the best

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operational forecast model provided by the National Center for Environmental Predictions (NCEP).

5.3 Calculation of pollutant transport

HYPACT is a state-of-the-art transport model that calculates temporal and spatial distributions of atmospheric pollutants using 3-D time dependent wind and turbulence fields. It allows assessment of the impact of one or multiple sources emitted into highly complex local weather regimes, including complex terrain flows, land/sea breezes, urban areas. Species can include gases and a spectrum of aerosol sizes. Source geometry can include point, line, area, and volume sources of various orientations. HYPACT is driven by wind and potential temperature fields simulated in the RAMS. The turbulent characteristics are calculated diagnostically from available meteorological information using turbulent models developed in (Mellor and Yamada, 1974, 1982; Deardorff, 1980). The model is capable of accounting for deposition processes, chemical or radiological decay, and gas-to-particle conversion. Transport of heavy particles accounts for the effect of gravitational forces on the mean particle motion. The interaction of the particle with the ground surface is parameterized following Boughton et al. (1987). Above a certain height h, the probability of particle deposition is negligible. If the particle comes below this height, the probability that the particle is absorbed is computed from the transition probability density given by Monin (1959).

In order to accurately assess the transport of contaminants in this model, it is essential that both gas and particle phases be considered. Initially we anticipate entering contaminant data into the HYPACT model as a sum (gas + particle) of atmospheric PCBs or Hg. In later model runs, gas and particle phase contaminants will be treated separately, with parameterization for partitioning of gas-phase compounds onto aerosols. This represents an additional justification for quantifying PCBs (and Hg?) in the particle phase. Using the concentration data as input, the model will estimate the contaminant flux from the sediments necessary to produce the observed concentrations of PCBs and Hg at the sediment application and trailer sites.

5.4 Transport Model Applications

RAMS in combination with HYPACT or similar chemical transport model has been used in numerous applications including recent studies of the sulfur cycle and acid deposition in East Asia (Carmichael et al., 2002), and calculations of chemical production of tropospheric ozone (Varinou et al., 1999; Fast et al., 2000; Galanter et al., 2000).

6.0 PROJECT TIMELINE

Month / Task	02	04	06	08	10	12	14	16	18	20	22	24
Flux chamber design, construction, and preliminary testing	■											
Flux chamber experiments		■	■	■								
Flux chamber sample analysis & quantitation			■	■	■							
Quantitation of PCBs and Hg in particles	■	■	■	■	■	■	■	■	■			
Purchase & installation of computing equipment	■											
Transport model calibration		■	■	■						■	■	
Transport model runs							■	■	■	■	■	■
Reports				■		■			■			■

7.0 DELIVERABLES
Progress reports will be submitted at the ends of the 6th, 12th, and 18th months of the project and a final report will be submitted within two months after the end of the project.

8.0 PERSONNEL
Dr. George P Korfiatis has over 18 years experience in research and consulting and has participated in over 60 large-scale research projects funded by government, and the private sector. His expertise lies in the area of fate and transport of contaminants in soils and sediments, investigation of contaminate soil, water, and sediment. He has published over 75 papers in the open literature on these topics.

Dr. Richard I. Hires is a professor with over 25 years experience in filed investigations of ocean, estuarine, and water atmosphere dynamics and transport processes. Dr. Hires has worked extensively with consultants in these areas.

Dr. Tsan Liang Su is a Research Assistant Professor with expertise in the field of Analytical Chemistry. He will be responsible for development of laboratory methods, and oversight of extraction and analysis of PCB samples.

Robert Miskewitz is a Research Engineer who has conducted field measurements of PCB fluxes from dredged sediment and performed laboratory analysis of the air samples for the Bayonne project. He will be responsible for design and operation of the flux chamber as well as data collection and analysis.
Dr. Lisa Totten is an assistant professor of environmental organic chemistry and has extensive experience in environmental monitoring of PCBs and other semi-volatile organic contaminants.

She will be responsible for overseeing analysis of particle-phase PCBs, and for compiling PCB data for input into the RAMS model. She will also assist in model development.

Dr. John Reinfelder is professor of environmental inorganic chemistry, is an expert on Hg cycling, and has been monitoring ambient Hg concentrations for the Bayonne project and for the New Jersey Atmospheric Deposition Network. He will be responsible for Hg analysis in conjunction with the flux chamber studies and in particle samples from the sediment disposal and trailer sites, and for compiling Hg data for input to the RAMS model.

Dr. Georgiy Stenchikov is a professor of Environmental Sciences and an expert in climate modeling and climate change. He will oversee compilation and calibration of the RAMS/HYPACT model.

Cari Gigliotti is a research staff scientist with more than 5 years experience in environmental measurement of PCBs. She has overseen the monitoring of PCB concentrations at Bayonne since the inception of the project, and personally collected all of the samples to be analyzed in the proposed work. She will conduct the analysis of particle-phase PCBs and assist in the compilation of the data for input into the model.

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EMISSIONS AND ATMOSPHERIC TRANSPORT OF PCBs AND Hg FROM STABILIZED HARBOR SEDIMENTS

Stevens Institute of Technology

		Year 1	Total
A. Labor			
Project Manager (G Korfiatis)	0.25 Month	$3,560	$3,560
Investigators			
Hires	0.5 Month	$5,955	$5,955
Su	2.0 months	$11,766	$11,766
Sidhoum	2.0 months	$14,664	$14,664
Miskewitz	6.0 months	$22,050	$22,050
Total Labor		$57,995	$57,995
Fringe Benefits @ 30%		$17,399	$17,399
Total Labor & Benefits		$75,394	$75,394
B. Materials			
Material for Volatilization Chamber		$2,500	$2,500
Thermocouples		$500	$500
Calibration of Datalogger and Soil Moisture Probe		$200	$200
Air Sampler Calibration		$150	$150
Drums		$300	$300
A6. LI-COR Water Vapor Sensor		$3,000	$3,000
Mixer		$800	$800
Expendables		$500	$500
MaterialsTotal		$7,950	$7,950
C. Laboratory Analyses			
C1. PUF Samples [112 x $40 per sample]		$4,480	$4,480
C2. PUF Sample Analysis			
Quartz Filter [112 x $5 per sample]		$560	$560
Extraction (SIT) [128 x $50 per sample]		$6,400	$6,400
Analysis (Rutgers) [128x $150 per sample]		$19,200	$19,200
Analysis Total		$30,640	$30,640
D. Machine Shop Services		$4,000	$4,000
E. TOTAL DIRECT COSTS		$117,984	$117,984
F. Overhead			
On A+B+C+D at 10%		$11,798	$11,798
G. NJMSC Administration		$10,383	$10,383
H. Total Project Cost (E+F) STEVENS		$140,165	$140,165

EMISSIONS AND ATMOSPHERIC TRANSPORT OF PCBs AND Hg FROM STABILIZED HARBOR SEDIMENTS

Rutgers University

	Year 1	Year 2	Total
A. Salaries			
Dr. Totten (0.2 FTE)	$11,000	$11,000	$22,000
Dr. Reinfelder (1 month)	$7,082	$7,082	$14,164
Dr. Stenchikov (1 month)	$10,171	$10,171	$20,342
Laboratory technician (0.3 FTE)	$14,314	$6,159	$20,473
Graduate Students (1.5)	$24,453	$16,302	$40,755
Total Salaries	**$67,020**	**$50,714**	**$117,734**
B. Fringe	**$13,678**	**$11,740**	**$25,418**
C. Equipment			
Cluster computer	$25,000		$25,000
D. Travel	**$3,000**	**$3,000**	**$6,000**
E. Other Direct Costs			
1. Chemical & Laboratory Supplies	$6,000	$3,000	$9,000
2. Computer services fee	$1,016	$1,016	$2,032
3. Publication services		$1,000	$1,000
4. Other services	$1,000	$1,000	$2,000
5. Tuition	— $9,990	$4,994	$14,984
Total Other Costs	**$18,006**	**$11,010**	**$29,016**
F. Total Direct Costs	**$126,704**	**$76,464**	**$203,168**
G. Indirect Costs			
(10% of F minus C and E5)	$9,171	$7,147	$16,318
H. NJMSC Administration	**$10,870**	**$6,689**	**$17,559**
I. Total Project Costs	**$146,745**	**$90,300**	**$237,045**

$286,910