Department of Natural Resources MARYLAND GEOLOGICAL SURVEY Jeffery P. Halka, Director

## **COASTAL ENVIRONMENTAL GEOSCIENCES PROGRAM FILE REPORT NO. 11-05**

# **Deep Creek Lake Sediment Study: Physical and Chemical Characteristics of Lake Sediments**

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Prepared For **Department of Natural Resources** Maryland Park Service

> **September, 2011 Revised October, 2011**

## **Deep Creek Lake Sediment Study: Physical and Chemical Characteristics of Lake Sediments**

### **EXECUTIVE SUMMARY**

The State of Maryland, Department of Natural Resources (DNR) recently added Deep Creek Lake to its public land holdings. Increased development of the surrounding land and a growing public concern over lake sedimentation has prompted a detailed examination of this resource. While Deep Creek Lake appears generally healthy based on existing water quality data, there are gaps in data, particularly with regard to sediments. Additional information is needed on the bottom sediments contained within this system, capacity of the lake itself, and identification, where possible, of the impacts of changing land use patterns on sedimentation and sediment character within the lake

In order to characterize the bottom sediments in Deep Creek Lake, the Maryland Geological Survey (MGS), a program within DNR's Resource Assessment Service, collected surficial sediments at 50 locations throughout the lake. The sediments were analyzed for textural properties, total nitrogen (N), total carbon (C), reactive carbon  $(C_R)$ , total phosphorus (P), and 48 additional elements including arsenic (As), antimony (Sb), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), nickel (Ni), lead (Pb), sulfur (S), zinc (Zn). These data provide baseline information on the bottom sediments.

Based on the textural analyses of the 50 surficial sediment samples (representing the top 5 cm of the sediment column), the majority of the samples collected are fine-grained sediments, with an average textural content of 18% sand, 39% silt and 43% clay. Clay represents a major component of the collected sediments, which is not unexpected given the abundance of shale in the underlying formations within the watershed. Sediments with the highest clay content were collected in the deepest part of the lake. Sand is a relatively minor component with only four samples classified as sand and silty-sand. The sand sample is the only sample to contain appreciable amounts of gravel (i.e., particles with diameter> 2mm). This sample was collected in the upstream reach of Cherry Branch.

Average N, C, and P in Deep Creek Lake sediments fall within the range of those measured in other Maryland freshwater lakes. On average, 70% of the total C contained in Deep Creek Lake sediment is reactive, readily available to the biological community. Coarser sediments (*i.e.,* low clay content) tend to contain a lower portion of reactive C. Sediments in the northern portion of the lake contain overall higher C content (both total and reactive C) compared to the southern end. There is less variation in the non-reactive C content with regard to distribution.

Total C has little correlation with grain size; the poor correlation due to inclusion of nonreactive C which has no association with any particular sediment type. However, reactive C has higher correlation with clay as well as with N and P (compared to total C), indicating that a significant portion of the reactive carbon in the sediment comes from primary productivity (plankton and algae blooms). Total N has the highest correlation with reactive C indicating that

most N is associated with organic material, most likely from primary productivity (algae). Total P is associated with the clay content of the sediment as well as many of the metals and S. When comparing the relative amounts of C, N and P in the Deep Creek Lake sediments to those of dried algae, P appears to be the limiting nutrient. In other words, mean C:P and N:P ratios are greater than those ratios of dried algae.

Compared to other freshwater lakes, S is significantly higher in most of the Deep Creek Lake sediments, particularly those collected in the deepest area at the downstream end (north end). The very dark color (black and dark grey) of some sediments collected indicated the presence of S, in the form of mono-sulfides (as FeS). Sources of S include sulfates from acid mine drainage, and atmospheric deposition in the watershed. Reduced S and sulfate  $(SO<sub>4</sub><sup>-2</sup>)$ concentration is an extremely important variable controlling P release from sediments. The increased P release from sediments at higher sulfate concentrations may help explain why primary production in freshwater systems (with relatively low S concentrations) tends to be P limited, whereas in many saline systems (with high sulfate concentrations) production is often P sufficient. Sulfur also plays an important role in arsenic cycling, which explains the high correlations between S, As, and Fe seen in this study.

Concentration and enrichment of most metals in Deep Creek Lake sediments are within normal range given the geology of the watershed. However, the sediments are significantly enriched in As, Cd, Cs, Hf, Pb, Sb, and Zn, with respect to average continental crust rock. The enrichments are higher than those reported for New Germany Lake, which is located in the same physiographic and atmospheric deposition regions and, thus is expected to be similar in geochemistry. The higher enrichments, particularly As, Cr, and Sb, in Deep Creek Lake sediments are attributed to contribution from coal deposits within the lake's watershed. Coal deposits are generally enriched with these metals as well as other rare earth elements. Acid mine drainage processes most likely mobilized these elements, resulting in higher concentration compared to New Germany sediments. It should be noted that sediments are enriched in Pb in all of the freshwater lakes studied in Maryland, illustrating the widespread anthropogenic sources for Pb. Nevertheless, Pb concentrations in Deep Creek Lake sediments are low as to not be a threat to the benthic community.

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### **INTRODUCTION**

The State of Maryland, Department of Natural Resources (DNR) recently added Deep Creek Lake to its public land holdings. Increased development of the surrounding land and a growing public concern over lake sedimentation has prompted a detailed examination of this resource. While Deep Creek Lake appears generally healthy based on existing water quality data, there are gaps in data, particularly with regard to sediments (Kesley and Powell, 2011). Additional information is needed on the bottom sediments contained within this system, capacity of the lake itself, and identification, where possible, of the impacts of changing land use patterns on sedimentation and sediment character within the lake

Sediments accumulating on the lake bottom act as reservoir for nutrients and contaminants, including toxic metals and organic pollutants. The physical and chemical properties of sediments are a controlling factor in the absorption and release of nutrients and contaminants. Documentation of these characteristic and the concentrations of any existing contaminants are necessary for the effective management of Deep Creek Lake.

#### **Objectives**

In order to characterize the bottom sediment in Deep Creek Lake, the Maryland Geological Survey (MGS), a program within DNR's Resource Assessment Service, collected sediments within the lake and analyzed them for physical and chemical properties. The results of these analyses are presented in this report.

#### **Study Area**

Deep Creek Lake is located in Garrett County. The lake was formed in 1925 when the Youghiogheny Hydroelectric Company constructed a dam across Deep Creek. The lake is presently owned and managed by Maryland Department of Natural Resources.

The Deep Creek Lake watershed is located within the Appalachian Plateau Physiographic Region of Maryland. The bedrock of this region consists principally of gently folded sedimentary rock comprised of shale, siltstone, and sandstone of mixed marine and non-marine origins. Folding has produced elongated arches, or anticlines, trending NE to SW across the region that expose the oldest formations at the surface. In the intervening synclinal basins, coal-bearing strata of Pennsylvanian and Permian ages are preserved. The northern half of Deep Creek Lake is located on broad syncline, called the Casselman Basin. Meadow Mountain is the eastern border of this structure. The lake perimeter is steep within this structure. The rock exposed here are brown colored sandstones and shales of a Mississippian age formation called the Mauch Chunk. Within the State Park, the 200 to 300-foot thick Greenbrier Limestone underlies the lake, contributing calcium carbonate to the water. Calcium carbonate may buffer the lake from acidic runoff from adjacent coal deposits. The Cherry Branch tributary drains the coal bearing formations and is thought to contribute a significant portion of the acidity to the lake (MDE, 2002).

The dam and immediately adjacent areas lie within the Upper Youghiogheny coal basin. Here sandstones and shales of the Allegheny /Pottsville formation of Lower Pennsylvanian age (325 million years old) are exposed. Some lower coal beds may also be exposed.

The southern half of the lake lies within the Deer Park Anticline composed of the (1) brown colored sandstones and shales of the Pocono Formation of Lower Mississippian age (350 million years old) then (2) further southeast, red to reddish brown sandstones and shales of the Hampshire Formation of Upper Devonian age (365 million years old) and finally (3) Devonian series of formations, comprise of predominately greywacke, siltstone and shale, sandstones and conglomerates. Unlike the northern half of the lake, the topography along the perimeter of the lake within the anticline structure is flatter and gentler.

## **Other Studies**

In 2007 and 2008, the United States Geological Survey (USGS) collected 34 sediment cores from Deep Creek Lake to determine the amount of sediment accumulation since the lake was established. The cores were collected in areas where sediment accumulation was expected to be the highest: in coves and mouths of streams draining into the lake. However, the USGS did not collect any cores in the deepist areas of the lake due to limitation of their coring equipment. The USGS analyzed seven cores for grain size and five cores for <sup>137</sup>Cs activity to determine sedimentation rates. USGS results showed variable sedimentation rates over the the 83 year history of Deep Creek Lake, with the higher average sedimentation rates occurring early after the lake was contructed (between 1925 and 1963). (Banks et al., 2010).

Concurrent with this study, a reconnaissance study of sediment accumulation in ten selected coves within Deep Creek Lake was conducted. The results of that study are presented in a separate report (Ortt and Manship, 2011).

#### **METHODS**

#### **Field Collection of Sediment Sample**

MGS collected 50 surficial sediment grab samples for this study. Sampling sites were selected to achieve spatial coverage; some sites were co-located at existing Maryland DNR water quality (WQ) and sub-aquatic vegetation (SAV) monitoring stations and Garret County Health Department (GCHD) stations (Figure 1).

Samples were collected in October 19 and 20, 2010. Locations of the sediment samples were document using a Lowrance GlobalNav 212 GPS interfaced to a Lowrance DGPS beacon receiver. Location coordinates were recorded in UTM, NAD83, meters. Location coordinates and water depths for the sediment locations are listed in Appendix I (Table 11).

Sediment samples were collected using a hand-operated LaMotte stainless-steel dredge which sampled a bottom surface area of 19 cm x 14 cm and a mean sediment depth of 10 cm.

Upon collection, the samples were placed in Whirl-Pak<sup>TM</sup> bags and kept cool until delivery to the MGS laboratory where they were refrigerated at 4˚ C. until analyses.



**Figure 1**. Sample locations. MGS sampling sites are represented by yellow circles, labeled with Site number. Also shown are existing Maryland DNR water quality (WQ) and sub-aquatic vegetation (SAV) monitoring stations and Garret County Health Department (GCHD) stations.

## **Laboratory Analyses**

## **Textural Analyses**

All sediment samples were analyzed for water content, bulk density, and grain size (sand, silt, clay contents, as well as gravel, when present). Two homogeneous splits of each sample are processed, one for bulk property analyses and the other for grain-size characterization. Analyses were performed as soon as possible after sample collection, and all samples were refrigerated in

sealed Whirl-Pak<sup>TM</sup> plastic bags prior to analysis.

Water content was calculated as the percentage of water weight to the weight of the wet sediment using equation 1.

$$
\%Water = \frac{W_w}{W_t} * 100
$$
 Equation 1

where:  $W_w$  is the weight of water; and  $W_t$  is the weight of wet sediment.

Water content was determined by weighing 20-30 g of sediment; the sediment was dried at 65°C, and then re-weighing the dried sediment. Dried sediments were saved for chemical analyses (see **Chemical Analyses** section).

Bulk density ( $\rho_B$ ) was calculated from water content utilizing equation (2) by assuming an average grain density ( $\rho_s$ ) of 2.72 g/cm<sup>3</sup> and saturation of voids with water of density  $\rho_w = 1.0$ g/cm<sup>3</sup>. This method was adopted from the work of Bennett and Lambert (1971):

$$
\rho_B = \frac{W_t}{W_d / 2.72 + W_w}
$$
 Equation 2

where  $W_d$  is the weight of dry sediment.

Sand, silt and clay contents were determined using the textural analysis detailed in Kerhin and others, (1988). Grain size analysis consisted of cleaning the samples in solutions of 10 percent hydrochloric acid and 6 or 15 percent hydrogen peroxide (determined by water content) with subsequent rinsing with deionized water. This process removed soluble salts, carbonates, and organic matter that could interfere with the disaggregation of the individual grains. The samples were then treated with a 0.26 percent solution of the dispersant sodium hexametaphosphate ( $(NaPO<sub>3</sub>)<sub>6</sub>$ ) to ensure that individual grains did not re-aggregate during analysis.

The separation of sand and silt-clay portions of the sample was accomplished by wetsieving through a 4-phi mesh sieve (0.0625 mm, U.S. Standard Sieve #230). The sand fraction was dried and weighed. The finer silt and clay-sized particles were suspended in a 1000 ml cylinder in a solution of 0.26 percent sodium hexametaphosphate. The suspension was agitated and, at specified times thereafter; 20 ml pipette withdrawals were made (Carver, 1971; Folk, 1974). The rationale behind this process was that larger particles settle faster than smaller ones (Stoke's law). By calculating the settling velocities for different sized particles, times for withdrawal can be determined at which all particles of a specified size will have settled past the point of withdrawal. Sampling times were calculated to permit the determination of the amount of silt (4 phi) and clay sized (8 phi) particles in the suspension. Withdrawn samples were dried at 65°C and weighed. From these data the percentages by dry weight of sand, silt, and clay were calculated for each sample and classified according to Shepard's (1954) nomenclature (Figure 2). Results of textural analyses are presented in Appendix II (Table 12).



**Figure 2.** Shepard's (1954) classification of sediment types

#### **Chemical Analyses**

Sediments dried for water content determination were saved for elemental analyses. The dried sediments were pulverized in tungsten-carbide vials using a ball mill, then placed in Whirl-Pak<sup>TM</sup> bags and stored in a desiccator.

#### *Nitrogen, Carbon and Sulfur Analyses*

The sediments were analyzed for total nitrogen (N), total carbon I, non-reactive C and total sulfur (S) using a Carlo Erba NA1500 analyzer. Untreated dried sediments were analyzed for total nitrogen, carbon and sulfur (NCS) contents. A split of dried sample was treated with15% hydrogen peroxide  $(H_2O_2)$  to remove "reactive" C (*i.e.*, carbon associated with labile or biologically active organic matter). This peroxide- treated sample was analyzed for non-reactive C which consists of inorganic or mineral C and non-labile C (Hennessee and others, 1986). Reactive C was calculated as the difference between total C and non-reactive C.

Approximately 10-15 mg of dried sediment (treated and untreated) was weighed into a tin capsule. The exact weight (to the nearest  $\mu$ g) of the sample was recorded. To ensure complete

combustion during the analysis, 15-20 mg of vanadium pentoxide  $(V_2O_5)$  was added to the sediment. The tin capsule containing the sediment and vanadium pentoxide mixture was then crimped to seal and stored in air-tight vial until analysis.

The sediment sample, contained in a tin capsule, was dropped into a combustion chamber where the sample was oxidized in pure oxygen. The resulting combustion gases (N, C, H, and S), along with pure helium used as a carrier gas, were passed through a reduction furnace to remove free oxygen and then through a sorption trap to remove water. Separation of the gas components was achieved by passing the gas mixture through a chromatographic column. A thermal conductivity detector was used to measure the relative concentrations of the gases.

The NA1500 Analyzer was configured for NCS analysis using the manufactu'er's recommended settings. As a primary standard, Sulfanilamide was used. Blanks (tin capsules containing only vanadium pentoxide) were run at the beginning and end of sample set. Replicates of every fifth sample were run. As secondary standards, one or more reference materials (NIST SRM #164–a - Estuarine Sediment and NIST SRM #87–4 - Buffalo River Sediment) were run every 5 samples. Comparisons of results of SRMs to their certified values are presented in Appendix I. Results of the NCS analyses are presented in Appendix II.

### *Elemental Analyses*

Two to three-gram splits of the dried sediments were shipped to Activation Laboratories, Ltd. (Actlabs) in Ontario, Canada, to be analyzed for 48 elements including total phosphorus (P) (Table 1). Sediments were analyzed either by neutron activation technique (INAA) or using a four-acid, "near total" digestion process, followed by analysis of the digestate by inductively coupled plasma emission spectroscopy (ICP-OES). The four-acid digestion employed perchloric (HClO4), hydrochloric (HCl), nitric  $(HNO<sub>3</sub>)$ , and hydrofluoric (HF) acids. Forty-three (43) of the 50 sediment samples were also analyzed for mercury using cold vapor extraction followed by Fluid-Injection Mercury System (FIMS). However, the recommended protocols for handling samples prior to the mercury analyses were not followed (*i.e.,* sediment samples were dried in open containers and holding time prior to analyses exceeded 14 days). Therefore, the reported Hg concentrations represent a portion of the total Hg originally contained in the sediments.

SRM NIST #8704 and #1646a were included as double- blind samples with the lake sediments submitted to Actlabs. The Actlabs' results of the analyses of the SRMs are listed in Appendix I (Table 10). Elemental analysis results for the surficial samples are listed in Appendix II (Table 14).

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			<b>Detection</b>				<b>Detection</b>				
<b>Analyte</b>	<b>Symbol</b>	Unit	Limit	Analyte	<b>Symbol</b>	Unit	Limit				
Silver	Ag	ppm	0.3	Manganese	Mn	ppm	1				
Gold	Au	ppb	$\overline{2}$	Molybdenum	Mo	ppm	$\mathbf{1}$				
Aluminum	Al	$\%$	0.01	Sodium	Na	$\%$	0.01				
Arsenic	As	ppm	0.5	Neodymium	Nd	ppm	5				
<b>Barium</b>	Ba	ppm	50	Nickel	Ni	ppm	$\mathbf{1}$				
Beryllium	Be	ppm	$\mathbf{1}$	Phosphorus	$\mathbf{P}$	$\%$	0.001				
<b>Bismuth</b>	Bi	ppm	$\overline{2}$	Lead	Pb	ppm	3				
<b>Bromide</b>	Br	ppm	0.5	Rubidium	<b>Rb</b>	ppm	15				
Calcium	Ca	$\%$	0.01	Sulfur	S	$\%$	0.01				
Cadmium	Cd	ppm	0.3	Antimony	Sb	ppm	0.1				
Cerium	Ce	ppm	3	Scandium	Sc	ppm	0.1				
Cobalt	Co	ppm	$\mathbf{1}$	Samarium	Sm	ppm	0.1				
Chromium	Cr	ppm	$\overline{2}$	Tin	Sn	$\%$	0.01				
Cesium	$\mathbf{C}\mathbf{s}$	ppm	$\mathbf{1}$	Strontium	Sr	ppm	1				
Copper	Cu	ppm	$\mathbf{1}$	Tantatum	Ta	ppm	0.5				
Europium	Eu	ppm	0.2	Terbium	Tb	ppm	0.5				
Iron	Fe	$\%$	0.01	Thorium	Th	ppm	0.2				
Hafnium	Hf	ppm	$\mathbf{1}$	Titanium	Ti	$\%$	0.01				
Mercury	Hg	ppb	$\mathbf{1}$	Uranium	U	ppm	0.5				
Iridium	Ir	ppb	5	Vanadium	$\mathbf V$	ppm	$\mathbf{2}$				
Potassium	$\bf K$	$\%$	0.01	Tungsten	W	ppm	$\mathbf{1}$				
Lanthanum	La	ppm	0.5	Yttrium	$\mathbf Y$	ppm	$\mathbf{1}$				
Lutetium	Lu	ppm	0.05	Ytterbium	Yb	ppm	0.2				
Magnesium	Mg	$\%$	0.01	Zinc	Zn	ppm	$\mathbf{1}$				

**Table 1.** List of the elements analyzed by Actlabs, Inc. along with units reported and laboratory detection limits.

### **RESULTS AND DISCUSSION**

### **Physical Characteristics**

Based on the textural analyses of the 50 surficial sediment samples (representing the top 5 cm of the sediment column), the majority of the samples collected are fine-grained sediments, with an average textural content of 18% sand, 39% silt and 43% clay. Thirty-two samples fall within the clayey-silt and silt-clay classifications (Table 2). Fourteen samples are classified as sand-silt-clay. Clay represents a major component of the collected sediments, which is not unexpected given the abundance of shale in the underlying formations within the watershed. Sand is a relatively minor component with only four samples classified as sand and silty-sand. The sand sample is the only sample to contain appreciable amounts of gravel (i.e., particles with diameter> 2mm). This sample was collected in the upstream reach of Cherry Branch (Figure 3) and contained abundant leaf litter, and organic matter which contributed to the higher water content. Typically, the water content of sand sized sediments averages around 20%. Water content increases with decreasing sediment size, with clayey sediments having water contents greater than 70%.



**Table 2.** Summary of sediment types for samples collected in Deep Creek Lake for this

The physical and chemical behavior of sediment is reflected in its texture. Particle diameter reflects the energy environment in which the sediment was deposited. Generally, coarse grained sediments (*i.e.*, sand and gravel) are found in higher energy environments, such as areas subjected to wave activity or high water currents, which tend to winnow out any fine grained sediment. Fine-grained sediments, which are transported further from the source and take a long time to settle, are usually found in areas that are not subjected to high waves or winds, or below deep of wave motion, such as deeper areas in the central area of the lake, or coves that are sheltered from high waves and winds. In Deep Creek Lake, the sediment distribution follows this pattern. The finest-grained sediments (i.e., sediment with highest clay content) are in the deepest part of the lake, whereas sandier sediments are found in shallower depths and in the up-stream areas.

Size also reflects the mineral composition of the sediments, which, in turn, is a product of the parent rock. The Deep Creek Lake watershed lies within sedimentary rock comprised of

mixed marine and non-marine shale, siltstone, and sandstone. The gravel- and sand- sized particles deposited in the lake consist primarily of quartz minerals, sand and gravel sized pieces of unweathered (intact) parent rock, large micas, feldspar, heavy minerals and sand sized coal particles. The quartz is a stable mineral and generally chemically inert. Clay minerals are abundant in the lake since shale is a common parent rock. Shales are lithified mud deposits, composed of silt sized particles (mainly quartz) and clay minerals which are the end product of weathering of other minerals. Clay particles are small, in the sub-micron range, platelike particles with a relatively large surface area. Thus, clay minerals comprise a significant portion of the clay size component of sediments. Depending on the crystalline lattice, clays have an enormous capacity to incorporate both organic and metal cations onto the lattice surface, and water and organic compounds within lattice layers. These bound substances, in turn, contribute to the cohesiveness of the clays. Organic rich clays, in turn, support active benthic bacteria and plankton communities.





**Figure 3.** Sediment distribution by sand content.

## **Nitrogen, Carbon, Sulfur and Phosphorus**

Total C contents measured in Deep Creek Lake sediments range from 1.5% to 9.6% (dry weight), with a mean of 4.1%. These values are lower than the average C reported for New

Germany Lake but higher than other fresh water reservoirs located in Central Maryland (Table 3). The total C in Deep Creek Lake consists of both reactive and non-reactive components. The non-reactive component is comprised of coal fragments, inorganic C contained in mineral such as limestone, and organic C containing high cellulose content such as woody debris associated with terrigenous (allochthonous) material. Reactive C contents average 2.7%, with values ranging from 0.6 % to 5.2%. Reactive C consists of aqueous algal matter as well as the more labile component of terrigenous carbon such as fresh leaf litter, decomposing animal and plant debris. On average, 70% of the total carbon contained in Deep Creek Lake sediment is reactive, readily available to the biological community. Coarser sediments (*i.e.,* low clay content) tend to contain a lower portion of reactive C (Figure 4).

Sediments in the northern portion of the lake contain overall higher carbon content (both total and reactive carbon) compared to the southern end (Figure 4). There is less variation in the non-reactive carbon content with regard to distribution.

Total carbon has little correlation with grain size; the poor correlation due to inclusion of non-reactive carbon which has no association with any particular sediment type (Table 4). However, reactive carbon has higher correlation ( $r = 0.4$ ) with clay as well as with N and P (compared to total C), indicating a significant portion of the reactive carbon in the sediment comes from primary productivity within the lake (plankton and algae blooms).



Total N measured in Deep Creek Lake sediments average 0.33%, with values ranging from 0.12% to 0.62%. Total N has the highest correlation with reactive C (Figure 5) indicating most N is associated with organic material, most likely from primary productivity (algae). Sources of N include atmospheric input, septic flow and fertilizers. As organic matter is "cycled through the natural system", relative proportions of P and, to a lesser degree, N increase as C decreases. Table 5 lists the Redfield ratios for N, C, and P for different sources and in sediment from several Maryland fresh and marine environments. The Deep Creek Lake ratios listed in Table 5 are calculated using total C. Ratios are smaller using reactive C: C:N=8.3; C:P=54.3, closer to the ideal Redfield ratio endpoint.



Deep Creek Lake -- Sediment Samples

16

 $0.00$ 

20.00

40.00

% Clay

60.00

80.00



Deep Creek Lake -- Sediment Samples

**Figure 5**. Nitrogen (% dry weight) distribution in sediments. Nitrogen is strongly associated with reactive carbon  $(R^2 = 0.72)$ , suggesting that the nitrogen is organic in nature, a significant portion likely from algae.

<b>Table 4.</b> Correlation matrix for textural, nutrient, and selected metals data based on 50 surficial sediment samples collected in Deep																		
Creek Lake. $C_T$ and $C_R$ denote total carbon and reactive carbon contents, respectively. The correlations were done using Pearson																		
product-moment technique (Johnson and Wichern, 1982). Values listed in the table are Pearson product-moment correlation																		
coefficienI(r). Values shown in regular type are significant at the 95% confidence level ( $p < 0.05$ ); values in red are not significant (p																		
$> 0.05$ ). The strongest correlations ( $r > 0.8$ ) are highlighted in <b>bold</b>																		
	Water	Sand	<b>Silt</b>	Clay	N	$C_T$	$C_R$	S	${\bf P}$	C <sub>d</sub>	Cu	Cr	Fe	Mn	Ni	Pb	Zn	As
Water		$-0.73$	0.28	0.79	0.74	0.41	0.63	0.63	0.77	0.58	0.78	0.55	0.81	0.17	0.75	0.72	0.75	0.75
<b>Sand</b>	$-0.73$		$-0.68$	$-0.92$	$-0.68$	$-0.28$	$-0.44$	$-0.42$	$-0.69$	$-0.34$	$-0.80$	$-0.63$	$-0.65$	$-0.01$	$-0.56$	$-0.61$	$-0.51$	$-0.54$
<b>Silt</b>	0.28	$-0.68$		0.33	0.35	0.22	0.29	0.06	0.16	0.21	0.33	0.52	0.24	0.20	0.30	0.20	0.23	0.11
<b>Clay</b>	0.79	$-0.92$	0.33		0.68	0.23	0.40	0.51	0.79	0.32	0.85	0.53	0.70	$-0.10$	0.56	0.67	0.53	0.64
${\bf N}$	0.74	$-0.68$	0.35	0.68		0.74	0.85	0.72	0.63	0.60	0.52	0.34	0.53	0.07	0.52	0.45	0.57	0.61
$C_T$	0.41	$-0.28$	0.22	0.23	0.74		0.94	0.45	0.26	0.44	0.02	$-0.01$	0.08	0.13	0.18	0.10	0.28	0.29
$C_R$	0.63	$-0.44$	0.29	0.40	0.85	0.94		0.60	0.46	0.63	0.25	0.17	0.34	0.17	0.44	0.33	0.50	0.51
S	0.63	$-0.42$	0.06	0.51	0.72	0.45	0.60		0.72	0.65	0.53	0.31	0.70	0.01	0.57	0.60	0.69	0.70
${\bf P}$	0.77	$-0.69$	0.16	0.79	0.63	0.26	0.46	0.72		0.52	0.81	0.48	0.86	$-0.04$	0.66	0.80	0.72	0.75
Cd	0.58	$-0.34$	0.21	0.32	0.60	0.44	0.63	0.65	0.52		0.47	0.43	0.61	0.27	0.82	0.62	0.88	0.58
Cu	0.78	$-0.80$	0.33	0.85	0.52	0.02	0.25	0.53	0.81	0.47		0.68	0.84	0.02	0.76	0.85	0.71	0.70
Cr	0.55	$-0.63$	0.52	0.53	0.34	$-0.01$	0.17	0.31	0.48	0.43	0.68		0.65	0.36	0.68	0.59	0.61	0.49
Fe	0.81	$-0.65$	0.24	0.70	0.53	0.08	0.34	0.70	0.86	0.61	0.84	0.65		0.11	0.80	0.87	0.84	0.77
Mn	0.17	$-0.01$	0.20	$-0.10$	0.07	0.13	0.17	0.01	$-0.04$	0.27	0.02	0.36	0.11		0.45	0.07	0.24	0.13
Ni	0.75	$-0.56$	0.30	0.56	0.52	0.18	0.44	0.57	0.66	0.82	0.76	0.68	0.80	0.45		0.79	0.90	0.72
Pb	0.72	$-0.61$	0.20	0.67	0.45	0.10	0.33	0.60	0.80	0.62	0.85	0.59	0.87	0.07	0.79		0.84	0.72
Zn	0.75	$-0.51$	0.23	0.53	0.57	0.28	0.50	0.69	0.72	0.88	0.71	0.61	0.84	0.24	0.90	0.84		0.69
As	0.75	$-0.54$	0.11	0.64	0.61	0.29	0.51	0.70	0.75	0.58	0.70	0.49	0.77	0.13	0.72	0.72	0.69	

**Table 4.** Correlation matrix for textural, nutrient, and selected metals data based on 50 surficial sediment samples collected in Deep



Although total P does not directly undergo reduction-oxidation processes in sediments, its cycling within the lake is controlled, in part, by the redox state of certain metals, particularly S and Fe, and by the concentration of organic matIal (C). Sources of P include weathering of natural soils and rocks, runoff from agricultural land and seepage from septic systems. Phosphate  $(PO<sub>4</sub><sup>-3</sup>)$  from fertilizers binds to soils, which erode during storm events adding suspended phosphate to streams that drain into the lake. Septic seepage may contribute phosphate in the form of orthophosphate and organic phosphorus. Unlike N and C, P has no gaseous form. Therefore, P does not cycle out of the system like N by way of denitrification or C by respiration. Thus P tends to accumulate in the sediments. Once in the sediments, P is slowly released into the interstitial water as organic material is oxidized. Free phosphate is rapidly bound to ferric oxyhydroxides and oxidized manganese which are found in the upper, oxidized layer of the sediments (i.e., oxidized flocculant layer on sediment surface). Deeper in the sediment column where anoxic conditions prevail and metals oxides have been reduced, P is released into the interstitial water and, if sulfide is low or absent, reacts with reduced forms of metals, particularly Fe, forming hydrous phosphates. However, if present, free sulfide will bind more readily to the reduced Fe and the phosphate remains free to diffuse upward to the oxidized layer whe"e it is "captured" by excess ferric oxyhydroxides (FeOOH ) and manganese oxides found in the upper sediment layer. If the overlying water column becomes anoxic, the "captured" P may be released in the overlying water column where it can contribute to increased algae/plankton production. The portion of total P active in this cycle includes the loosely sorbed phosphate, fresh, leachable, organic P, and iron-bound phosphate. These available forms of P make up 40% to 50% of the total P in the upper 1 cm of sediments and are largely depleted below 3 cm in the sediment column (Jorgensen, 1996). Any P below this depth usually consists of the more stable forms, bound to clay minerals, or associated with apatite or calcium carbonate minerals, and become permanently buried in the sediments.

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Figure 6. Total P (% dry weight) distribution in sediments. P is strongly associated with sediment clay content  $(R^2=0.63)$ . The three highest P contents were measured in sediments collected from the deepest part of the lake (depth > 15 m).

Total P measured in the sediments average 0.06%, with values ranging from 0.01% to 0.13%. These values are similar to those found in New Germany Lake (Table 3). Total P is associated with the sediment clay content (Figure 6). P also shows a high correlation with many of the metals as well as S (Table 4). When comparing the relative amounts of C, N and P in the Deep Creek Lake sediments to those of dried algae, P appears to be the limiting nutrient. In other words, mean C:P and N:P ratios are greater than those of dried algae.

Sulfur measured in the sediments average 0.26% with values ranging from trace to 1%. S is significantly higher in some of the Deep Creek Lake sediments, particularly those collected in the downstream end (north end) (Figure 7). The very dark color (black and dark grey) of some sediments collected indicated the presence of S, in the form of mono-sulfides (FeS). Sources of S include sulfates from acid mine drainage and sulfide bearing minerals (pyrites) in the marine shales and siltstones, and atmospheric deposition within the watershed. Another source could be from slow release fertilizer (granules) that use sulfur coatings.



Deep Creek Lake -- Sediment Samples



Reduced S and sulfate  $(SO<sub>4</sub><sup>-2</sup>)$  concentration is an extremely important variable controlling P release from sediments (Caraco *et al.* 1989; Wetzel, 1983). The increased P release from sediments at higher sulfate concentrations may help explain why primary production in freshwater systems (with relatively low S concentrations) tends to be P limited, whereas in many saline systems (with high sulfate concentrations) production is often P sufficient. Sulfur also plays an important role in arsenic cycling (Fisher et al., 2008), which explains the high correlations between S, As, and Fe (Table 4).

## **Metals**

Table 6 lists summary statistics for those metals having reported threshold limits listing in NOAA Screening Quick Reference Tables (Buchman, 2008). Most elements listed in Table 6 are above background levels. Because the Deep Creek Lake samples were analyzed using a near total decomposition method (four-acid digestion), caution is warranted when comparing the resulting concentration values for some metals to threshold limits given in the NOAA tables. The values listed in the NOAA tables are based on EPA methods which allow partial decomposition of sediment samples and thus reflects that portion of any element that may become biologically available/mobile under extreme environmental conditions. For example, the NOAA tables list background levels in soil/sediments for Al as 0.26% which reflects the average Al biologically available. However, Actlabs' results for Al range from 1.06% to 7.33%, reflecting total recovery of the element by the digestions method used. Al is a major component of most minerals found in native rock and soils. Likewise, average concentrations of Fe and Mn exceed the NOAA background levels for the same reasons given for Al.

**Table 6**. Summary statistics of select metal concentration measured in Deep Creek sediments. All values are ppm unless otherwise indicated. For comparison, benchmark levels for freshwater sediments are included along with the number of lake samples exceeding the respective limit values. These benchmarks for freshwater sediments are based upon chronic, long-term impacts of contamination to benthic organisms (Buchman, 2008). The Lowest Effect Level (LEL) is a level of sediment contamination that can be tolerated by the majority of benthic organisms. The Severe Effect Level (SEL) is that at which pronounced disturbance of the sediment-dwelling community can be expected. This is the concentration that would be detrimental to the majority of the benthic community.



Hg, on the other hand, may be compared to the NOAA tables. Hg, was analyzed using cold vapor extraction followed by FIMS method. However, because standard handing protocols for Hg sample analyses were not followed; maximum recoveries most likely were not achieved. Nevertheless, reported Hg concentrations for many of the Deep Creek Lake sediments exceeded the upper background level for fresh water sediments. Hg concentration reported for one sample, Sta. #2, exceeded the LEL threshold value. Generally, reported Hg concentrations increased with clay content (Figure 8). The levels of Hg concentration in the sediments are not unexpected and probably are higher than reported.





While, most metals of concern (As, Cd, Cr, Cu, Hg, Ni, Pb, Zn) are above the LEL in most sediments (Table 6), very few exceeded the SEL value. Most sediments containing metals above the LEL correspond the fine-grained sediments in the deepest part of the lake. Many of the metals show a significant correlations with clay content and nutrient content (N and P) as well as with other metals (Table 4).

#### *Enrichment Factors*

Because of the wide range of sediment types analyzed, comparisons of absolute metal concentrations between the surficial sediments are difficult. Therefore, metal concentrations are discussed in terms of enrichment factors (EF). The use of enrichment factors also allows for comparisons of sediments from different environments and the comparisons of sediments whose trace metal contents were obtained by different analytical techniques (Cantillo, 1982; Hill *et al*., 1990; Sinex and Helz, 1981). However, the use of enrichment factors to assess metal data does not entirely eliminate the influence of textural variation.

Enrichment factor is defined as:

$$
EF_{(x)} = X/Fe_{(sample)} / X/Fe_{(reference)}
$$
 Equation 3

where:  $EF_{(x)}$  is the enrichment factor for the metal X;

 $X/Fe$ <sub>(reference)</sub> is the ratio of the concentrations of metal X to Fe in a reference material, such as an average continental crust rock.

Fe is chosen as the element for normalizing because anthropogenic sources for Fe are small compared to natural sources (Helz, 1976). Taylor's (1964) average continental crust is

used as the reference material. Average crustal abundance data may not be representative of Deep Creek Lake sediments because there is a higher proportion of clay and silt in the sediments compared to the average crustal rock. However, abundance data is useful as a relative indicator when comparing the data with other studies.

The average EF values for most metals are within those values obtained for other freshwater lakes and reservoirs (Table 7). The sediments in Deep Creek Lake are significantly enriched (*i.e.*, EF>3) in As, Cd, Cs, Hf, Pb, Sb, and Zn, with respect to average continental crust rock. The EF values are higher than those reported for New Germany Lake, which is located in the same physiographic and atmospheric deposition regions and, thus is expected to be similar in geochemistry. The higher enrichments, particularly As, Cd, and Sb, in Deep Creek Lake sediments are attributed to contribution from coal deposits within the lake's watershed. Coal deposits are generally enriched with these metals. Issues with documented acid mine drainage processes most likely further mobilized these elements. However, when EF values are plotted on a map, there appears to be not general pattern as to the distribution of the highest EF values. This may be due to several factors. There may be more than one source of the elements. The elements may have different geochemical behaviors during and after deposition in the lake. It should be noted that EF values for Pb are significantly high for all of the lakes listed illustrating the widespread anthropogenic sources for Pb.



**Table 7**. Comparisons of average enrichment factors in several Maryland fresh water lakes

### **SUMMARY AND CONCLUSIONS**

Deep Creek Lake bottom sediment reflect the geology of the watershed. In general, the surficial sediments sampled in Deep Creek Lake are primarily fine-grained, ranging from silty clays to clayey silts. The predominance of clay size sediments is not unexpected given the abundance of shale in the underlying formations within the watershed.

Total C, N and P concentrations measured in Deep Creek Lake sediments are within the range of those reported for other Maryland freshwater lakes. Reactive carbon accounts for 70% of total carbon. Comparisons of total and reactive C content with N and P in terms of Redfield ratios for plankton and algae suggest that a significant portion of the reactive carbon is from primary productivity. Also, P appears to be the limiting nutrient in Deep Creek Lake sediments.

Unlike other Maryland freshwater lakes, total S content is high in some of the lake sediments. Sources of S include sulfates from acid mine drainage and sulfide bearing minerals (pyrites) in the marine shales and siltstones, and atmospheric deposition within the watershed. Under anoxic conditions, sulfur may contribute to the increased release of P from sediments, which, in turn, may increase primary productivity (i.e., algae blooms).

Concentration and enrichment of most metals in Deep Creek Lake sediments are within normal range given the geology of the watershed. The sediments are significantly enriched in As, Cd, Cs, Hf, Pb, Sb, and Zn, with respect to average continental crust rock. The EF values are higher than those reported for New Germany Lake, which is located in the same physiographic and atmospheric deposition regions and, thus is expected to be similar in geochemistry. The higher enrichments, particularly As, Cr, and Sb, in Deep Creek Lake sediments are attributed to contribution from coal deposits within the lake's watershed. Coal deposits are generally enriched with these metals as well as other rare earth elements. Documented acid mine drainage processes most likely mobilized these elements, resulting in higher concentrations compared to New Germany, where acid mine drainage may not be an issue. It should be noted that EF values for Pb are significantly high for all of the lakes listed, illustrating the widespread anthropogenic sources for Pb. Nevertheless, Pb levels in Deep Creek Lake sediments are well below the SEL benchmark threshold for freshwater sediments.

Reported Hg concentrations for many of the Deep Creek Lake sediments exceeded the upper background level for fresh water sediments. Generally, reported Hg concentrations increased with clay content. The levels of Hg concentration in the sediments are not unexpected and probably are higher than reported.

## **ACKNOWLEDGEMENTS**

The authors wish to thank Stephen VanRyswick for his assistance in collecting the sediment samples, and Nicholas Kurtz for assisting in the physical and chemical analyses of the sediments.

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# **Appendix I QA/QC**

## **Textural Analyses**

Although the techniques used to determine grain size are based on traditional analytical methods developed for the sedimentology laboratory, some analytical error is inherent to the techniques. For example, results can be affected by level of technician skill and/or changes in laboratory conditions (such as sudden temperature changes). Furthermore, there is no standard reference material available that includes the broad range of particle sizes and shapes contained in natural sediment. To maximize consistency of textural analysis, several "checks" are used to monitor results. The calculated sand, silt, clay and gravel (when present) percentages are checked against 1) sample field descriptions; 2) calculated water contents; and 3) calculated weight loss of sample during processing. These comparisons are made to determine if the size components match the visual description of the sample and/or fall within an expected classification with respect to water content and weight loss. Any discrepancy is "flagged" and the results are reviewed further to determine if re-analysis is warranted.

## **Elemental Analyses**



**Table 8.** Results of nitrogen, carbon, and sulfur analyses of the standard reference materials (SRMs) compared to the certified or known values. MGS values were obtained by averaging the results of all SRM analyses run during this study.

other laboratories (Haake Buchler Labs and U.S. Dept. of Agriculture).

<sup>2</sup>For SRM the value for sulfur are certified values reported by NIST; nitrogen and carbon values were obtained from repeated analyses in-house and by Actlabs

**Table 9.** Results of analyses of Standard Reference Material (NIST SRM #8704 - Buffalo River Sediment) submitted as blind unknowns with the Deep Creek Lake surficial samples. Also given are the method detection limits for each element reported by Actlabs, Inc..



**Table 9.** Results of analyses of Standard Reference Material (NIST SRM #8704 - Buffalo River Sediment) submitted as blind unknowns with the Deep Creek Lake surficial samples. Also given are the method detection limits for each element reported by Actlabs, Inc..



**Table 10.** Results of analyses of Standard Reference Material (NIST SRM #1646a- Estuarine Sediment) submitted as blind unknowns with the Deep Creek Lake surficial samples. Also given are the method detection limits for each element reported by Actlabs, Inc..





**Table 10.** Results of analyses of Standard Reference Material (NIST SRM #1646a- Estuarine Sediment) submitted as blind unknowns with the Deep Creek Lake surficial samples. Also given are

## **Appendix II Textural and Chemical Data**







**Table 12**. Physical characteristics of the surficial sediment sample collected in Deep Creek Lake.



Lake.								
	Water		Size component (% dry weight)					
	content	<b>Bulk</b>						
	$%$ wet	<b>Density</b>					Shepard's <b>Classification</b>	
<b>Station</b>	weight	$g/cm^3$	<b>Gravel</b>	<b>Sand</b>	<b>Silt</b>	<b>Clay</b>	Sand-Silt-	
DCL-29	58.55	1.36	0.00	36.09	42.04	21.88	Clay	
							Sand-Silt-	
$DCL-30$	65.96	1.27	0.00	20.38	37.63	41.98	Clay	
$DCL-31$	72.59	1.21	0.00	2.08	41.63	56.28	Silty-Clay	
<b>DCL-32</b>	69.14	1.24	0.00	3.72	35.03	61.26	Silty-Clay	
DCL-33	65.22	1.28	0.00	3.19	42.82	53.99	Silty-Clay	
DCL-34	74.00	1.20	0.00	12.41	38.55	49.04	Silty-Clay	
							Sand-Silt-	
<b>DCL-35</b>	68.59	1.25	0.00	30.29	29.47	40.23	Clay	
<b>DCL-36</b>	72.77	1.21	0.00	1.22	33.78	65.00	Silty-Clay	
<b>DCL-37</b>	75.34	1.18	0.00	4.65	38.50	56.85	Silty-Clay	
DCL-38	71.48	1.22	0.00	0.66	40.59	58.75	Silty-Clay	
DCL-39	74.59	1.19	0.00	1.25	45.38	53.37	Silty-Clay	
$DCL-40$	62.54	1.31	0.00	7.75	53.65	38.61	Clayey-Silt	
							Sand-Silt-	
DCL-41	67.44	1.26	0.00	33.14	39.02	27.84	Clay	
<b>DCL-42</b>	66.40	1.27	0.00	5.33	55.55	39.12	Clayey-Silt	
DCL-43	69.39	1.24	0.00	1.72	42.14	56.14	Silty-Clay	
DCL-44	59.47	1.34	0.00	58.95	23.72	17.33	Silty-Sand	
<b>DCL-45</b>	73.70	1.20	0.00	5.14	53.06	41.81	Clayey-Silt	
<b>DCL-46</b>	68.78	1.25	0.00	1.09	40.30	58.61	Silty-Clay	
							Sand-Silt-	
DCL-47	51.15	1.45	0.00	33.62	38.88	27.51	Clay	
DCL-48	68.25	1.25	0.00	1.22	42.65	56.14	Silty-Clay	
<b>DCL-49</b>	70.60	1.23	0.00	0.90	43.76	55.34	Silty-Clay	
$DCL-50$	53.38	1.42	0.00	29.51	49.92	20.56	Sand-Silt- Clay	

**Table 12**. Physical characteristics of the surficial sediment sample collected in Deep Creek















