



North Carolina Mercury TMDL

Sept 13, 2012
Modeling and TMDL Unit



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SUMMARY

North Carolina Mercury Total Maximum Daily Load (TMDL)

1. 303(d) Listed Waterbody Information

State: North Carolina

County: All counties in North Carolina

Major River Basin: All river basins in North Carolina, including Broad, Cape Fear, Catawba, Chowan, French Broad, Hiwassee, Little Tennessee, Lumber, Neuse, New, Pasquotank, Roanoke, Savannah, Tar-Pamlico, Watauga, White Oak, Yadkin-Pee Dee

Watershed: All watersheds in North Carolina

Impaired Waterbody (2010 303(d) List): All 13,123 Waters in North Carolina are in category 5-303(d) List due to statewide fish consumption advisory for several fish species.

Constituent(s) of Concern: Mercury

Designated Uses: Fish Consumption

Applicable Water Quality Target:

The U.S. Environmental Protection Agency (EPA) and Food and Drug Administration (FDA) recommended fish tissue water quality criterion of 0.3 mg methylmercury (MeHg) / kg fish is selected as the target level for this TMDL. Since fish tissues were monitored for total mercury in North Carolina, and studies have shown that the majority of mercury concentrations in fish tissues are in the form of methylmercury, the 0.3 mg MeHg/kg fish tissue mercury target is applied to total mercury in fish tissues in this TMDL study. To protect water bodies from impairment, the 90th percentile standardized-length Largemouth Bass (LMB) fish tissue total mercury concentration is selected to meet the target level.

TMDL expression: This TMDL is expressed as both annual and daily loads.

2. TMDL Development

Development Tools (Analysis/Modeling): Basic statistics and Community Multi-scale Air Quality (CMAQ) model

Seasonal Variation: Mercury deposition and concentrations in water vary due to seasonal differences in rain and wind patterns, but this variation is not relevant because mercury concentrations in fish represent accumulation over their life spans. Factors such as size and waterbody conditions have greater effect on mercury concentrations than seasonal variation. The mercury concentration in the fish represents an integration of all temporal variation up to

the time of sample collection. Variability among fish because of differences in size, diet, habitat, and other undefined factors are expected to be greater in sum than seasonal variability. This TMDL is expressed as an average annual load.

Critical Conditions: Critical conditions in this TMDL are related to sensitivities of water bodies to mercury loading because of their water chemistry. Fish mercury concentrations tend to be higher in the eastern coastal plain regions of North Carolina than in the mountain and piedmont regions. This aspect of critical conditions has been addressed in this TMDL by using the 90th percentile of the standardized-length Largemouth Bass mercury concentration over the entire State of North Carolina. The mercury concentrations in the most popular and most likely consumed fish species in eastern North Carolina are usually less than those found in Largemouth Bass, and much less than the 90th percentile of the standardized-length Largemouth Bass. It is reasonable to expect that mercury concentrations in the most likely consumed fish species in eastern North Carolina will be lower than the target level once the Largemouth Bass mercury concentrations would decline below the target level. The 90th percentile is calculated from standardized-length Largemouth Bass mercury data, which has also avoided the selection of rare incidences from original samples of large-sized, long-living fish. By taking into consideration the most sensitive water bodies, the relatively insensitive water bodies will be protected as well.

3. TMDL Allocation Summary

In order to protect North Carolina waters from mercury impairment and ultimately remove the fish consumption advisory, a 67% reduction is needed from the 2002 baseline mercury loading. The final TMDLs for North Carolina are shown in the following table for both annual and daily loads.

Table 1. TMDL allocation summary

	Annual Load*		Daily Load	
	(kg/yr)	(lbs/yr)	(kg/day)	(lbs/day)
Baseline Point Source	112	247	0.3	0.7
Baseline Nonpoint Source	5,437	11,961	14.9	32.8
Baseline Total	5,549	12,208	15.2	33.5
Margin Of Safety	Implicit	Implicit	Implicit	Implicit
Wasteload Allocation	37	81	0.1	0.2
Load Allocation	1,794	3,948	4.9	10.8
Total Maximum Daily Load	1,831	4,029	5.0	11.0

*Annual load is included to facilitate implementation of the daily allocations as appropriate in NPDES permits and nonpoint source directed management measures, see Section 6.7.

Point source discharges are considered a small contribution to mercury concentrations in fish since cumulative baseline loading of all wastewater point sources to the receiving waters accounts for only 2% of total mercury loadings. Significant decreases in mercury loading will require reductions in atmospheric deposition. The TMDL does not regulate mercury loading from atmospheric deposition; achieving those reductions will require strategies that fall outside the scope of the NPDES permitting program.

For this mercury TMDL, the wasteload allocation (WLA) is defined as 2% of the TMDL to ensure that water point source mercury loads remain small and continue to decrease. Due to the small percentage contribution from point sources, the WLA is statewide and not specified to individual source, thereby providing a cap for the state. Instead of allocating the WLA among sources with individual limits, mercury reduction will be accomplished primarily through mercury minimization plans (MMPs) as needed and ancillary efforts that reduce point source particulate loading (e.g., phosphorus controls, biochemical oxygen demands (BOD) and total suspended solids (TSS) reductions, etc).

The primary nonpoint source of mercury is from deposition of air emissions and hence load is allocated to air deposition. The allowable loads from atmospheric nonpoint sources are allocated proportionally to their existing contributions. Load allocations (LA) and expected reductions are listed in the following table.

Table 2. TMDL Load Allocation and Expected Reduction

Nonpoint Source		Percentage Contribution	Baseline Load		Allowable Load		Expected Reduction
			(kg/yr)	(lbs/yr)	(kg/yr)	(lbs/yr)	
Global*		66%	3,661	8,054	1,208	2,658	67% [#]
Regional**		15%	844	1,857	278	612	67% [#]
In-State	Natural	1%	56	123	56	123	N/A
	Anthropogenic	16%	876	1,927	252	555	71%
Total		98%	5,437	11,961	1,794	3,948	67%

* In this TMDL, mercury air sources coming from outside the CMAQ model 12 km model domain are referred as global sources.

**Mercury air sources within the CMAQ model 12 km model domain but outside North Carolina are referred as regional sources.

[#]Expected percent reductions from global and regional air sources are reductions in total air deposition of mercury.

As noted above, however, the NPDES permitting program does not regulate air emissions and emission reductions necessary to meet the load limit would require other mercury reduction strategies at the state, national and international level.

4. Public Notice Date:

5. Submittal Date: to be determined

6. Establishment Date: to be determined

7. EPA Lead on TMDL (EPA or blank):

8. Endangered Species (yes or blank):

9. MS4s Contributions to Impairment (Yes or Blank):

10. TMDL Considers Point Source, Nonpoint Source, or both: Both

1. INTRODUCTION

Mercury is a naturally occurring element that cannot be created or destroyed. The same amount of mercury has existed on the planet since the earth was formed (EPA, 1997). Mercury, however, can cycle in the environment through both natural (*e.g.*, volcanoes, fires, surface emissions) and human activities (*e.g.*, combustion, commercial products). Human activities have increased the amount of mercury that is available in the atmosphere, in soils and sediments, and in various water bodies (EPA, 2006b). Measured data and modeling results indicate that the amount of mercury mobilized and released into the biosphere has increased since the beginning of the industrial age (EPA, 1997).

Humans are exposed to mercury primarily through consumption of fish that contain methylmercury. Methylmercury is a neurotoxin that is biomagnified in aquatic food webs so that fish, wildlife and humans that consume fish, are potentially at greater risk of exposure to methylmercury. Research shows that fish consumption does not cause a health concern for most people. However, outbreaks of methylmercury poisonings have demonstrated that high levels of methylmercury in the bloodstream of unborn babies and young children may harm the developing nervous system, making the child less able to think and learn (EPA, 1997).

Mercury can also impact the ecological systems that humans rely on for other food sources and for recreation. Birds and mammals that eat fish are more exposed to mercury than other animals in water ecosystems. Similarly, predators that eat fish-eating animals may be highly exposed. Research suggests that at high levels of exposure, methylmercury's harmful effects on these animals include death, reduced fertility, slower growth and development, and abnormal behavior (EPA, 1997).

Fish Consumption Advisory in North Carolina

Based on fish tissue data routinely collected by N.C. Department of Environment and Natural Resources (NC DENR), the N.C. Department of Health and Human Services (NC DHHS) has issued a statewide fish consumption advisory for fish that contain mercury, advising people to either limit consumption or avoid eating those kinds of fish. NC DHHS advises that most fish are good to eat and good for people, but some kinds of fish contain high amounts of mercury that can cause health problems in people, especially children. The following table is a summary of the advisory as of January, 2012 (<http://www.epi.state.nc.us/epi/fish/current.html>).

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Table 1-1. North Carolina Fish Consumption Advisory Summary

	Fish Low in Mercury	Fish High in Mercury
Women of Childbearing Age (15-44 years), Pregnant Women, Nursing Women, and Children under 15	Eat up to 2 meals per week	Do not eat
All Other Individuals	Eat up to 4 meals per week	Eat only 1 meal per week

Fish high in mercury include:

Ocean fish: Albacore (white) tuna** fresh or canned, Almaco jack, Banded rudderfish, Cobia, Crevalle jack, Greater amberjack, South Atlantic grouper (gag, scamp, red and snowy), King Mackerel, Ladyfish, Little tunny, Marlin, Orange roughy, Shark, Spanish mackerel, Swordfish, Tilefish, and Tuna (fresh or frozen**)

Freshwater fish: Blackfish (bowfin)*, Black crappie***, Catfish (caught wild)*, Jack fish (chain pickerel)*, Largemouth bass (statewide), Walleye from Lake Fontana and Lake Santeetlah (Graham and Swain counties), and from Lake Gaston (Warren, Halifax and Northampton Counties), Warmouth*, and Yellow Perch*

*High mercury levels have been found in blackfish (bowfin), catfish, jack fish (chain pickerel), warmouth, and yellow perch caught south and east of Interstate 85.

**Different species from canned light tuna

***High mercury levels have been found in black crappie caught south and east of Interstate 95.

303(d)/Total Maximum Daily Loads (TMDLs)

Section 303(d) of the Clean Water Act (CWA) requires states to develop a list of waters not meeting water quality standards or which have impaired uses. This list is submitted biennially to the U.S. Environmental Protection Agency for review.

Due to the statewide fish consumption advisory in North Carolina, the designated uses of all statewide water bodies are impaired by mercury. Therefore, all named water bodies in NC are included in the 2008 and 2010 303(d) list for mercury impairment.

The 303(d) process requires that a Total Maximum Daily Load (TMDL) be developed for each of the listed waters, where technically feasible. EPA characterizes the TMDL as the sum of the wasteload allocation (WLA), load allocation (LA), and a margin of safety (MOS), or

$$TMDL = WLA + LA + MOS$$

The wasteload allocation portion of the TMDL accounts for the loads from existing and future point sources. The load allocation portion of the TMDL accounts for the loads from existing and future nonpoint sources and natural background. The margin of safety addresses uncertainties in the data collection and modeling techniques (EPA, 1998). The objective of a TMDL is to allocate allowable pollutant loads to known sources so that actions may be taken to restore the water to its intended uses (EPA, 1991). The ultimate objective of this TMDL is to reduce fish tissue levels of mercury so that fish consumption advisories can be removed and the fish can be safely consumed.

EPA (2008) has identified the primary components of a TMDL where mercury loadings are predominantly from air deposition, as follows:

a. Identification of Waterbodies, Pollutant Sources

For regional or statewide approaches, this would include identification of the geographic area and specific waterbodies covered and not covered by the TMDL; description of factors such as land use, water quality, fish tissue data, sources, and loadings within each region in order to support a regional approach; and rationale for how and why waterbodies can be grouped.

b. Water Quality Standards and TMDL Target

TMDLs must describe applicable water quality standards (WQS) and identify a numeric TMDL target, a quantitative value used to attain and maintain the applicable WQS.

c. Loading Capacity – Linking Water Quality and Pollutant Sources

TMDLs must identify loading capacity and reductions needed to meet water quality standards. A linkage analysis is usually provided to link between the numeric TMDL target and mercury sources, including both point sources and nonpoint sources.

d. Allocation of Pollutant Loads

Allocating pollutant control responsibility to the sources of impairment. The wasteload allocation portion of the TMDL accounts for the loads associated with existing and future point sources. Similarly, the load allocation portion of the TMDL accounts for the loads associated with existing and future nonpoint sources and natural background.

e. Margin of Safety (MOS)

The margin of safety addresses uncertainties associated with pollutant loads, modeling techniques, and data collection. Per EPA (2000), the margin of safety may be expressed explicitly as unallocated assimilative capacity or implicitly due to conservative assumptions.

f. Seasonal Variation & Critical Conditions

TMDLs should take into account critical conditions for stream flow, loading and water quality parameters on the TMDL calculation. For mercury, critical conditions might include impacts of land use, erosion, sulfates, dissolved organic carbon (DOC), and pH on mercury methylation and bioaccumulation, as well as the impacts of meteorology on mercury deposition. As mercury bioaccumulates over time, annual variations are usually considered more important than seasonal variations, particularly if a fish tissue target is used. For regional or statewide approach, mercury TMDLs should also take into account differences in critical conditions & sensitivity to methylation between waterbodies or regions.

Section 303(d) of the CWA and the Water Quality Planning and Management regulation (EPA, 2000) require EPA to review all TMDLs for approval or disapproval. Once EPA approves a TMDL, then the waterbody may be moved to Category 4a of the Integrated Report. Waterbodies remain in Category 4a until compliance with water quality standards is achieved.

2. BACKGROUND INFORMATION

2.1 Mercury Species and the Mercury Cycle

Detailed descriptions about mercury species and the mercury cycle can be found in a number of publicly-available articles including those published by EPA (<http://www.epa.gov/hg/about.htm>), U.S. Geological Survey (<http://www.usgs.gov/mercury/>), and The Encyclopedia of Earth (<http://www.eoearth.org/article/Mercury>). Some related information is summarized in the following:

Forms of Mercury

Mercury is a naturally occurring element that is found in air, water and soil. It exists in several forms: elemental or metallic mercury, inorganic mercury compounds, and organic mercury compounds.

Metallic mercury is the elemental or pure form of mercury and it is a liquid at room temperature. Metallic mercury is traditionally used in thermometers and some electrical switches. At room temperature, some of the metallic mercury will evaporate and form mercury vapors. Mercury vapors are colorless and odorless.

Inorganic mercury compounds occur when mercury combines with elements such as chlorine, sulfur, or oxygen. These mercury compounds are also called mercury salts. Most inorganic mercury compounds are white powders or crystals.

When mercury combines with carbon, the compounds formed are organic mercury compounds. There are a potentially large number of organic mercury compounds. The most common organic mercury compound in the environment is methylmercury.

Sources of Mercury

Mercury naturally enters the environment as the result of the normal breakdown of minerals in rocks and soil from exposure to wind and water, and from volcanic activity. It is believed that mercury releases from natural sources have remained relatively constant in recent history.

Human activities since the start of the industrial age have resulted in additional release of mercury to the environment. Approximately 80% of the mercury released from human activities is elemental mercury released to the air, primarily from fossil fuel combustion, mining, and smelting, and from solid waste incineration. Coal-burning power plants are the largest man-made source of mercury emissions to the air in the United States, accounting for over 50% of all domestic human-caused mercury emissions. About 15% of the total is released to the soil from fertilizers, fungicides, and municipal solid waste (for example, from waste that contains discarded batteries, electrical switches, or thermometers). Discharges of industrial wastewater account for an additional 5% of mercury released to surface waters.

Annual mercury releases due to human activities are estimated to account for between one-third and two-thirds of total annual mercury releases. A major uncertainty in these estimates is the amount of re-emission of mercury that was previously deposited due to human activities as opposed to new natural releases. The continuous release of mercury has resulted in current levels that are three to six times higher than the estimated levels in the preindustrial era atmosphere.

Mercury Cycle

Most of the mercury found in the environment is in the form of metallic mercury and inorganic mercury compounds. In air, the elemental mercury vapor can be changed into other forms of mercury and further transported to water or soil in rain or snow. The levels of mercury in the atmosphere are very low and do not pose a health risk.

Mercury in the air eventually settles into water or onto land where it can be washed into water. Once deposited, certain microorganisms can change it into highly toxic methylmercury. Methylmercury released from microorganisms can enter the water or soil and remain there for a long time. Methylmercury can also enter and accumulate in the food chain. Methylmercury is stored in the tissue of small fish that eat aquatic organisms containing methylmercury, when a larger fish eats the smaller fish, most of the methylmercury in the small fish will then be stored in the body of the larger fish. As a result, the larger and older fish living in contaminated waters build up the highest amounts of methylmercury in their bodies. Fish and shellfish are the main sources of methylmercury exposure to humans.

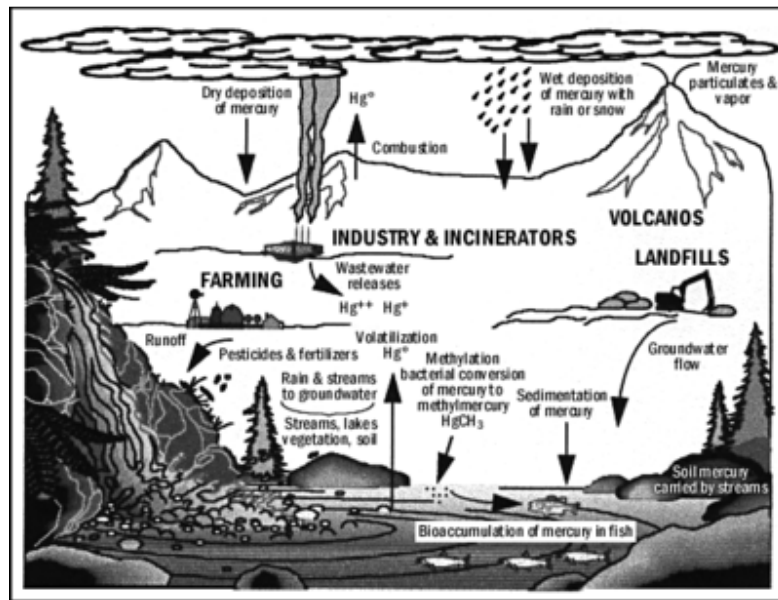


Figure 2-1. Mercury Cycling.

2.2 Data Collection and Assessment

Several types of data are used in this TMDL:

- NC Division of Water Quality (DWQ) fish tissue mercury data
- DWQ Eastern Regional Mercury Study data,
- National Pollutant Discharge Elimination System (NPDES) data
- NC Division of Air Quality (DAQ) CMAQ (Community Multi-scale Air Quality) modeling air deposition data

Data collection programs and the methods used are described in this section while the data analysis results are discussed in the following sections.

DWQ Fish Tissue Mercury Data

The Environmental Sciences Section of DWQ has collected fish tissue samples for total mercury analysis since 1978. Data are usually reported starting from 1990 for consistency in laboratory analysis protocols (<http://portal.ncdenr.org/web/wq/ess/bau/fish-tissue-data>). From 1990 to 2008, the Division processed and analyzed 6,436 fish tissue samples for mercury analysis. The dataset covers about 275 statewide sampling locations.

Largemouth Bass (*Micropterus salmoides*, *LMB*) embody the largest data subset within the DWQ fish tissue mercury database, representing 2,311 or 36% of the 6,436 records collected from 1990 to 2008. The average fish tissue concentration for total mercury in Largemouth Bass was 0.52 mg/kg, much higher than the state's fish consumption advisory action level of 0.4 mg/kg. Other frequently recorded fish species in the 1990-2008 DWQ mercury database include Bowfin (*Amia calva*), Bluegill (*Lepomis macrochirus*), Redear Sunfish (*Lepomis microlophus*), and Channel Catfish (*Ictalurus punctatus*). Collective records for these four species represent approximately 24% of the DWQ fish tissue mercury data collected from 1990 to 2008.

The seven most common species that exceed the state's fish consumption advisory action level (in addition to Largemouth Bass) are: Bowfin, Chain Pickerel (*Esox niger*), King Mackerel (*Scomberomorus cavalla*), Warmouth (*Lepomis gulosus*), Yellow Perch (*Perca flavescens*), Spotted Sucker (*Minytrema melanops*), and Walleye (*Stizostedion vitreum*). The average fish tissue mercury concentrations for these species ranged from 0.42 (Spotted Sucker) to 0.95 mg/kg (King Mackerel). However, the number of samples collected for these species were much less than those for Largemouth Bass. In addition, some of these species (e.g. Bowfin and Walleye) are only found in specific regions within the state, which makes them less representative (than Largemouth Bass) of state-wide mercury issues in North Carolina.

Figure 2-2 shows the number of samples collected for Largemouth Bass and Bowfin at stations across North Carolina from 1990 to 2008.

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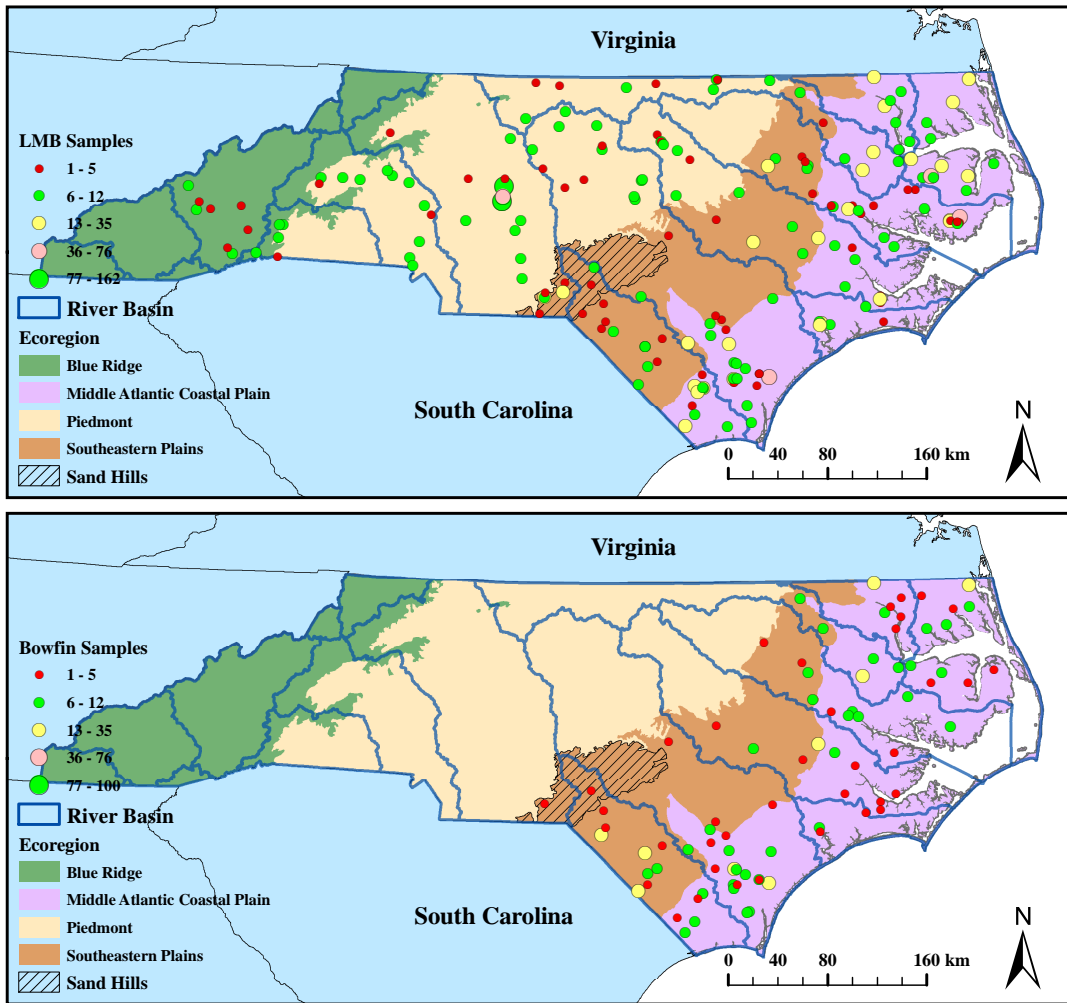


Figure 2-2. The number of samples collected for Largemouth Bass (upper panel) and Bowfin (lower panel) at stations across North Carolina during 1990 to 2008.

DWQ Eastern Regional Mercury Study and Mercury Study Extension

The Eastern Regional Mercury Study (ERMS) was conducted from November 2002 to July 2003 to determine low level mercury concentrations in surface waters and sediments at 11 sites in eastern North Carolina (DWQ, 2003). In 2004, the study was extended to include additional sampling at three of the 2002-2003 sites and at eight new sites (Mercury Study Extension (MSE)) (DWQ, 2007). The study was expanded to include waters in the central and western portions of the state. Quarterly (fall, winter, spring and summer) sampling was conducted during both studies to address seasonal variations at the study sites. Parameters collected at the sites included total and monomethyl mercury, sediment mercury analysis, sulfate, dissolved organic carbon, nutrients and physical parameters.

Trace-level mercury sampling (total and monomethyl) was conducted using EPA's Method 1669 (EPA, 1996). This method, together with the EPA's analytical Method 1631E (EPA, 2002), allowed mercury quantitation level (0.5 ng/l) to be four-hundred times lower than the method previously used by DWQ for water and sediment. This water sampling methodology includes the use of clean hands/dirty hands procedures and peristaltic pumping of the sample through Polytetrafluoroethylene (PTFE) tubing into laboratory cleaned and certified Teflon bottles. The method is performance-based with strict adherence to quality assurance procedures including field and laboratory blanks. Brooks Rand LLC in Seattle, Washington performed trace-level analysis and equipment cleaning and certification. This methodology significantly reduces the risk of contamination at these low levels of quantitation.

NPDES Data

National Pollutant Discharge Elimination System (NPDES) data were retrieved to assess the mercury point source load within all of North Carolina. Effluent mercury monitoring was required in North Carolina for facilities with potentially significant impacts to local streams. Both monitored point source load and total point source load were estimated from the dataset. The monitored point source load represents the point source load from facilities where mercury monitoring is required and available. The total point source load is the estimated point source load from all NPDES facilities in North Carolina. Therefore, the monitored point source load serves as a lower bound for estimating the total point source load of mercury in NC.

Starting from September 1, 2003, EPA Method 1631E (or subsequent low-level mercury methods approved by EPA in 40 CFR Part 136) with a quantitation limit of 0.5 ng/l (EPA, 2002) and clean sampling techniques (EPA Method 1669) were required when analyzing for total mercury for facilities that discharge greater than 6% of the stream volume. The requirement affected approximately 155 facilities that have mercury limits and/or monitoring requirements.

EPA Method 245.1 (with a quantitation limit of 200 ng/l) was used by the rest of the permitted facilities.

Atmospheric Deposition Modeling and Monitoring Data

Mercury atmospheric deposition is composed of wet deposition and dry deposition. Mercury wet deposition involves the transfer of mercury from the atmosphere to land or water through precipitation. Several chemical species of mercury exist in ambient air as a result of both natural and man-made emissions and the water-soluble forms of mercury may be scrubbed out of the atmosphere by cloud water or rain and snowfall. For many sensitive surface waters, atmospheric wet deposition constitutes a significant route of mercury input. Dry deposition of particles and gases occurs by complex processes such as settling, impaction, and adsorption. Dry deposition processes also contribute to the overall rate of atmospheric deposition. Together, these phenomena can contribute to raise methylmercury levels in fish in mercury-sensitive waters (http://daq.state.nc.us/toxics/studies/mercury/wet_dep.shtml).

Monitoring data are available for wet deposition through the collection and subsequent analysis of rainfall for total mercury concentration. The DAQ has operated two sites for measurement of mercury in rainfall since 1996. Both deposition monitoring sites are in the eastern part of the state near mercury-sensitive waters; one at Pettigrew State Park on the shores of Phelps Lake in Washington County (NC42), and the other at Waccamaw State Park in Columbus County (NC08). Data were also collected at Candor in Montgomery County (NC26) during a shorter period of time. Data collected from these stations are provided to the National Atmospheric Deposition Program Mercury Deposition Network (NADP-MDN: <http://nadp.sws.uiuc.edu/MDN/>) to aid in the identification of geographical and temporal trends in mercury deposition across the U.S. In this TMDL study, we also looked at wet deposition recorded at Great Smoky Mountains National Park-Elkmont in Sevier County, Tennessee (TN11, close to the border between Tennessee and North Carolina). Rainfall is collected weekly in a bucket sampler and sent to a laboratory for quantitative analysis. Mercury levels are measured using EPA Method 1631E for total mercury analysis and undergo full quality assurance/quality control procedures before being reported.

No monitoring data are available for dry deposition of mercury in North Carolina.

In this TMDL study, mercury atmospheric deposition (including both wet and dry deposition) is assessed using the CMAQ modeling system, which is developed and maintained by EPA Office of Research Development and analyzed and processed through NC Division of Air Quality. For the purpose of this TMDL study, the CMAQ model (version 4.71), the emissions data, and the meteorological simulations for the entire year of 2005 were provided by EPA. Model runs were performed by DAQ to estimate nonpoint source (air deposition) loading of total mercury from in-

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state, regional, and global sources. Detailed information about model setup and scenario runs are provided in Appendix B.

The CMAQ model, designed to simulate various chemical and physical processes that are important for understanding atmospheric trace gas transformations and distributions, was initially released to the public by EPA in 1998. CMAQ has been extensively used by EPA and the states for air quality management analyses, by the research community for studying relevant atmospheric processes, and by the international community in a diverse set of model applications (<http://www.epa.gov/asmdnerl/ModelDevelopment/index.html>).

3. NORTH CAROLINA WATER QUALITY STANDARDS

North Carolina water quality standards are state regulations or rules that protect lakes, rivers, streams and other surface water bodies from pollution. The rules are in Title 15A of the North Carolina Administrative Code (NCAC). These rules include beneficial use designations (classifications) and numeric levels and narrative statements protective of the use designations.

The fresh surface water quality standards applicable to the waters covered in this mercury TMDL (15A NCAC 02B .0211) states:

- (1) *Best Usage of Waters: aquatic life propagation and maintenance of biological integrity (including fishing and fish), wildlife, secondary recreation, agriculture and any other usage except for primary recreation or as a source of water supply for drinking, culinary or food processing purposes;*
- (2) *Conditions Related to Best Usage: the waters shall be suitable for aquatic life propagation and maintenance of biological integrity, wildlife, secondary recreation, and agriculture. Sources of water pollution which preclude any of these uses on either a short-term or long-term basis shall be considered to be violating a water quality standard;*

North Carolina has also adopted water column criteria for mercury of 0.012 µg/l for fresh surface waters. The water column criterion was derived using the 1984 U.S. Food and Drug Administration (FDA) fish tissue action level and divided by a bioconcentration factor for the chemical. The FDA action level was derived for methylmercury in fish. Most of the mercury in fish is methylmercury and mercury is readily methylated by both aerobic and anaerobic bacteria in fish mucus, liver, and intestines, the FDA fish tissue action level for methylmercury is therefore used to derive the surface water quality standard for total mercury.

Assessment Methodology for 303(d) List

The water quality standards discussed above were used to assess water quality conditions for mercury in North Carolina. The definition of fishing includes fish consumption (15A NCAC 02B .0202). The fish consumption use is assessed based upon fish consumption advisories. Fish consumption advisories are issued by NC Department of Health and Human Services. These public advisories are based upon review of fish tissue data collected by DWQ.

4. NORTH CAROLINA'S APPROACH

4.1. Statewide Approach

In North Carolina, a statewide fish consumption advisory exists for mercury in Largemouth Bass. This advisory was set because mercury concentrations in Largemouth Bass exceed the state's fish consumption advisory action level across the entire state.

Based on the fish tissue data collected by DWQ from 1990 to 2008, the average fish tissue concentration for total mercury in Largemouth Bass was 0.52 mg/kg, much higher than the state's fish consumption advisory action level of 0.4 mg/kg. In addition, mercury concentrations in Largemouth Bass that exceeded the state's fish consumption advisory action level have occurred statewide (Figure 4-1). All waters in North Carolina were listed in Category 5 of the 2010 303(d) list for mercury impairment due to the statewide fish consumption advisory.

Given that the mercury loadings are predominantly from air deposition, and mercury transported by air could reach thousands of miles away from their emission sources, developing a regional or statewide TMDL for mercury would be a reasonable approach (EPA, 2008).

Considerations on Eco-regional, Basin-wide, and Statewide Approaches

Dividing the entire state into multiple eco-regions or addressing mercury loading based on individual river basins are recognized as alternatives to statewide approach.

Although elevated Largemouth Bass mercury concentrations occur statewide, most of the elevated mercury concentrations occur in the eastern part of the state, within Coastal Plains Ecoregion (Figure 4-1, Middle Atlantic Coastal Plain and Southeastern Plains). The highest mercury concentrations of Largemouth Bass have been found in the southernmost part of the state in the Lumber River Basin, with mercury concentrations reaching a maximum of 3.6 mg/kg. The Sandhills Ecoregion, which includes the upper reaches of the Lumber River Basin in Scotland, Richmond, Hoke, and Moore counties, also holds numerous Largemouth Bass samples that are well above the state's fish consumption advisory action level. The data shows that Largemouth Bass mercury concentrations tend to be higher in the Coastal Plains Eco-region than in the Mountains (Figure 4-1, Blue Ridge) and Piedmonts.

However, streams spanning through multiple eco-regions may have fish swimming across the boundaries of an individual eco-region; river basins containing different fish populations appear to be more manageable units than eco-regions for addressing mercury.

Among the seventeen river basins in North Carolina, Largemouth Bass mercury data were collected in the twelve major river basins. Relatively higher Largemouth Bass mercury levels were found in the Lumber (averaged at 1.03 mg/kg), Pasquotank (averaged at 0.64 mg/kg) and Cape Fear (averaged at 0.59 mg/kg) river basins, while relatively lower mercury concentrations were found in the Catawba (0.17 mg/kg) and French Broad (0.23 mg/kg) river basins (Figure 4-2). However, not all river basins that are located in the coastal plains have much higher Largemouth Bass mercury concentrations than those located in the mountains and piedmont areas. The Yadkin (average Largemouth Bass mercury concentration = 0.34 mg/kg) and Broad (0.32 mg/kg) river basins (mostly in the piedmont area) have average Largemouth Bass mercury concentrations similar to those found in the Tar-Pamlico (0.40 mg/kg) and White Oak (0.37 mg/kg) river basins (mostly in the coastal plains). In summary, Largemouth Bass mercury concentrations are in general higher in river basins residing in the coastal plains than those located in the mountain and piedmont regions.

Based on the spatial pattern found in Largemouth Bass mercury, dividing the entire state into multiple river basins and calculating a TMDL for each river basin seemed a natural choice. Development of multiple TMDLs based on a basin-wide approach does not seem to be an effective strategy, however, since mercury sources are dominated by atmospheric deposition. Minnesota took an eco-regional approach for their mercury TMDL (Minnesota Pollution Control Agency, 2007), however, by assuming mercury deposition to be uniform across the state, Minnesota eventually chose the greater regional reduction goal as an overall statewide emissions goal. Therefore, a statewide mercury TMDL, which protects the most sensitive water bodies within the State would help inform restoration efforts to remove the statewide fish consumption advisory.

In addition to Largemouth Bass, elevated mercury concentrations were also observed in other fish species whose spatial patterns differed from those observed in Largemouth Bass. For example, the average fish tissue mercury concentrations observed in Walleye was 0.46 mg/kg. The walleye samples were collected in the western part of the state in the mountains and piedmont. The average mercury concentration in Walleye was 0.63 mg/kg in Lake Fontana (in the mountains) and 0.96 mg/kg in Lake Santeetlah (in the mountains), similar to the mercury level found in Largemouth Bass in the river basins of Lumber, Pasquotank and Cape Fear (mostly in the coastal plains). In contrast with the spatial pattern observed in Largemouth Bass, mercury concentrations observed in Golden Redhorse (*Moxostoma erythrum*) appeared to be higher in the river basin of French Broad (in the mountains) than those in Roanoke, Tar-Pamlico and Neuse (coastal plains) (Figure 4-3). Therefore, no universal spatial pattern of fish mercury concentration was found for all the fish species collected in the DWQ fish mercury database, likely (or partly) due to the differences in fish habitat preferences and sample sizes.

The higher mercury concentration observed in Largemouth Bass in the eastern part of NC is likely due to enhanced mercury methylation and bioaccumulation processes in the local environments (discussed in Section 5.3), rather than higher mercury loading.

As discussed in Section 5, so far no linkages between elevated fish mercury concentrations and local large water and air sources were identified. Sackett *et al.* (2010) found that lower tissue mercury and higher tissue selenium concentrations were measured in fish collected near power plants. Fish tissue mercury concentrations will be continually monitored and evaluated by DWQ to investigate potential local impacts of point sources in NC waters. In the case of locally elevated fish mercury concentrations that are caused by local sources, DWQ will develop a site-specific mercury TMDL as needed.

A Phase I mercury TMDL study was completed in 1999 for the Lumber River Basin in North Carolina (http://portal.ncdenr.org/c/document_library/get_file?uuid=a97c560f-e3d2-4d01-adeb-f968e6faf199&groupId=38364), the quantification of point source loading was limited at the time by the high quantitation limit (200 ng/l) of the analytical method (EPA Method 245.1) used to monitor effluent mercury concentrations. A regional air quality model study was proposed to be included in Phase II of the TMDL but has not been conducted. This statewide mercury TMDL, which addresses the shortcomings of the Phase I mercury TMDL for Lumber River Basin, will override the previously developed mercury TMDL.

In addition to freshwater fish species, some estuarine and saltwater predator fish also have high mercury levels. For example, King Mackerel caught in the coastal Atlantic Ocean off the North Carolina coast have an average mercury concentration of 0.95 mg/kg. This mercury TMDL covers all waters within North Carolina. However, mercury sources for high mercury saltwater fish species that travel through or live predominately in the coastal oceans off the coast of North Carolina are likely different from those within North Carolina waters. As a result, ocean waters off the North Carolina coast are excluded from this TMDL.

A statewide, universal mercury TMDL, which conservatively considers the necessary mercury reduction goal to remove fish consumption advisory across the state, is appropriate for mercury TMDL development in North Carolina.

North Carolina Mercury TMDL

September 13, 2012

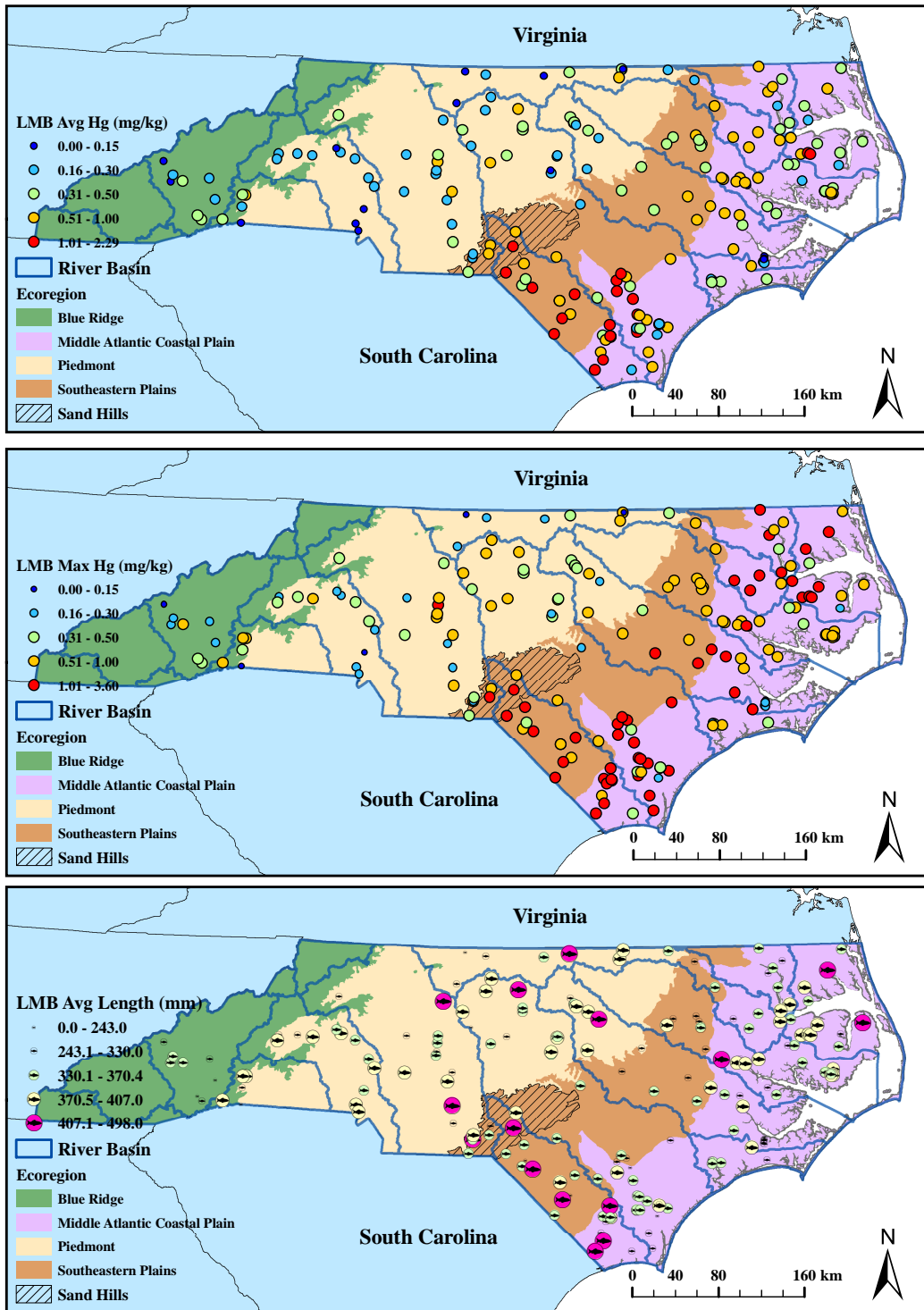


Figure 4-1. North Carolina distributions of station-averaged (top panel) and station-maximum (mid panel) Largemouth Bass fish tissue total mercury concentrations, and station-averaged fish length (bottom panel) during 1990-2008.

North Carolina Mercury TMDL

September 13, 2012

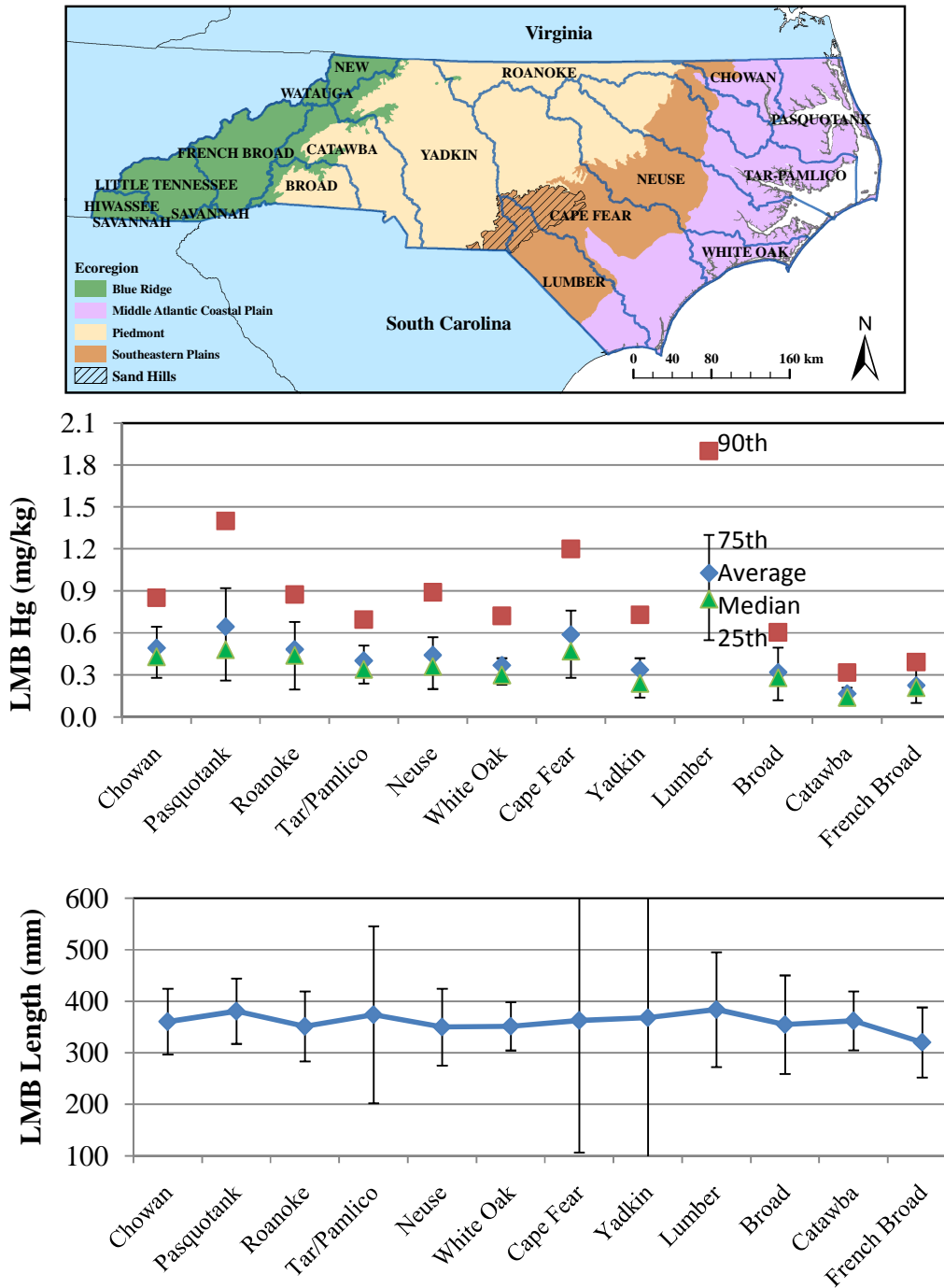


Figure 4-2. River basins and ecoregions in North Carolina (top panel), Largemouth Bass mercury concentrations (middle panel) and average fish length (bottom panel, error bars indicate one standard deviation) in different river basins.

September 13, 2012

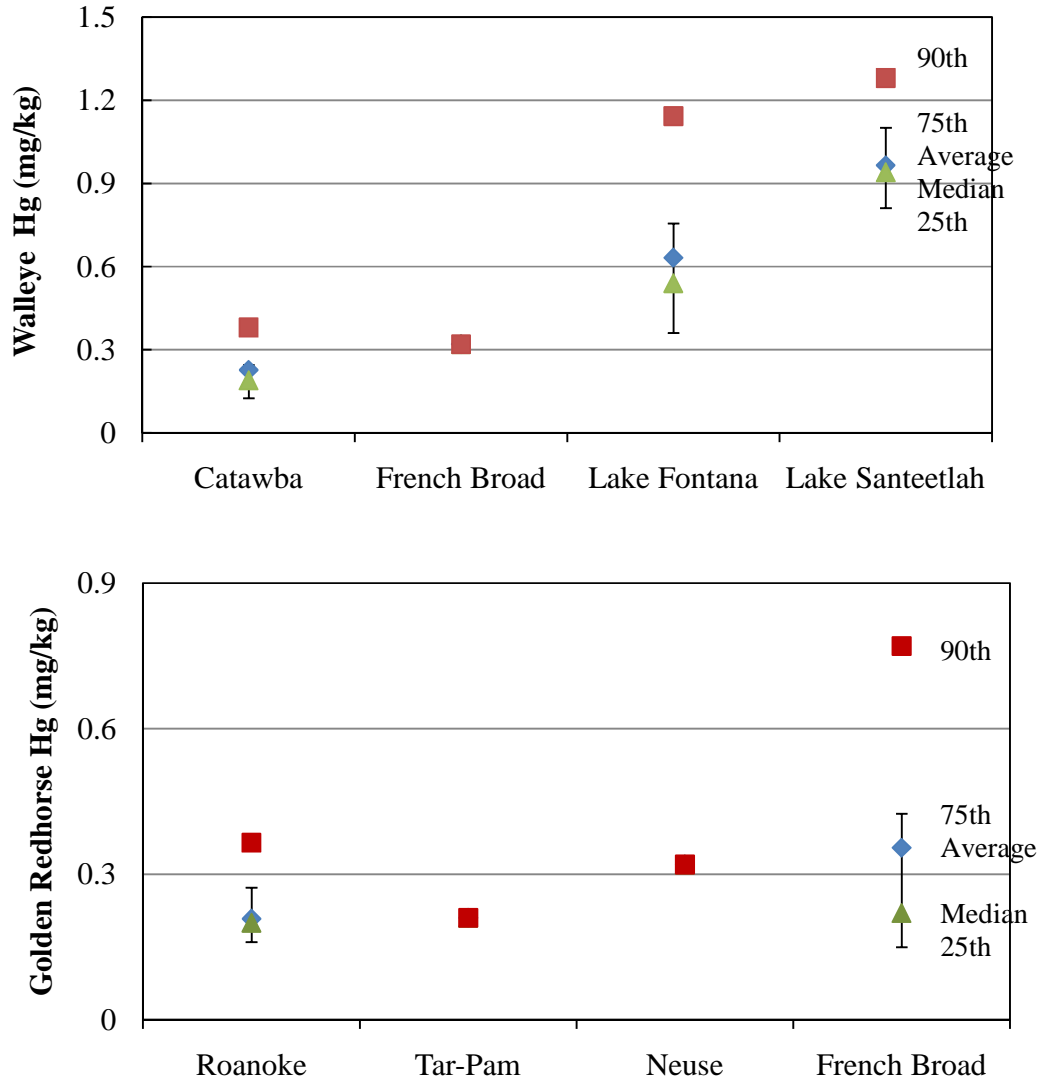


Figure 4-3. Mercury concentration observed in Walleye (upper panel) and Golden Redhorse (lower panel).

4.2. Baseline Year: 2002

The Division of Water Quality selected calendar year 2002 as the baseline year for the North Carolina mercury TMDL. In June 2002, the N.C. General Assembly enacted Session Law 2002-4 (the “Clean Smokestacks Act”), which required significant actual emissions reductions from coal-fired power plants in North Carolina. As a result, 2002 has become a well-established baseline for mercury emissions inventories in North Carolina. Since then, mercury air emissions from sources in North Carolina have significantly declined.

In addition, global emissions stayed relatively stable from 1990-2005 (Pacyna and Pacyna, 2002; Pacyna et al., 2006). The year of 2002 represents the end of a period when mercury emissions and fish concentrations were most likely in a steady state.

4.3. Water Quality Target

4.3.1 Applicable Water Quality Target

The ultimate goal of this TMDL is to have safe-level mercury concentrations in fish caught in North Carolina waters so that the fish consumption advisory in NC can be removed. No numeric fish tissue water quality standard for mercury is established in North Carolina, a fish tissue mercury target is hence needed for this TMDL. The following are some fish tissue criteria used by national or North Carolina agencies.

EPA/FDA recommended fish tissue criterion for mercury

The human health Ambient Water Quality Criterion for methylmercury recommended by U.S. EPA and FDA is 0.3 mg methylmercury (MeHg) /kg fish (<http://www.epa.gov/waterscience/criteria/methylmercury/pdf/mercury-criterion.pdf>). The EPA/FDA published a joint federal advisory for mercury in fish in 2004 using this criterion (<http://water.epa.gov/scitech/swguidance/fishshellfish/fishadvisories/publicinfo.cfm>).

The methylmercury water quality criterion is a concentration in fish tissue. It was calculated using the criterion equation in the 2000 Human Health Methodology (<http://www.epa.gov/waterscience/criteria/humanhealth/method/complete.pdf>) rearranged to solve for a protective concentration in fish tissue rather than in water.

The resulting tissue residue criterion is 0.3 mg MeHg/kg fish. This is the concentration in fish tissue that should not be exceeded based on a total fish and shellfish consumption-weighted rate of 0.0175 kg fish/day, which is equivalent to about 4 oz fish per week or 19 oz per month. EPA strongly encourages States and authorized Tribes to develop a water quality criterion for

methylmercury using local or regional data rather than the default values if they believe that such a water quality criterion would be more appropriate for their target population.

Food and Drug Administration also has an action level of 1 ppm methylmercury in commercially caught fish. An action level represents a limit at or above which FDA will take legal action to remove products from the market. This action level is considered by EPA and FDA to be inappropriate for establishing local advisory needs and EPA does not support its use for that purpose (<http://www.gpo.gov/fdsys/pkg/FR-2001-01-08/pdf/01-217.pdf>) (66 FR 1344, January 8, 2001).

North Carolina fish consumption advisory action level for mercury

The North Carolina Department of Health and Human Services considers fish that have on average methylmercury levels between 0.1 to 0.3 mg MeHg/kg as fish low in methylmercury, and recommends that women of childbearing age and children less than 15 years of age eat up to two meals a week of fish low in methylmercury. NC DHHS considers average methylmercury levels in fish tissue of 0.4 mg/kg as potentially unsafe for women of childbearing age and children (Williams, 2006).

Using the data from the Faroes Islands study and EPA standardized equations and recommended doses, the North Carolina Occupational and Environmental Epidemiology Branch of NC DHHS determined the action level for issuing fish advisories in North Carolina is 0.4 mg methylmercury/kg fish. If the average methylmercury level for a given species at a given location is 0.4 mg/kg or higher then no consumption is recommended for women of childbearing age and children less than 15 years of age and no more than one meal a week for the general public.

4.3.2 Water Quality Target for Mercury TMDL

The EPA and FDA fish tissue mercury criterion of 0.3 mg methylmercury / kg fish is selected as the target level for this TMDL development. Since fish tissues were monitored for total mercury in North Carolina, and studies have shown that the majority of mercury concentrations in fish tissues are in the form of methylmercury, the 0.3 mg MeHg / kg fish tissue mercury target is applied to total mercury in fish tissues in this TMDL study.

The water quality target of 0.3 mg/kg mercury in fish is also consistent with the NC DHHS action level for fish consumption advisory. It is the upper bound of mercury concentration in fish that NC DHHS considers as low in mercury, and hence a fish consumption advisory is not issued.

To demonstrate that meeting the fish tissue target will achieve water quality standards (40 CFR 130.7(c)), the bioaccumulation factor (BAF) was estimated from the data of DWQ Eastern

Regional Mercury Study and Mercury Study Extension (see Section 5.3). The estimated BAF ranged between 0.6 - 5.4×10^5 l/kg. The fish tissue mercury target of 0.3 mg/kg would be equivalent to a total mercury concentration target of 0.6 - 5 ng/l in surface waters. Therefore, by meeting the target for this TMDL, the numerical water column criterion for total mercury in North Carolina (12 ng/l) will be met simultaneously.

Standardized-length Predator Fish

Various studies have shown that fish MeHg concentrations varied greatly with fish species and their corresponding trophic levels (Sackett et al., 2009). Predator fish with longer life spans tend to bio-accumulate more MeHg inside their bodies. As described in Section 2.2, based on the fish data collected 1990-2008, Largemouth Bass is the most commonly found predator fish in the waters of North Carolina, representing approximately thirty-six percent of the fish tissue mercury data in the entire database and sixty-nine percent of fish samples that are of concern (i.e. fish having average fish mercury concentration > 0.4 mg/kg). In addition, data available for Largemouth Bass are widely spread across the state, while other fish species of concern were found typically within a smaller region in NC.

For the southeastern region of North Carolina, due to a higher mercury methylation potential (discussed under Section 5.3), the general public likely faces a higher health risk from fish consumption. A creel survey of the recreational fishery on the Cape Fear River (a typical southeastern NC river) showed that one third of the anglers targeted catfish. A smaller percentage of anglers targeted other fish species: Largemouth Bass (16 percent), Sunfish (12 percent), Striped Bass (4 percent), American Shad (2 percent), and Crappie (less than 1 percent). The remaining 31 percent fished for a combination of species or anything they could get. Altogether, Sunfish accounted for 59 percent of the total harvest followed by Catfish, which accounted for 31 percent of the total harvest. Sunfish and Catfish are the two fish species that are most likely (~90%) consumed by local anglers on the Cape Fear River.

North Carolina Wildlife Resources Commission listed the most popular fish species in the coastal region of North Carolina as: Striped Bass, American and Hickory Shad, Largemouth Bass, Crappie, assorted Sunfish (Redear, Redbreast, Bluegill, Warmouth and Pumpkinseed), White and Yellow Perch, and Channel, Blue, White and Flathead Catfish (http://www.ncwildlife.org/Portals/0/Fishing/documents/Coastal_Region_Fishing_booklet.pdf). A comparison of the mercury concentrations in these fish species (from the 1990-2008 DWQ fish monitoring dataset) is provided in Table 4-1.

As shown in Table 4-1, the mercury concentrations in the most popular and most likely consumed fish species in eastern North Carolina are usually much less than those found in Largemouth Bass. It is reasonable to expect that mercury concentrations in the most likely

consumed fish species such as Sunfish and Catfish will be lower than the target level once the Largemouth Bass mercury concentrations decline below the target level as a consequence of mercury loading reductions. As a result, Largemouth Bass was selected to be the target fish species for this TMDL.

Within a specific fish species, mercury concentrations tend to be higher in larger fish. Figure 4-4 shows that mercury concentrations in Largemouth Bass usually vary as a function of the size of the fish. To account for this size-dependency of mercury concentrations, the mercury concentration is compared for fish of the same size. To avoid biases caused by different prevailing fish sizes at different sampling time and place, we calculated the standardized-length Largemouth Bass mercury concentration for each sampling event. Standardized length fish mercury concentrations were also used in other regional mercury TMDLs approved by EPA (Minnesota Pollution Control Agency, 2007; Northeast, 2007; New Jersey Department of Environmental Protection, 2009). The standardized Largemouth Bass length in this TMDL was calculated as the median Largemouth Bass length in all the samples, which is 353 mm. A standardized-length fish is also the fish one would most often encounter in the environment (highest sample frequency, Figure 4-4).

A standardized-length Largemouth Bass mercury concentration was calculated for each survey during 1990-2008. Multiple fish samples were usually collected during each survey. Linear regression was used to generate the relationship between fish length and mercury (Hg) concentration for that particular survey. A standardized-length Largemouth Bass mercury concentration was then calculated as the mercury concentration that corresponds to 353 mm fish length in the linear function. This exercise brought over 2000 Largemouth Bass mercury concentration data down to 172 data points of standardized-length Largemouth Bass mercury concentrations. The standardized-length Largemouth Bass mercury concentration data were then used in assessing the reduction goals needed to meet the TMDL target.

North Carolina Mercury TMDL

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Table 4-1. Mercury concentrations (mg/kg) of largemouth bass, catfish and sunfish species

Fish Species	Fish Hg Concentration (mg/kg)		
	Average	75 th Percentile	90 th percentile
Largemouth Bass (LMB)	0.52	0.68	1.10
Warmouth (Sunfish)	0.44	0.56	0.73
Yellow Perch	0.43	0.52	0.67
Flathead Catfish	0.34	0.28	0.55
Channel Catfish	0.24	0.30	0.47
Redbreast (Sunfish)	0.22	0.30	0.46
Redear (Sunfish)	0.22	0.30	0.44
White Perch	0.21	0.27	0.38
White Catfish	0.21	0.26	0.44
Black Crappie	0.20	0.27	0.45
Blue Catfish	0.20	0.26	0.33
Flier (Sunfish)	0.20	0.24	0.36
Green Sunfish	0.19	0.26	0.33
Hickory Shad	0.19	0.22	0.27
Bluegill (Sunfish)	0.18	0.23	0.34
Striped Bass	0.18	0.20	0.26
Pumpkinseed (Sunfish)	0.15	0.23	0.31
White Crappie	0.06	0.09	0.10
American Shad	0.05	0.05	0.06

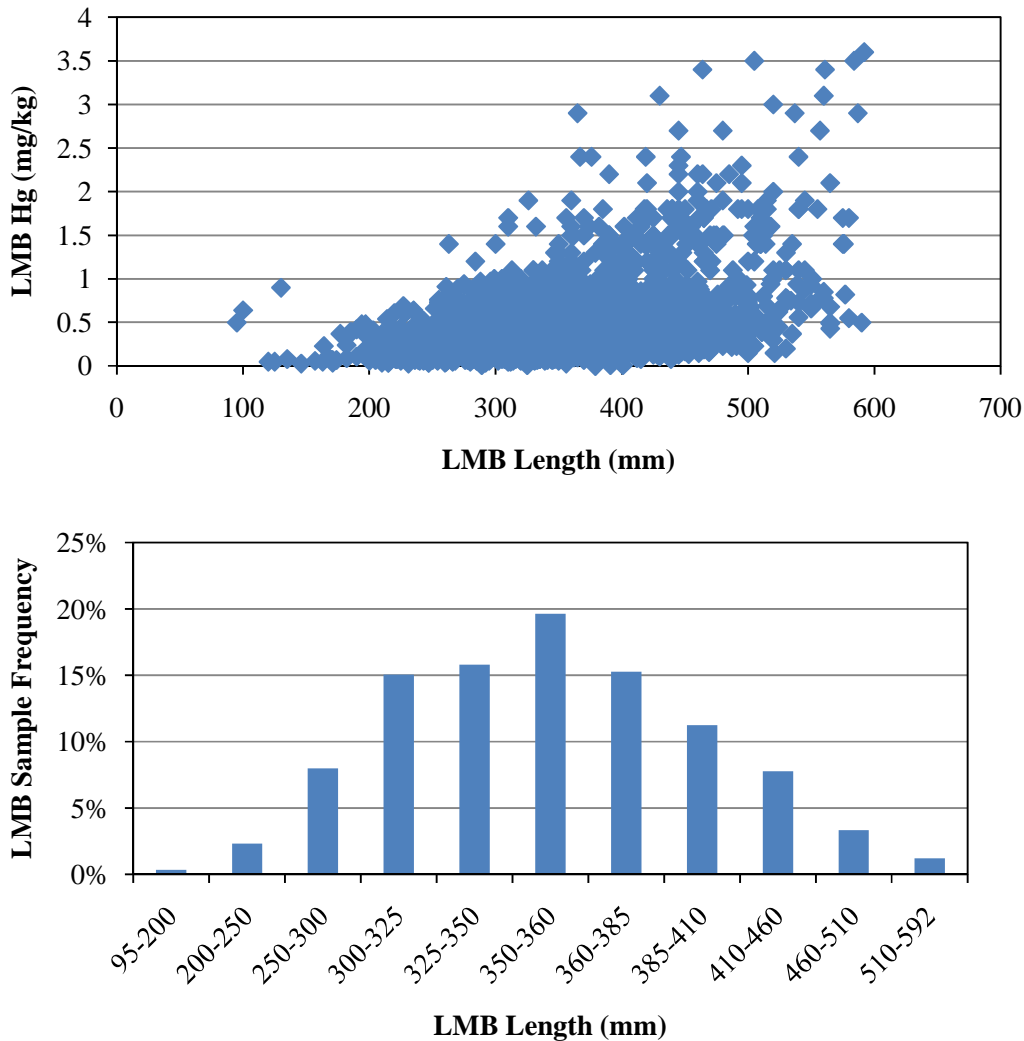


Figure 4-4. The relationship between Largemouth Bass mercury concentrations and the fish lengths (upper panel) and the Largemouth Bass sample frequency distributions at different Largemouth Bass size groups (lower panel).

Applying the Target Level to the 90th Percentile Fish concentration

To achieve water quality standards and protect water bodies from impairment, an appropriate statistic must be selected to meet the target level of 0.3 mg/kg. Following the practices of statewide mercury TMDL development in Minnesota and New Jersey, the 90th percentile of the standardized-length predator fish samples was selected to meet the target level mercury

concentration. The selection of the 90th percentile of samples is also consistent with the assessment guidance by the EPA (i.e. no more than 10% of the samples can exceed the standard).

Rather than using a measure of central tendency, such as the mean or the median, the 90th percentile of Largemouth Bass samples was selected to provide greater protection. The 90th percentile is calculated from standardized-length Largemouth Bass mercury data, which has avoided the selection of rare incidences from original samples of large-sized, long-living fish. Achieving the target level for the 90th percentile of standardized-length Largemouth Bass ensures that the smaller predator fish and fish at lower trophic levels will meet the target level.

Due to significant inter-annual variations on sampling sites, sampling numbers, weather, and natural variability in fish populations, multi-year data are used to provide the assessment of the baseline year fish mercury conditions. Figure 4-5 shows that although median or average standardized-length largemouth bass mercury concentrations varied considerably within 1990-2008 period, the 90th percentile stayed relatively the same in the 1990's and 2000's. The 90th percentile of the standardized-length Largemouth Bass mercury concentrations in 1990-2008, which is 0.9 mg/kg, is selected to represent the baseline fish mercury condition.

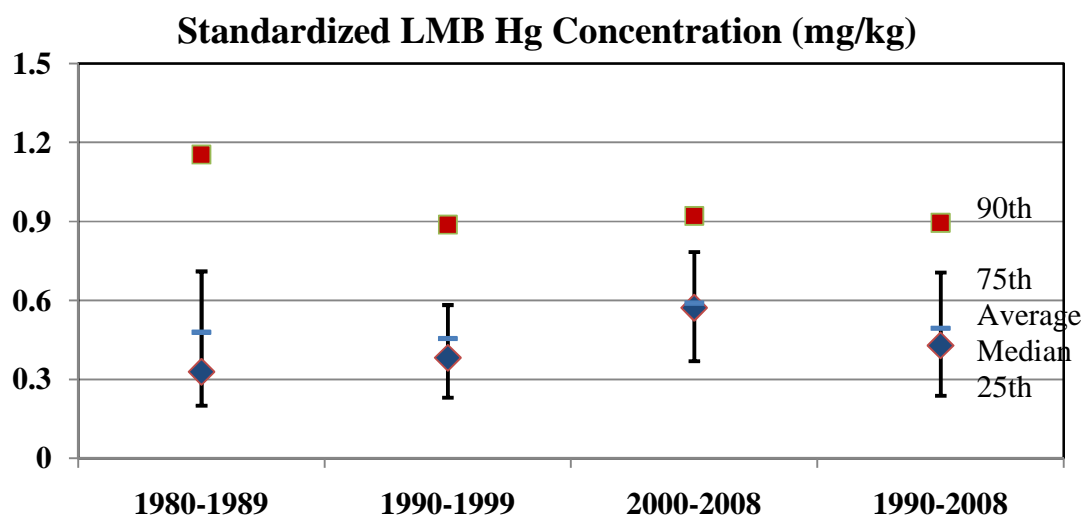


Figure 4-5. Decadal variations of standardized largemouth bass mercury concentrations.

5. MERCURY SOURCE ASSESSMENT AND TRENDS

5.1 Sources of Mercury in North Carolina Fish

As discussed in the preceding sections, mercury is a natural element but human activities have increased the amount of mercury that is biologically available.

Fish mercury concentration is known to be affected by three major consecutive processes: 1. mercury deposition/transportation to aquatic systems; 2. conversion to biologically active methylmercury (mercury methylation); and 3. bioaccumulation in aquatic systems (Wiener et al., 2006; Peterson et al., 2007).

Mercury in fish mostly comes from mercury emitted into the air, which is deposited into waters or onto adjacent lands, where it is washed off into surface waters when it rains. For most aquatic ecosystems, the primary source of mercury is atmospheric deposition of inorganic mercury (about 0.3 to 30 $\mu\text{g m}^{-2} \text{yr}^{-1}$; EPA, 1997). This includes both wet deposition (rainfall) and dry deposition and is affected by a series of complex factors including mercury air emission sources, local or regional meteorology and atmospheric chemistry. Under the Clean Water Act, atmospheric deposition of mercury into surface waters is regarded as a nonpoint source.

Some mercury is discharged directly into surface waters from industrial and municipal point sources, although the amounts are usually very small compared to air emission sources.

5.2 Trends in Mercury Emissions and Deposition

Mercury Emission Sources and Trend

Three types of mercury air emission sources were defined in the Mercury Study Report to Congress (EPA, 1997):

- *Natural mercury emissions* -- the mobilization or release of geologically bound mercury by natural processes, with mass transfer of mercury to the atmosphere;
- *Anthropogenic mercury emissions* -- the mobilization or release of geologically bound mercury by human activities, with mass transfer of mercury to the atmosphere; or
- *Re-emitted mercury* -- the mass transfer of mercury to the atmosphere by biologic and geologic processes drawing on a pool of mercury that was deposited to the earth's surface after *initial* mobilization by either anthropogenic or natural activities.

The magnitude of the natural emissions versus re-emissions is poorly understood because it is usually not feasible to distinguish between natural emissions and re-emissions. Deposition to the surface, whether land or sea, is complicated by the fact that deposited mercury can be re-emitted

to the atmosphere as elemental mercury. In addition, there are few measurements available and current estimates are to a large extent extrapolated from a few data points and constrained by global mass balance estimates. Studies suggested that ocean emissions were between 770-2300 tonnes / yr, volcano emissions were between 20-447 tonnes/yr, emissions from soil were between 500-3200 tonnes / yr, emissions from vegetation were between 850-2000 tonnes / yr, and up to 100 tonnes / yr for emissions from fires (<http://www.geiacenter.org/>; Fitzgerald, 1986; Pacyna, 1986; Nriagu, 1989; Lindberg et al., 1998; Ebinghaus et al., 1999; Nriagu, 1999). Pirrone (et al., 2010) reported that on an annual basis, natural and re-emission sources account for 5207 tonnes of mercury released to the global atmosphere. Re-emission estimates, on a global scale are on the order of 1/3 to 1/2 of the combined anthropogenic and natural emissions.

The quantities of mercury in environmental reservoirs (i.e. the global pool) in both the preindustrial and present day cycles are uncertain. However, the ratio between present-day and preindustrial mercury deposition suggested that human activities, such as coal burning, have increased the amount of mercury cycling among the land, atmosphere, and ocean by a factor of three to five (Selin, 2009).

For anthropogenic mercury emissions, the Arctic Monitoring and Assessment Programme (AMAP) analyzed global mercury inventories from 1990, 1995, 2000 and 2005 (Pacyna and Pacyna, 2002; Pacyna et al., 2006) and reported that the level of mercury emissions in the air on the global scale has been relatively stable from 1990-2005, although contributions from Europe and North America were reduced whereas emissions in Asia were increased. They estimated the global emission inventory for anthropogenic mercury to be around 1921 tonnes in 2005. A slightly higher 2005 value (2320 tonnes per year) was estimated by Pirrone (et al., 2010). In 2005, AMAP estimated that the Asian countries contributed about 67 percent to the global mercury emissions to air from anthropogenic sources, followed by North America and Europe (AMAP/UNEP, 2008).

Depending on the form of the mercury emitted, the location of the emission source, and the weather, atmospheric mercury can be transported over a range of distances before it is deposited, potentially resulting in deposition on local, regional, continental and/or global scales. EPA has estimated that about one third of U.S. emissions are deposited within the contiguous U.S. and the remainder enters the global cycle. Current estimates are that less than half of all mercury deposition within the U.S. comes from U.S. sources, although deposition varies by geographic location.

North Carolina Mercury TMDL

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Top 50 Mercury Air Emission Point Sources, 1998 - 1999

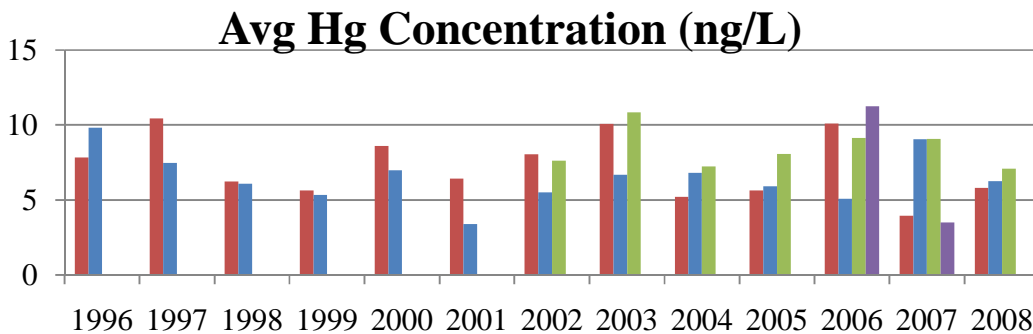
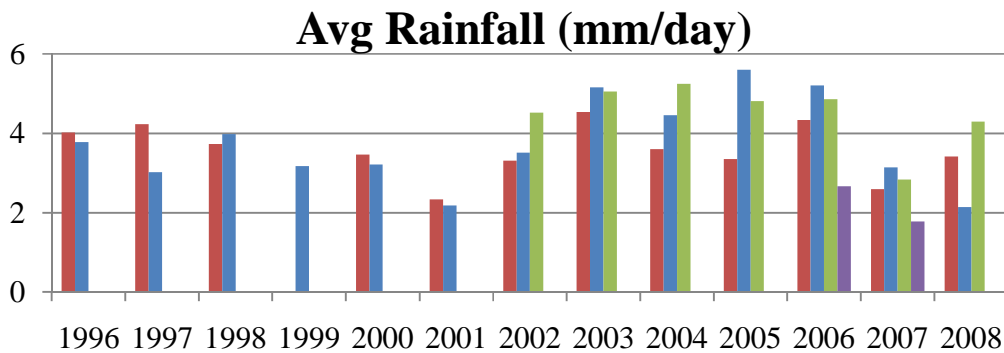
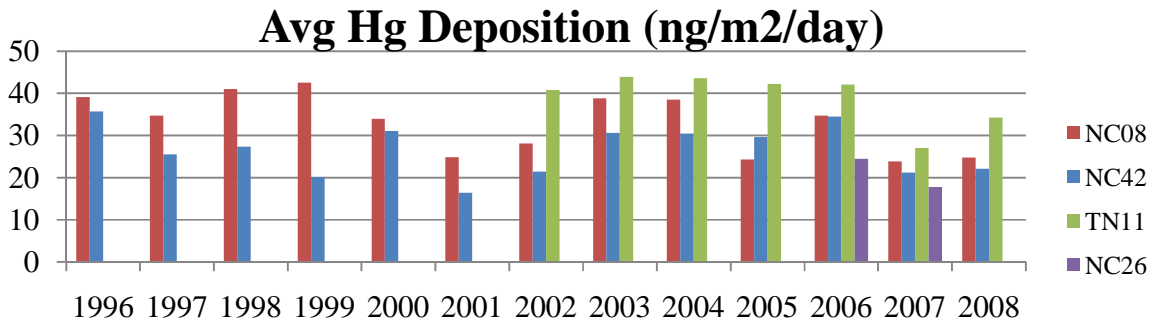
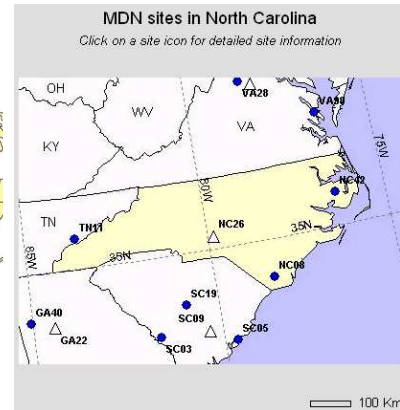
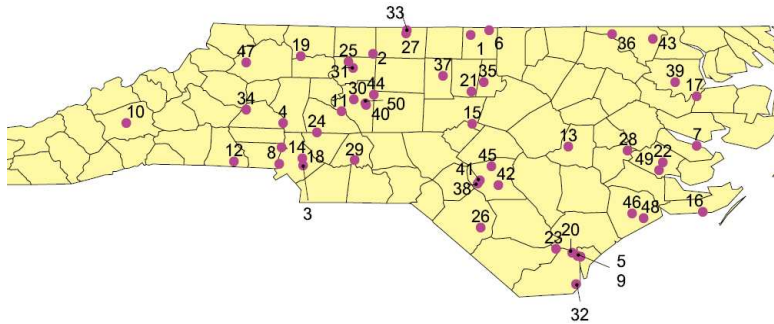


Figure 5-1. Mercury air emission sources and wet deposition data from National Atmospheric Deposition Program-Mercury Deposition Network.

North Carolina Mercury TMDL

September 13, 2012

In North Carolina, as shown in Figure 5-1, air emission sources are located across the entire state. In 2002, approximately 5,300 lbs of mercury were emitted from permitted stationary sources of air pollution in North Carolina. Sixty-six percent of the emissions were attributed to coal-fired utility electric generating units (EGUs) from the two primary utility companies: Duke Energy Carolinas, LLC and Progress Energy Carolinas, LLC. The remaining 34% of statewide mercury emissions is attributed to various non-EGU industrial coal-fired boilers, steel mills, incinerators, and other sources (Table 5-1).

Table 5-1. NC's mercury emissions from permitted air sources

Source Type	2002 lbs/year	2010 lbs/year	2016* lbs/year	2002-2016 Reduction
Electric Generating	3,500	963	700	80%
Other Point	1,800	881	800	56%
Total	5,300	1,844	1,500	72%

*2016 projected emission include EPA's Electric Generating Units Maximum Achievable Control Technology (MACT) Rules and planned shutdowns/fuel conversions

State Requirements for Emissions Reductions

In June 2002, the N.C. General Assembly enacted Session Law 2002-4 (the "Clean Smokestacks Act"), which requires significant actual emissions reductions from coal-fired power plants in North Carolina. Under the act, nitrogen oxides (NOx) emissions must be reduced (from 1998 levels) by 77% by 2009 and sulfur dioxide (SO2) emissions by 73% by 2013. A significant co-benefit resulting from the controls being put in place to reduce NOx and SO2 is a reduction in mercury emissions (<http://daq.state.nc.us/news/leg/hg/>).

An important feature of the Clean Smokestacks Act is that North Carolina's two largest utility companies, Duke Energy and Progress Energy, must achieve these emissions cuts through actual reductions at their 14 power plants in the state - not by buying or trading emissions credits from utilities in other states, as allowed under federal regulations. The utilities also cannot sell credits for the emission reductions, ensuring that utilities in neighboring states don't negate the gains achieved in North Carolina by purchasing the rights to increase or to avoid controlling their own emissions.

North Carolina also has two state mercury rules that deserve to be highlighted. They are 15A NCAC 02D .2509, Periodic Review and Reallocations, and 15A NCAC 02D.2511, Mercury Emission Limits. Under 02D .2509, NC Division of Air Quality shall report to the Environmental Management Commission (EMC) information on the regulation of mercury emissions in 2008 and 2012. Based upon the upcoming 2012 report, the EMC will review the state of mercury technology and decide if any rule changes are needed.

Mercury emissions from NC's stationary point sources continue to decline as shown in the table below. Among the fourteen (electric generating) power plants in NC, seven of them are being converted to natural gas or being retired, and additional controls are expected. By the year 2016, mercury air emissions from stationary point sources in North Carolina are expected to be reduced by 72% (Table 5-1).

Monitored Trends in Mercury Deposition in North Carolina

Measurement of long-term deposition from ice cores and lake sediments suggest that in the Northern Hemisphere deposition has increased from pre-industrial levels by a factor of 3 to >10 (Bindler et al. 2001, Schuster et al. 2002). Recent peak deposition probably occurred sometime in the 1970's to 1980's. However emissions from Asia may lead to higher global emission levels in the future.

Wet deposition of mercury is monitored regularly at sites across the U.S. by the National Atmospheric Deposition Program-Mercury Deposition Network (MDN). As discussed in Section 2.2, three sites are located within North Carolina: NC08, NC42, and NC26 (presently inactive) (Figure 5-1). In addition, one station is located within Tennessee but close to its border with North Carolina: TN11. A close examination to the MDN data at these sites reveals that mercury wet deposition appears to be highest at western NC (TN11, mountain area) and lowest in central NC (NC26, piedmont area). These differences are due at least partly to differences in rainfall. However, the inter-annual variations of wet depositional fluxes of mercury are pretty high, undermining the spatial pattern discerned from the data set (data are available at NC08 and NC42 after 1996; at NC26 during 2006-2007; at TN11 after 2002) (Figure 5-1).

Assuming wet deposition of mercury over the entire State of North Carolina could be represented by the average condition of the existing three MDN monitoring stations (NC08, NC42 and TN11), *the total wet deposition within North Carolina was estimated to be around 1533 kg (3373 lbs) during the baseline year of 2002, slightly less than the long-term (2002-2008) average of the annual wet deposition in North Carolina (1639 kg or, 3606 lbs) and that during the year of 2005 (1631 kg or, 3588 lbs).* The inter-annual difference in wet deposition is partly due to the differences in precipitation. The baseline year of 2002 is a relatively dry year

according to its negative average annual 12-Month standardized precipitation index (<-1.5) (<http://www.nc-climate.ncsu.edu/>).

Although the wet deposition of mercury has been widely studied and monitored, limited information is available on the contribution of dry deposition of mercury to total atmospheric mercury deposition. *It has been reported that the the ratio of dry to wet deposition could range between 0.5 and 10*, and vary with season, the form of mercury in local sources, the methods used for dry deposition approximations, and the places of concern (Miller et al., 2005; Sakata and Marumoto, 2005; Lyman et al., 2007).

CMAQ Model-simulated Mercury Deposition in North Carolina

The EPA's CMAQ modeling system (<http://www.cmaq-model.org>) was used to calculate mercury depositional fluxes in North Carolina. A brief description of the CMAQ model can be found at Section 2.2. The model was a special version of CMAQ version 4.71 that included mercury chemistry. The model and associated inputs were used for modeling impacts of the EPA MACT (maximum achievable control technology) rule for EGUs. The version uses gridded area emissions and stack emissions from various sources. EPA ran a national 36 km CMAQ run with GEOS-CHEM boundary conditions and a nested 12 km CMAQ run with boundary conditions from the 36 km run. For this TMDL study, DAQ conducted all model runs at 12 km model domain using the EPA provided 2005 emissions, boundary conditions from the 36 km EPA run, and MCIP meteorological files obtained from EPA. Figure 5-2 shows the 36 km and 12 km model domains. Due to the special model setup, in this TMDL, mercury sources coming from outside the 12 km model domain are referred as *global sources*, sources within the 12 km model domain but outside North Carolina as *regional sources*, and sources within North Carolina as *in-state mercury sources*.

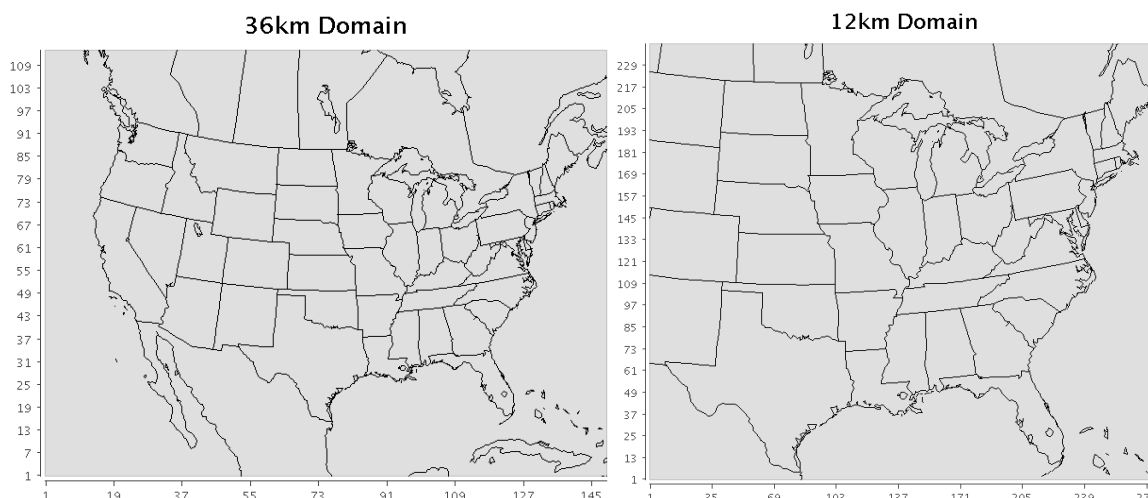


Figure 5-2. CMAQ 36 km and 12 km model domains.

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Using the CMAQ model, *the model-simulated total atmospheric mercury deposition for 2005 is 5,239 kg/yr, with 1588 kg/yr from wet deposition and 3651 kg/yr from dry deposition.* The model-simulated total mercury deposition is uniformly distributed except at several model grids where local air sources (e.g. power plants) exist. The locally elevated air depositional fluxes are likely due to under-predicted horizontal dispersion fluxes in the model in vicinity of local sources. According to Sackett et al. (2010), fish tissue mercury concentrations were found to be lower close to power plants than those farther away from power plants in North Carolina. No linkage between higher air depositional fluxes and locally elevated fish mercury concentrations were found.

The CMAQ-model-simulated wet deposition appears to agree with the estimated wet deposition (1631 kg during 2005) from MDN monitoring data. In addition, the model-simulated ratio of dry to wet deposition appears to fall within the literature-reported range (as discussed under “Monitored Trends in Mercury Deposition in North Carolina” under Section 5.2). Sensitivity model runs were also conducted to assess global, regional and in-state contributions to the total mercury depositional flux within North Carolina. Model results show that *approximately 16% of the total depositions could be attributed to in-state air emissions.* The global contributions were estimated by zeroing out the boundary conditions (*i.e.* here the global is not literally “global”, rather it refers to the contributions from outside of the 12 km model domain). The model suggested that *approximately 70% of the deposited mercury is coming from global sources. The remaining 14% was estimated coming from regional sources* (*i.e.* from other states of the U.S. that are within the 12 km model domain).

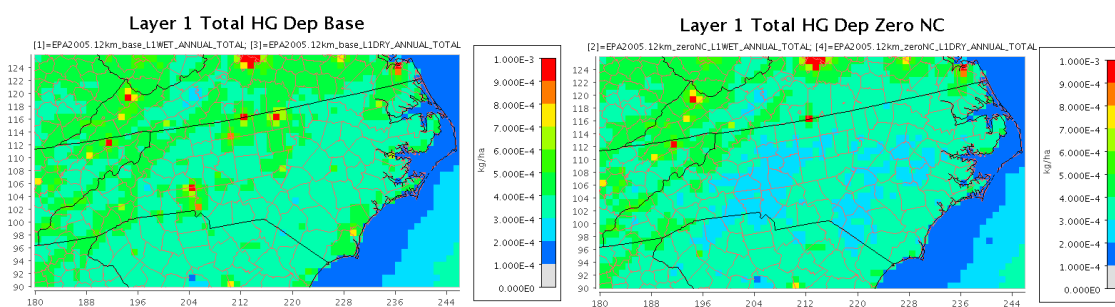


Figure 5-3 CMAQ simulated total mercury deposition in 2005 with all emission sources (left) and with emission sources outside NC (right).

While sensitivity tests were conducted to assess contributions from emission sources of different geographical locations, contributions from natural and anthropogenic sources cannot be readily differentiated with the current CMAQ settings. In fact, as discussed in the previous sections, the

magnitude of the natural emissions and re-emissions is poorly known, most likely because it is usually not feasible to distinguish between natural emissions and re-emissions. For the purpose of this TMDL, due to lack of natural sources such as volcanoes and relatively very low amount of biomass burning within North Carolina, natural emissions (including biogenic and forest fire emissions) are estimated to be approximately 6% of the total air emissions of mercury in North Carolina. Correspondingly, it is assumed that natural sources in North Carolina contribute 6% of the air deposition of mercury caused by NC air emission sources.

5.3 Mercury Methylation and Bioaccumulation

Studies have shown that fish mercury concentration is not only affected by the availability of total mercury in the water column, but also mercury methylation and bioaccumulation in aquatic systems (Wiener et al., 2006; Peterson et al., 2007). Mercury is normally deposited onto water surfaces and land in the form of inorganic mercury and turned into organic methylmercury by biota through the methylation process the methylmercury then biomagnifies up the food chain, where it reaches high concentrations in some of the higher-trophic and longer-lived fish.

A USGS study suggested that mercury concentrations in Largemouth Bass from basins with no mining operations increase as the percentages of evergreen forest and woody wetlands increase, especially where the sampling site is closer to these kinds of land cover. Mercury methylation and bioaccumulation appear more likely to occur in these types of settings (Scudder, et al., 2009).

Table 5-2 shows the percentage of forest and wetland within the river basins in North Carolina, where Largemouth Bass data are available. The percentage of wetlands varies greatly in these river basins, from 0.1% to 28%. The three river basins with highest Largemouth Bass mercury concentrations (Lumber, Pasquotank, and Cape Fear) all have extensive wetlands along their river banks. This suggests that biogeochemical features of the surface water system have a greater influence on the spatial pattern of fish mercury concentration than localized mercury emissions sources.

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Table 5-2. Land cover and Largemouth Bass Hg concentrations in NC river basins (highest three are bolded).

Basin	Forest	Wetland	Year	Source	LMB Hg Conc. (mg/kg)*	
					Average	90 th percentile
Broad	61.0%	2.0%	2001	NLCD	0.32	0.60
	63.3%		1997	NRI		
Cape Fear	38.2%	14.9%	2001	NLCD	0.59	1.20
	56.3%		1997	NRI		
Catawba	55.0%	0.5%	2001	NLCD	0.17	0.32
	43.9%		1997	NRI		
Chowan	46.3%	5.5%	2001	NLCD	0.49	0.85
	54.9%		1997	NRI		
French Broad	75.7%	0.1%	2001	NLCD	0.23	0.39
	46.9%		1997	NRI		
Lumber	25.7%	26.1%	2001	NLCD	1.03	1.90
	60%		1997	NRI		
Neuse	32%	16.8%	2001	NLCD	0.44	0.89
	44.9%		1997	NRI		
Pasquotank	13.8%	23.5%	2001	NLCD	0.64	1.40
	23.9%		1997	NRI		
Roanoke	48.9%	8.7%	2001	NLCD	0.48	0.87
	62.1%		1997	NRI		
Tar-Pamlico	28.5%	15.0%	2001	NLCD	0.40	0.70
	38.2%		1997	NRI		
White Oak	23.0%	28.2%	2001	NLCD	0.37	0.72
	30.5%		1997	NRI		
Yadkin	52.7%	1.3%	2001	NLCD	0.34	0.73
	50.0%		1997	NRI		

NLCD: National Land Cover Database from Multi-Resolution Land Characteristics Consortium

NRI: National Resources Inventory from Natural Resource Conservation Service

*Largemouth Bass (LMB) Hg concentrations were calculated from the 1990-2008 DWQ fish mercury database.

Based on data from Eastern Regional Mercury Study (DWQ, 2003) and Mercury Study Extension (DWQ, 2007), the average methylmercury concentrations were highest in the Waccamaw and Cashie Rivers (average MeHg = 1.64 ng/l in both systems), followed by Black River (0.54 ng/l) and Lumber River (0.46 ng/l). Due to the lack of spatial representation within river basins, the ERMS and MSE study results are presented here as averages at each sampling station or combined close-by sampling stations that are located within the same stream (or lake).

The high-MeHg systems are all characterized as having relatively high mercury methylation rates (Figure 5-4). In these systems, MeHg concentrations were usually below 1.0 ng/l, but

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spikes of high MeHg concentrations sometimes occur, suggesting event-driven mercury methylation mechanisms may take place in such systems.

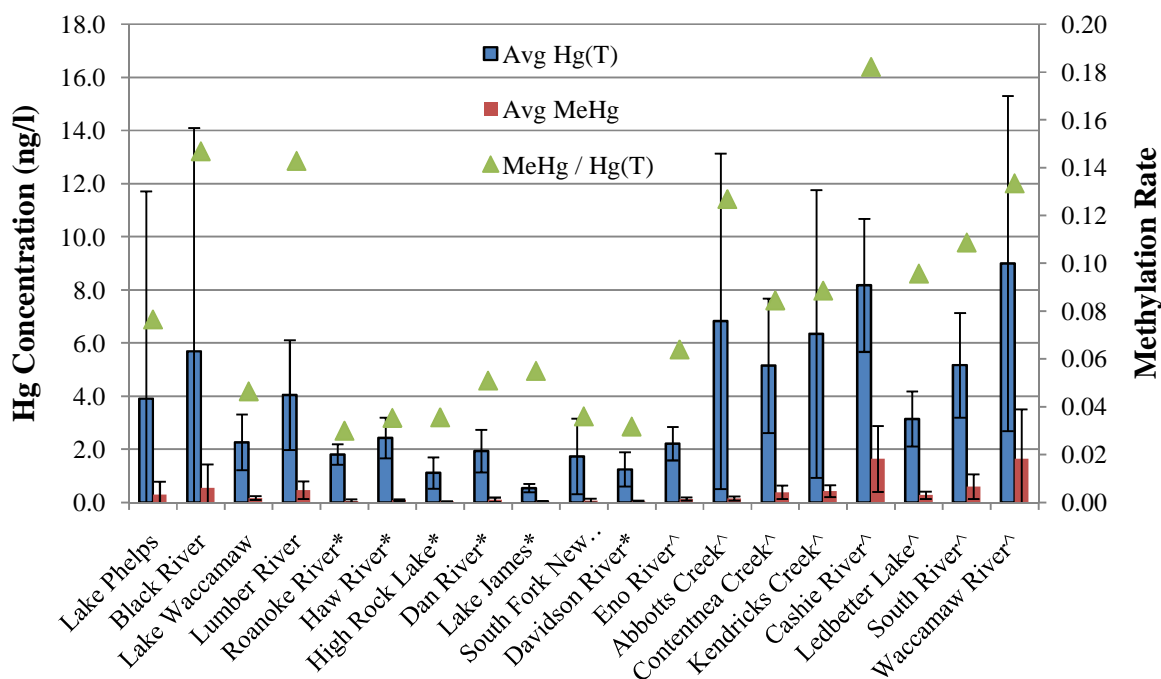


Figure 5-4. Average mercury concentrations and methylation ratio at NC surface waters. Error bars indicate the standard deviation. Data are from Eastern Regional Mercury Study (ERMS, 2002-2003) and Mercury Study Extension (MSE, 2005-2006) *Data from ERMS only; ^Data from MSE only.

Table 5-3. Correlation coefficients between observed parameters from the ERMS and MSE.

	Hg(T)	MeHg	DOC	Sulfate	MeHg/Hg(T)	Hg(T) _{sed}	MeHg _{sed}	TOC _{sed}
Hg(T)	1.00	<u>0.61</u>	<u>0.46</u>	0.03	0.11	0.34	0.20	0.10
MeHg		1.00	<u>0.69</u>	-0.14	<u>0.58</u>	-0.20	-0.03	-0.14
DOC			1.00	-0.15	<u>0.40</u>	0.04	0.09	0.17
Sulfate				1.00	-0.13	0.21	0.16	0.34
MeHg/Hg(T)					1.00	-0.37	-0.05	-0.17
Hg(T) _{sed}						1.00	<u>0.46</u>	<u>0.45</u>
MeHg _{sed}							1.00	<u>0.77</u>
TOC _{sed}								1.00

Note: the red underlined numbers indicate statistically significant correlations at 0.01 level; the bold numbers suggest statistically significant correlations at 0.05 level; T = total; sed = sediment; TOC=total organic carbon

Correlation analyses to the ERMS and MSE data (Table 5-3) suggest that the monomethyl mercury concentrations in NC surface waters are closely related with total mercury (dominated by inorganic mercury) (correlation coefficient (c) =0.61), dissolved organic carbon (DOC) concentrations (c=0.69) and the ratio between methylmercury and total mercury (as an indicator of mercury methylation rate) (c=0.58).

If we assume all the external sources of mercury to aquatic systems are in the form of inorganic mercury, and methylmercury are the products of methylation after mercury reaches aquatic environments, then the amount of methylmercury in the water column can be expressed as:

$$\text{MeHg} = r * \text{Hg(T)}$$

where Hg(T) is the total mercury concentration in a water column; r is the mercury methylation rate, which ranged between 0.03 (averaged for Roanoke River at Hwy 11) to 0.18 (averaged for Cashie River School Rd near Windsor). Much variation of the mercury methylation rate was observed spatially as well as temporally. Average mercury methylation rates were highest in Cashie River, followed by Black River, Lumber River and Waccamaw River.

Mercury methylation is reported to be influenced by a variety of environmental factors such as organic carbon availability, pH, sulfur cycling, biological productivity, and temperature (Wiener et al. 2006). Conventional approaches to mercury methylation research in riverine systems have focused on processes below the sediment-water interface, where hypoxic or anoxic conditions are most favorable for the conversion of inorganic Hg to methylmercury by anaerobic bacteria; however, no significant correlations were found between water column MeHg and Hg (for both total Hg and MeHg) concentrations in the sediment, suggesting more active processes involving MeHg within the water column.

Statistically significant correlations were found between mercury methylation rate and available DOC (but not sulfate) in the water column. Unfortunately, pH values were not measured during ERMS and MSE. The relationship between methylation rate and DOC gives ($R^2=0.15$):

$$r = \alpha \times \text{DOC} + \beta$$

$$\text{MeHg} = (\alpha \times \text{DOC} + \beta) \text{Hg(T)}$$

where $\alpha = 0.0033$ l/mg; $\beta = 0.0545$; MeHg and Hg(T) are in ng/l and DOC is in mg/l. As shown in Figure 5-5, the spatial pattern of mercury methylation between aquatic systems could be explained, at least partly, by the differences in DOC concentration. For example, with a relatively higher averaged DOC concentration, mercury methylation rate appears to be higher in the Cashie River than in the Black River. DOC is believed to enhance the mobility of Hg in a

system. In addition, a higher DOC concentration usually indicates a higher productivity in a system. In such a system, photolysis of methylmercury is usually inhibited by a lack of available light at the presence of relatively higher amount of organic matter. Therefore MeHg concentrations tend to be higher in such a system.

Variations of DOC concentrations could also affect temporal variations of mercury methylation within certain systems (e.g. Waccamaw River). However, in other systems such as the Lumber River, Black River and Abbotts Creek, the observed DOC values appear to be relatively stable. The differences in mercury methylation may not be explained by differences in DOC concentrations. In such systems, the variations of mercury methylation may be influenced by other factors such as pH.

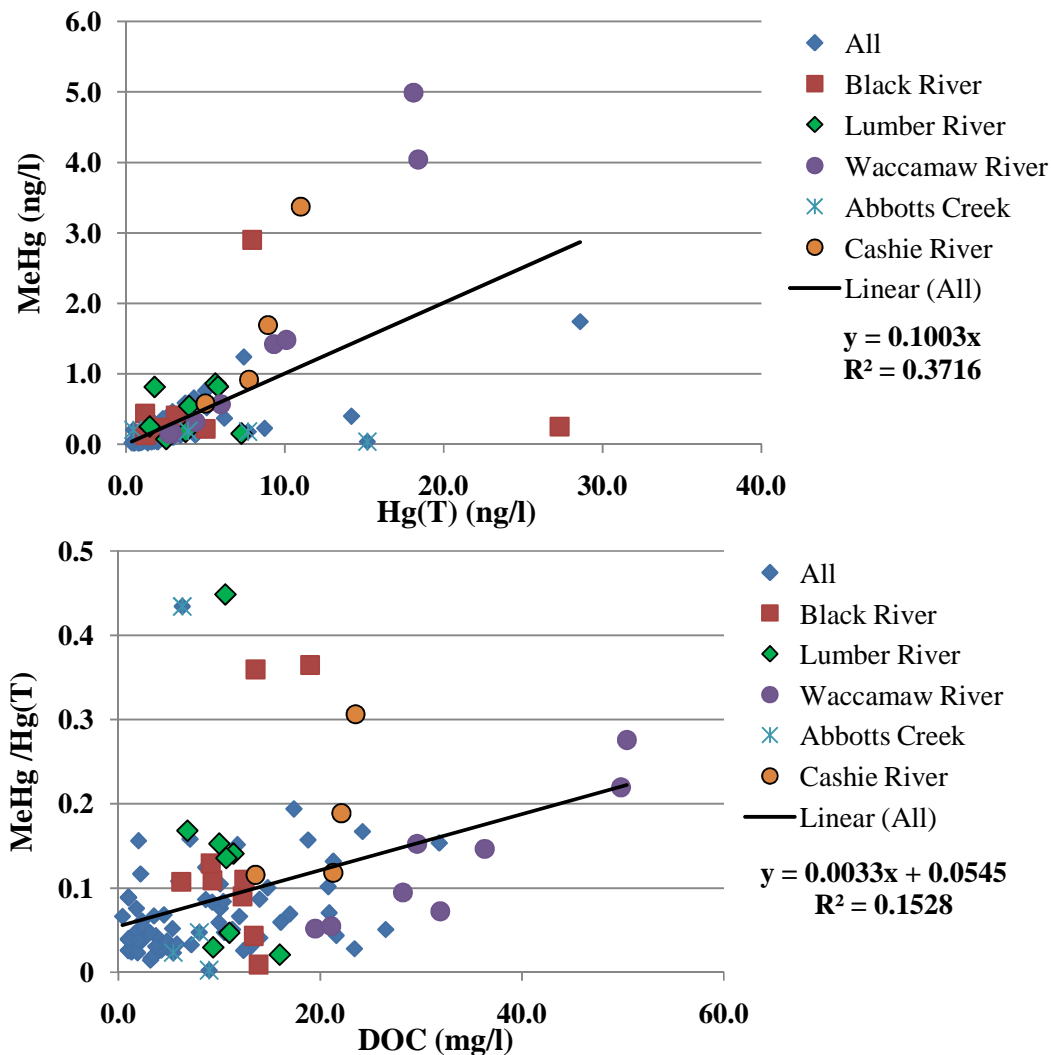


Figure 5-5. Linear regression between MeHg and Hg(T) concentrations (upper panel), and between methylation rate and DOC (lower panel).

Although Largemouth Bass mercury concentrations were measured at stations across the State, the location of the monitoring stations and the times of the surveys were not consistent with either ERMS or MSE. The data are not readily available to directly assess the bioaccumulation rate of mercury in each system. By contrast, a rough estimation on the magnitude of the bioaccumulation rate was calculated by separately averaging the Largemouth Bass mercury concentration and the water column total mercury concentration at stations (or close-by stations) where both types of data are available, and then using that ratio as the bioaccumulation factor (BAF). The calculated BAF values ranged between 0.6 - 5.4×10^5 l/kg (Figure 5-6). Some of these estimates were slightly higher than the BAF value of 0.817×10^5 l/kg, which was used in calculating NC surface water quality standard for total mercury.

Since a lot of temporal and spatial variations were observed for both the Largemouth Bass and water column Hg concentrations, and data were often scarce to represent such variations, only the magnitude of the estimated bioaccumulation factor, 10^5 l/kg, is recommended here as a reference value for mercury bioaccumulation in North Carolina waters.

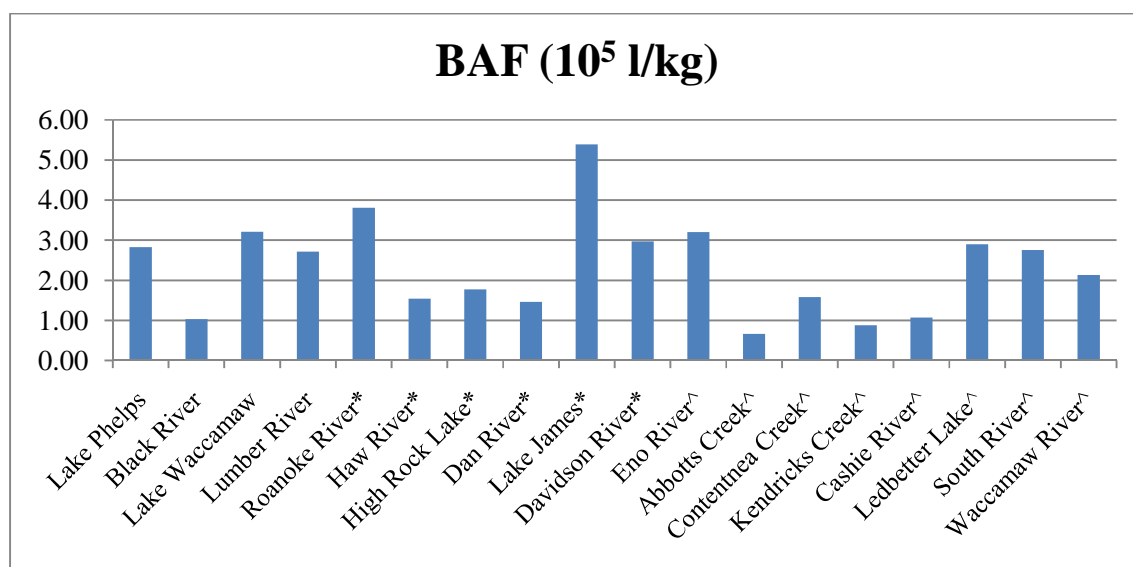


Figure 5-6. The calculated bioaccumulation factors at different streams/lakes in North Carolina. Data are from Eastern Regional Mercury Study (ERMS, 2002-2003) and Mercury Study Extension (MSE, 2005-2006) *Data from ERMS only; ^ Data from MSE only.

5.4 Point Sources to Surface Waters

There are currently 1258 NPDES permitted wastewater dischargers that discharge into North Carolina's surface waters. Four facilities discharge directly into the Atlantic Ocean and therefore have been excluded from this analysis. Approximately 18% of the NPDES permitted wastewater dischargers are industrial process and commercial facilities; those sources contribute 20% of the permitted flow in North Carolina. About 23% of the dischargers are municipal wastewater treatment plants; those discharges contribute around 67% of the permitted flow statewide. The remainder of the dischargers include small domestic wastewater dischargers, water plants and groundwater remediation dischargers. There are also approximately 1700 minor dischargers under general permits; the maximum flow from each of these permitted sites is generally below 1000 gallons per day. Excluding the facilities directly discharging into Atlantic Ocean, and including dischargers covered under general permits, the total permitted flow (i.e. maximum allowed) from all the existing facilities was estimated to be around 1913 million gallons per day (MGD).

Monitored Effluent Mercury Concentrations

Effluent mercury monitoring is required at facilities where discharge of mercury is a concern to the receiving streams. However, effluent mercury concentrations often could not be measured accurately until recent years. The EPA Method 245.1, which was normally used in effluent mercury monitoring, has a quantitation limit of 0.2 ug/l (200 ng/l), a value often much higher than the mercury NPDES permit limit for certain facilities. Beginning September, 2003, DWQ required approximately 150 facilities to use EPA Method 1631E for all effluent samples. The newer EPA Method 1631E has a quantitation level of approximately 0.5 ng/l, thus allowing compliance with the water quality standards and permit limits more feasible. The guidance on requiring analysis of mercury samples by EPA Method 1631E is explained at <http://portal.ncdenr.org/web/wq/swp/ps/npdes/guidance>.

DWQ obtained effluent mercury concentration data from Discharge Monitoring Reports (DMR) submitted by facilities where mercury monitoring is required. In order to have a more accurate assessment of the monitored mercury loading from water point sources, DWQ analyzed data from 2006 to 2009 because, by then, the use of EPA Method 1631E had been initiated at many facilities. For most of the facilities (>75%), average effluent mercury concentrations were below 12 ng/l, the current water quality standard for surface freshwaters in NC. The median of the effluent mercury concentrations among the monitored facilities is 5.2 ng/l (effluent concentrations reported below the quantitation limit of the EPA Method 245.1 (200 ng/l) are not included here). By contrast, the mean of the effluent mercury concentrations from the monitored facilities is calculated to be 42.5 ng/l, likely due to some very high concentrations reported by several facilities.

Monitored Mercury Point Source Load

To calculate the monitored mercury point source load for a facility, the monitored effluent mercury concentration was multiplied by the reported flow to give an instantaneous mercury load. An annually averaged load for the facility was obtained as the average of all the instantaneous loads that were available during the year of concern. The total monitored mercury point source load in NC was then calculated by summing all the annually averaged loads of the facilities where such data are available. While some of the power plants discharged a significant amount of flow into NC surface waters, they reported data as “less than the Practical Quantitation Limit (PQL)” of 200 ng/l using the EPA Method 245.1. The use of this analytical method and its higher quantitation level introduced uncertainty to this analysis. Using the median value (5.2 ng/l) of the effluent mercury concentrations to represent these “less than” values, the total monitored mercury point source load discharged into NC surface waters (the columns in Figure 5-7) is then averaged (2006-2009) to be around 13.3 kg (29.3 lb) per year. Among the facilities monitored for mercury, industrial process and commercial facilities appear to contribute the most.

In order to account for the uncertainty caused by using the median effluent mercury concentration to represent these “less than” values, the range of the actual monitored mercury point source load was estimated by both “zeroing out” and using the quantitation limit (200 ng/l) as the lower and upper boundaries of “less than” results. The resulting monitored mercury load estimation ranged from 7.4 kg/yr (16.3 lb/yr, zero out non-detected mercury load) to 71.5 kg/yr (157.6 lb/yr, using quantitation limit to account for the “less than” values).

Estimated Total Mercury Point Source Load

Most of the small domestic and minor municipal wastewater dischargers are not required to monitor mercury, since their contributions to total mercury loading are expected to be insignificant. The “Estimated Median Load” for mercury point sources was calculated by multiplying the median effluent mercury concentration of 5.2 ng/l (from monitored facilities) by the total permitted flow from all the NPDES facilities, resulting in a total load of about 13.7 kg/yr (30.1 lbs/yr). This number is likely under-representative on the point source loadings. On the other hand, if the mean effluent mercury concentration (from all monitored facilities) of 42.5 ng/l was chosen to represent a typical effluent concentration for all facilities, and multiply it with the total permitted flow, the “Estimated Mean Load” for mercury point sources is about 112.4 kg/yr (247.3 lbs/yr). This could serve as the upper bound of the estimated total point sources. Based on the calculation described above, the total mercury point sources in North Carolina likely range between 13.7 to 112.4 kg/yr.

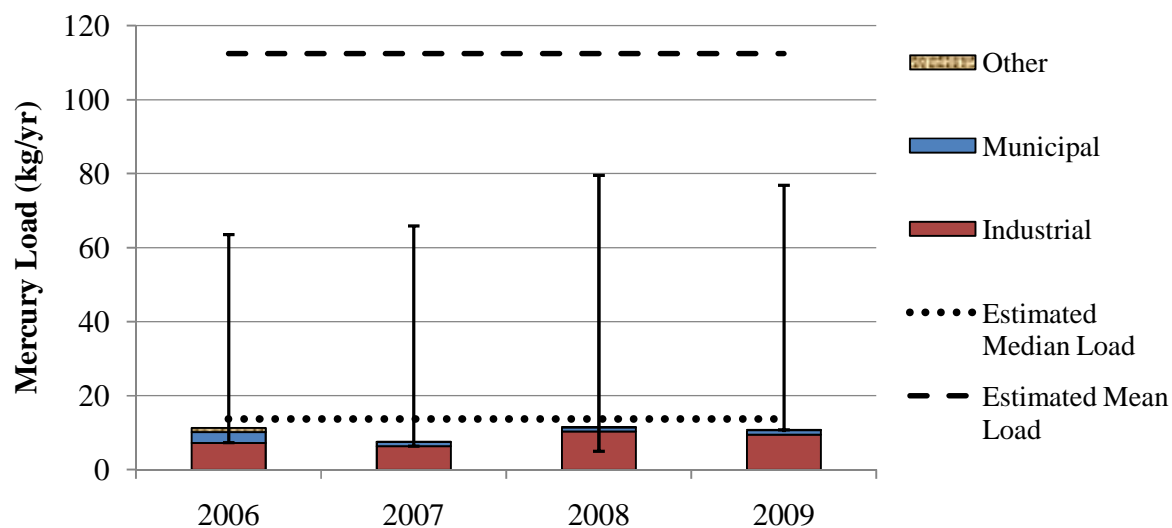


Figure 5-7. Monitored and estimated mercury point source loads. The columns represent 2006-2009 monitored mercury loadings (median effluent concentration of 5.2 ng/l was used for “less than the PQL” values). The error bars indicate the uncertainty ranges of the monitored loading due to the choices of reported concentrations below the PQL (lower error bars: “less than” values were excluded; upper error bars: the quantitation limit of 200 ng/l was used for “less than” values). Estimated Median/Mean Load is the estimated total mercury point source load as the product of the median/mean effluent concentration and the total permitted flow for all NPDES facilities.

Stormwater Mercury Load

When stormwater is addressed in a TMDL, it is generally included with the point source load and subsequently included in the wasteload allocation. However, most mercury in stormwater comes from atmospheric deposition and the exact contribution of stormwater to mercury loading is unknown. Currently, stormwater is monitored in NC at several facilities twice per year. The limited existing data indicates that mercury concentrations in stormwater varied from 0 to 10 ng/l, falling within the range of mercury concentrations normally observed in rainwater. In this TMDL, regulated stormwater is included in the WLA, and unregulated stormwater is included in the LA. Because the majority of mercury in stormwater originates from atmospheric deposition, reductions of mercury loading in stormwater will likely be addressed through reductions in atmospheric deposition. No reductions are required from NPDES stormwater permittees at this time.

6. TMDL DEVELOPMENT

6.1 Baseline Mercury Load for 2002

The total baseline mercury source load for 2002 (TSL) includes both nonpoint (NPL) and point source loads (PSL) that occurred during the baseline year of 2002.

$$\text{TSL} = \text{NPL} + \text{PSL} \quad (6-1)$$

As discussed in Section 5, the single largest nonpoint source of mercury in the U.S. surface waters is atmospheric emissions and subsequent deposition. Using CMAQ, the total air deposition of mercury within NC was estimated to be 5,239 kg for 2005. Approximately 16% of the total depositions could be attributed to in-state emissions (INPL=828 kg). Air deposition that is due to global contributions (GNPL) was estimated to be around 3661 kg; air deposition from regional contributions (RNPL) was estimated to be around 750 kg.

It was assumed that the global emissions stayed relatively stable between 2002 and 2005. The in-state and regional contributions were adjusted to account for differences between 2002 and 2005 emissions. The ratio between the actual 2002 North Carolina emissions (Table 5-1) and the modeled 2005 emissions for North Carolina (4,708 pounds) was used to adjust the modeled deposition for the regional and in-state contributions. The total NPL for the baseline year of 2002 is estimated using the following equation:

$$\begin{aligned} \text{NPL} &= \text{GNPL} + (\text{RNPL} + \text{INPL}) * (\text{In-State Emission}_{2002} / \text{In-State emission}_{2005}) \\ &= 3,661 + (750 + 828) * (5300 / 4708) \\ &= 3,661 + 844 + 932 \\ &= 5,437 \text{ (kg/yr)} \end{aligned} \quad (6-2)$$

Therefore global sources contributed approximately 67% of the total nonpoint source load, regional sources contributed approximately 16% of the total NPL, and in-state sources contributed around 17% of the total NPL during the baseline year of 2002. The in-state atmospheric deposition from natural sources was estimated to be 6% of the in-state deposition. Therefore, the in-state natural contribution is 56 kg/year.

As discussed in Section 5.4, the NPDES-regulated total point source load (including unmonitored facilities) directly into surface waters was estimated to range between 13.7 (estimated median load) and 112.4 kg/yr (estimated mean load) and the currently monitored mercury point source load was estimated to range between 7.4 and 71.5 kg/yr. An exact estimation of the total point source load is not currently available due to a number of reasons, including the analytical method used in monitoring and some reporting errors. A bounding

condition is that the total point source load should be greater than the monitored load (to count for effluent loadings from facilities that are not monitored for mercury). In this TMDL study, the estimated mean load is used as a conservative estimation of the total mercury point source load (PSL), which is 112.4 kg/yr. This number is likely over-predictive.

$$\text{PSL} = 112 \text{ kg/yr} \quad (6-3)$$

Therefore the total mercury load for the baseline year of 2002 (TSL) is estimated as:

$$\text{TSL} = \text{NPL} + \text{PSL} = 5,437 \text{ kg/yr} + 112 \text{ kg/yr} = 5549 \text{ kg/yr} \quad (6-4)$$

Based on these values, the total mercury baseline load for 2002 is 5549 kg/yr. The existing point source loads represent approximately 2% and existing nonpoint source loads represent 98% of the 2002 TSL. Figure 6-1 shows the relative contributions of different sources to the total 2002 baseline mercury load.

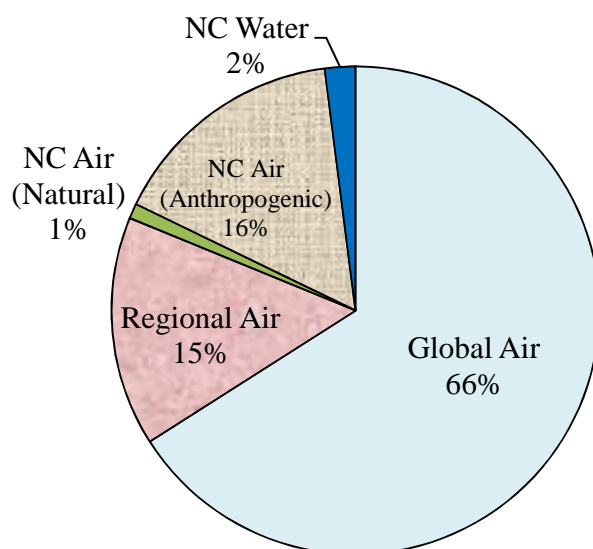


Figure 6-1. Relative contributions of 2002 total baseline mercury load.

6.2 Reduction Factor

The reduction factor (RF) is the percent reduction needed to achieve the target fish mercury concentration (Hg_{target}) for the existing fish mercury concentration (Hg_{fish}). As discussed in Section 4.3, the Hg_{target} is equal to 0.3 mg/kg, and the Hg_{fish} for this study is 0.9 mg/kg, which

represents the 90th percentile concentration based on standardized length for largemouth bass. RF is calculated using the following equation:

$$RF = \frac{Hg_{fish} - Hg_{target}}{Hg_{fish}} = \frac{0.9 - 0.3}{0.9} = 67\% \quad (6-5)$$

Therefore, a 67% of reduction in fish tissue mercury concentration is needed for this TMDL.

Proportionality of Mercury Reductions

At this time, neither the mechanisms linking emissions and mercury bioaccumulation nor the effect of a given emissions reduction on fish tissue concentrations are well understood. Study results and empirical evidence suggest that reductions in fish tissue mercury are likely to result from reductions in mercury inputs. Therefore it is reasonable to rely on certain assumptions regarding the relationships between mercury emissions, deposition, and fish tissue concentrations.

In environmental systems, steady state means that concentrations may vary from season to season or year to year, but that long term averages are constant. Several dynamic, ecosystem scale models such as the Mercury Cycling Model and IEM-2M assume that, at steady state (i.e., over long time periods), reductions in fish mercury concentrations will be proportional to reductions in mercury inputs. When atmospheric deposition is the main source of mercury to a given waterbody, these models predict a linear response between changes in deposition, ambient concentrations in water and sediments, and fish mercury levels.

The TMDL is based on the following assumptions: 1 a decrease in mercury emissions will result in a proportional decrease in mercury deposition; 2. a decrease in mercury deposition will result in a proportional decrease in mercury loading to waterbodies; and 3. ultimately, a decrease in mercury loading in waterbodies will result in a proportional decrease in mercury concentrations in fish. This follows the analyses presented by the EPA *Mercury Maps* Model, which is based on steady state formulations of the Mercury Cycling Model and IEM-2M Model (EPA, 2001).

An approach is outlined below for deriving a simplified relationship between percent reductions in air deposition load and fish tissue concentrations at steady state that draws on this same assumption of long-term proportionality.

As stated in Section 5.3, the mercury concentrations in fish (Hg_{fish}), resulting from the mercury bioaccumulation process, can be expressed using the following equation:

$$Hg_{fish} = BAF \times Hg_{water} \quad (6-6)$$

Where Hg_{water} is the total mercury concentration in the surface waters, and BAF is the bioaccumulation factor, which is a constant value under steady-state conditions. Assuming linear relationship between mercury air deposition loading (L_{air}) and Hg_{water} , we have

$$Hg_{water} = r \times L_{air} \quad (6-7)$$

Again, r is constant under long-term steady-state conditions. Combining Eq. 6-6 and 6-7, the total mercury air loading can be expressed as:

$$L_{air} = \frac{Hg_{fish}}{r \times BAF} \quad (6-8)$$

Since both r and BAF are constants, then we could have:

$$L_{target} = \frac{Hg_{target}}{r \times BAF} \quad (6-9)$$

Therefore,

$$\frac{L_{target}}{L_{air}} = \frac{Hg_{target}}{Hg_{fish}} \quad (6-10)$$

$$\frac{L_{air} - L_{target}}{L_{air}} = \frac{Hg_{fish} - Hg_{target}}{Hg_{fish}} = RF \quad (6-11)$$

Thus, under long-term steady-state condition and linear relationship assumption between mercury air emission sources and mercury in fish, the same reduction factor of 67% is required in mercury air emissions and atmospheric deposition.

6.3 TMDL Goal

Methods similar to those used in the Northeast Regional TMDL (2007) and Minnesota Statewide Mercury Total Maximum Daily Load (2007) are employed below to calculate the total maximum daily load. The total baseline mercury source load (TSL), described in Section 6.1, and reduction factor (RF), as described in Section 6.2, are used to define the TMDL by applying the reduction factor to the total source load. The total source load and reduction factor are then combined to give the total maximum daily load in units of mass per time.

$$TMDL = TSL \times (1 - RF) = 5549 \text{ kg/yr} \times (1 - 67\%) = 1831 \text{ kg/yr} \quad (6-12)$$

Therefore, the total allowable load from air deposition and the facilities discharging into NC waters is 1831 kg/yr, which is equivalent to 5 kg/day. TMDL must include a daily load, in addition to the annual load. However, annual loads are more appropriate than daily loads for mercury because the concern in this TMDL study is the long term accumulation of mercury rather than the short term acute toxicity events.

Ultimately, the TMDL is presented in the basic equation form

$$\text{TMDL} = \text{WLA} + \text{LA} + \text{MOS} \quad (6-13)$$

where WLA is Wasteload Allocation (wastewater & permitted stormwater sources), LA is Load Allocation (nonpoint sources), and MOS is Margin of Safety. Each of these TMDL components is discussed below.

6.4 Margin of Safety (MOS)

The Federal Clean Water Act requires that a MOS be included in a TMDL to account for uncertainty that may be present in the calculations. An MOS can either be explicit (e.g., additional percentage load reduction), implicit in the calculations, or a combination of the two. For this mercury TMDL, the MOS is implicit because of the following conservative assumptions used to develop this TMDL:

- The 90th percentile fish mercury concentration based on a standardized-length largemouth bass was used. Largemouth bass are predator fish and tend to have relatively higher concentrations of mercury among fish species commonly caught in North Carolina waters. The vast majority of fish have concentrations lower than this. According to Equation 6-5 and 6-12, the higher the fish-tissue mercury concentration, the higher the RF and the lower the TMDL. As many people eat a combination of fish, including many at lower trophic levels than Largemouth Bass, use of the 90th percentile Largemouth Bass incorporates a margin of safety into the analysis.
- The EPA fish tissue mercury criterion of 0.3 mg MeHg / kg fish is used as the target level for this TMDL development. The North Carolina Occupational and Environmental Epidemiology Branch within NC DHHS determined the action level for issuing fish advisories in North Carolina is 0.4 mg methylmercury/kg fish. The ultimate goal of this TMDL is to have safe-level mercury concentrations in fish caught in North Carolina waters so that the fish consumption advisory in NC can be removed. Although 0.4 mg MeHg / kg fish is used as the action level for issuing fish consumption advisories in

North Carolina, the 0.3 mg/kg criterion recommended by EPA and FDA is used here as a margin of safety.

- The transformation of mercury to methylmercury is dependent on sulfur, so it is believed that reductions in sulfur deposition will lead to reduced methylation of mercury. As ongoing federal and state programs are reducing sulfur emissions and deposition, methylation of mercury should also decrease. As the TMDL does not account for this potential reduction in mercury bioaccumulation, proposed mercury reductions based on the TMDL may be overestimated and therefore provide an extra level of protection.
- The EPA fish tissue criterion used as TMDL targets are based on concentrations of methylmercury, but the state is actually measuring total mercury in fish instead of methylmercury. It is assumed that approximately 90 percent of total mercury in fish is methylmercury, so if NC is meeting a concentration of 0.3 mg total mercury /kg fish, the concentration of methylmercury is actually about ten percent lower than this value, allowing for another level of protection.

6.5 Wasteload Allocation

According to Equation 6-13, the calculated permissible load (TMDL) of mercury that will not cause the applicable water quality standards to be exceeded is the sum of the wasteload allocation (point sources), load allocation (nonpoint sources), and MOS. As explained in Section 6.4, an implicit MOS is used for this study which infers an explicit MOS of zero. Therefore the TMDL is equal to the sum of the WLA and LA. As discussed in Section 5, point sources primarily consist of discharges from NPDES wastewater treatment facilities and the only significant nonpoint source is atmospheric deposition. Consequently, the total load is apportioned between wastewater and atmospheric loads.

The WLA includes the contributions from regulated stormwater sources. Mercury loading in stormwater primarily comes from atmospheric sources, but also includes small contributions from local sources within the watershed and natural sources. The vast majority of mercury in stormwater originates from air sources and will be controlled accordingly. Although regulated stormwater is considered to be part of the WLA, actual reductions in mercury loading in stormwater will have to be addressed through controls on atmospheric deposition sources that are necessary to meet the load allocation. These controls would be established through appropriate state or federal air laws and regulations. The state anticipates that once atmospheric deposition reductions are met, the only remaining regulated stormwater contributions would be solely attributed to natural sources and run-off from localized non-atmospheric sources. This residual stormwater contribution is considered to be an insignificant part of the WLA.

North Carolina is already engaged in controlling stormwater pollution using best management practices (BMPs) in accordance with Clean Water Act §402(p) and 40 CFR Part 122.44(k), and any residual mercury in stormwater that originates from non-atmospheric sources can be addressed by these programs. The six minimum control measures associated with permits for municipal separate storm sewer systems (MS4s) (<http://cfpub.epa.gov/npdes/stormwater/munic.cfm>) will contribute toward reducing mercury loading by reducing stormwater volume and sediment loading. Stormwater pollution prevention plans and associated BMP requirements for regulated industrial facilities where mercury may be a concern should also address residual mercury from non-atmospheric sources. For example, power plants with NPDES stormwater permits will need to employ efforts to prevent coal fly ash from contaminating stormwater discharges.

As discussed in Section 6.1, the existing point source load for the entire state is approximately 2% of the TSL for mercury, which is small (as compared to the nonpoint source load). According to EPA's *Draft Guidance for Implementing the January 2001 Methylmercury Water Quality Criterion* (EPA, 2006a), point source discharges are considered a small contribution if the loading or cumulative loading of all point sources to the receiving water are expected to account for a small or negligible portion of the total mercury loadings (EPA, 2006a). Therefore, all significant decreases in mercury loading to the region will come from reductions in atmospheric deposition (i.e., load allocation).

To maintain the low contribution from point source load, the WLA is set at 2% of the TMDL, which is equivalent to 37 kg/yr or 0.1 kg/day. While this percentage is based on the estimated relative contribution of wastewater point sources, the TMDL assumes regulated stormwater discharges can be contained within this 2% once emissions reductions are met.

Due to the low percentage contribution from point source dischargers, the WLA is statewide and is not specified to individual sources, thereby providing a cap for the state. Instead of allocating the WLA among sources with individual limits, mercury reduction will be accomplished through mercury minimization plans (MMPs) as needed and ancillary efforts that reduce point source particulate loading (e.g., phosphorus controls, biochemical oxygen demands (BOD) / total suspended solids (TSS) reductions, etc). Mercury minimization plans help ensure that discharges have no reasonable potential to cause or contribute to an exceedance of water quality standards. EPA believes that a requirement to develop a MMP may provide dischargers with sufficient information to voluntarily and economically reduce mercury discharges (EPA 2006a). Evaluation of progress will determine if MMPs and additional monitoring at point sources should be prescribed for dischargers that do not already have those programs in place. New or expanded point source discharges to surface waters will be addressed pursuant to the permitting strategy. All new or increased discharges will be required to stay below the statewide WLA.

No linkages between elevated fish mercury concentrations and local large water and air sources were identified. In order to avoid local impact from individual point sources, a cap of wastewater effluent mercury concentration will be developed by DWQ and included in wastewater permitting strategies. Fish tissue mercury concentrations will be continually monitored and evaluated by DWQ to investigate potential local impacts of point sources in effluent dominated streams. If necessary, DWQ will look to additional permit limitations and/or develop a site-specific mercury TMDL.

6.6 Load Allocation

Load Allocation Calculations

Subtracting the WLAs calculated in Section 6.5 from the TMDL calculated in Section 6.3 according to Equation 6-13, and including an implicit MOS as discussed in Section 6.4, yields the state's mercury LA as 1794 kg/yr or 4.9 kg/day.

The primary nonpoint source of mercury is from air emissions and hence load is allocated to air deposition. As discussed in Section 6.1, global sources contributed approximately 67% of the existing NPL (or, 66% of the TSL), regional sources contributed approximately 16% of the total NPL (or, 15% of the TSL), and in-state sources contributed around 17% of the total existing NPL (or, 17% of the TSL). The allowable loads from atmospheric nonpoint sources are allocated proportionally to their existing contributions. Load allocations are listed in Table 6-1.

Necessary Reductions to Meet LA

Natural sources cannot be controlled and are expected to remain at the same long-term average; therefore all mercury reductions must come from anthropogenic sources. Natural sources within North Carolina are estimated to be around 6% of the total emissions from NC (contributing approximately 1% of the total baseline load), which is around 56 kg/yr. Anthropogenic sources within NC are estimated to contribute around 876 kg/yr of mercury, as the difference between the INPL and the in-state natural source contributions.

In order to meet the allowable load for nonpoint sources (i.e., LA), the necessary reductions in anthropogenic atmospheric deposition within North Carolina can be calculated through the equation below:

$$\begin{aligned}
 \text{Percent reduction in anthropogenic deposition} &= \text{RF} / (1 - \text{percent of natural contribution}) \\
 &= 67\% / (1-6\%) \\
 &= 71\% \qquad \qquad \qquad (6-14)
 \end{aligned}$$

Therefore, the necessary reduction for anthropogenic air sources within NC is 71%.

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Since the contributions from global and regional natural emission sources to air deposition of mercury within NC is not readily known, this TMDL does not calculate reduction goals for air emissions from out-of-state anthropogenic sources. Instead, the out-of-state emission reduction goals are based on all air emission sources.

Table 6-1. TMDL Load Allocation and Expected Reduction.

Nonpoint Source		Percentage Contribution	2002 Baseline Load		Allowable Load		Expected Reduction
			(kg/yr)	(lbs/yr)	(kg/yr)	(lbs/yr)	
Global*		66%	3,661	8,054	1,208	2,658	67% [#]
Regional**		15%	844	1,857	278	612	67% [#]
In-State	Natural	1%	56	123	56	123	N/A
	Anthropogenic	16%	876	1,927	252	555	71%
Total		98%	5,437	11,961	1,794	3,948	67%

* In this TMDL, mercury air sources coming from outside the CMAQ model 12 km model domain are referred as global sources.

**Mercury air sources within the CMAQ model 12 km model domain but outside North Carolina are referred as regional sources.

[#]Expected percent reductions from global and regional air sources are reductions in total air deposition of mercury.

6.7 Daily Load

Because this TMDL addresses mercury accumulation in fish over long periods of time, annual loads are the technically appropriate approach for expressing mercury loading goals. Daily loads cannot be shown to correlate to fish tissue concentrations. Therefore, the calculations and compliance with this TMDL are based on annual loads. However, in order to comply with current EPA guidance, the TMDL is also expressed as a daily load.

6.8 Final TMDL

As discussed in Sections 6.1 and 6.2, in order to protect the North Carolina waters from mercury impairment and ultimately remove the fish consumption advisory, a total 67% of reduction is expected from the baseline mercury loading. The resulting TMDL goal is then 1,831 kg/yr or 5.0 kg/day.

As described in Section 6.4, a very conservative implicit MOS, based on several factors, is used for this TMDL, and therefore, it is not necessary to include an explicit MOS in the calculations.

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Calculation of the WLA and LA are described in Sections 6.5 and 6.6 respectively. The final TMDLs for North Carolina are shown in Table 6-2 for both annual and daily loads.

The WLA is defined for this mercury TMDL as 2% of the TMDL to ensure that water point source mercury loads remain small and continue to decrease.

The allowable loads from atmospheric nonpoint source (LA) are allocated proportionally to their existing contributions. Load allocations and reductions needed to achieve the target level are listed in Table 6-1 under Section 6.6.

Table 6-2. TMDL allocation summary.

	Annual Load*		Daily Load	
	(kg/yr)	(lbs/yr)	(kg/day)	(lbs/day)
Baseline Point Source	112	247	0.3	0.7
Baseline Nonpoint Source	5,437	11,961	14.9	32.8
Baseline Total	5,549	12,208	15.2	33.5
Margin of Safety	Implicit	Implicit	Implicit	Implicit
Wasteload Allocation	37	81	0.1	0.2
Load Allocation	1,794	3,948	4.9	10.8
Total Maximum Daily Load	1,831	4,029	5.0	11.0

*Annual load is included to facilitate implementation of the daily allocations as appropriate in NPDES permits and nonpoint source directed management measures, see Section 6.7.

7. SEASONAL VARIATION AND CRITICAL CONDITION

Seasonal variations and “...critical conditions for stream flow, loading, and water quality parameters” are discussed in 40 CFR Part 130.7(c)(1). The regulation states that: “for pollutants other than heat, TMDLs shall be established at levels necessary to attain and maintain the applicable narrative and numerical WQS with seasonal variations and a margin of safety which takes into account any lack of knowledge concerning the relationship between effluent limitations and water quality. Determinations of TMDLs shall take into account critical conditions for stream flow, loading, and water quality parameters”.

Mercury deposition and concentrations in water vary due to seasonal differences in rain and wind patterns, but this variation is not relevant because mercury concentrations in fish represent accumulation over their life spans. Factors such as size and waterbody conditions have greater effect on mercury concentrations than seasonal variation. The mercury concentration in the fish represents an integration of all temporal variation up to the time of sample collection. Variability among fish because of differences in size, diet, habitat, and other undefined factors are expected to be greater in sum than seasonal variability. This TMDL is expressed as an average annual load.

Critical conditions in this TMDL are related to sensitivities of water bodies to mercury loading because of their water chemistry. Fish mercury concentrations tend to be higher in the eastern coastal plain regions of North Carolina than those in the mountain and piedmont regions. This aspect of critical conditions has been addressed in this TMDL by using the 90th percentile of the standardized-length Largemouth Bass mercury concentration over the entire State of North Carolina. The mercury concentrations in the most popular and most likely consumed fish species in eastern North Carolina are usually less than those found in Largemouth Bass and much less than the 90th percentile of the standardized-length Largemouth Bass. It is reasonable to expect that mercury concentrations in the most likely consumed fish species in eastern North Carolina will be lower than the target level once the Largemouth Bass mercury concentrations would decline below the target level. The 90th percentile is calculated from standardized-length Largemouth Bass mercury data, which has also avoided the selection of rare incidences from original samples of large-sized, long-living fish. By taking into consideration the most sensitive water bodies, the relatively insensitive water bodies will be protected as well.

8. REASONABLE ASSURANCE

A complete TMDL evaluation requires reasonable assurance that the impaired waters will attain water quality standards. In this TMDL, nonpoint sources are the major source of the pollutant. Studies have shown that reductions in mercury air emissions on state, federal, and international levels will significantly reduce mercury concentrations in fish (e.g., <http://www.newmoa.org/prevention/mercury/MercurySuccessStorySummary.pdf>). Reasonable assurances are provided in the following to show what actions are available and will continue to reduce mercury contamination in North Carolina, and what new or proposed actions will further reduce mercury in fish to fulfill the goal of this TMDL.

Based on the programs listed below, the expected reductions from North Carolina air emission sources are expected to exceed the reduction goal proposed in this TMDL. DWQ expects that these initiatives will eventually lead to reductions sufficient to reduce fish mercury concentrations in North Carolina water bodies. Uncertainties about implementation of reduction efforts world-wide and the complexity of mercury cycling make it difficult to predict when the effects of these actions will result in significant improvements in North Carolina.

8.1 State Level Assurances

a) Air Quality

- **Clean Smokestacks Act and Expected Reduction in Mercury Emissions in NC**

(<http://daq.state.nc.us/news/leg/hg/>)

North Carolina's General Assembly enacted legislation in 2002 in controlling multiple air pollutants from coal-fired power plants. The Clean Smokestacks Act requires power companies to reduce their smog- and haze-forming emissions by approximately three-fourths over the next decade. Under the act, coal-fired power plants must achieve a 77% cut in NO_x emissions by 2009 and a 73% cut in SO₂ emissions by 2013. A significant co-benefit resulting from the controls being put in place to reduce NO_x and SO₂ is a reduction in mercury emissions.

Although the Clean Smokestacks Act does not set caps on mercury, DAQ has estimated that the controls needed to meet the NO_x and SO₂ limits will reduce mercury significantly – 72% reductions (from baseline year 2002) of mercury emissions from stationary point sources are projected by 2016 (see Section 5.2). This level of reduction exceeds the reduction goal (71%) identified in this TMDL for in-state anthropogenic air emissions.

- **Expected Reduction in “Deposition-prone Mercury” Emissions in NC**

Organic methylmercury in fish largely comes from inorganic airborne mercury that is deposited into waters. Mercury emitted from industrial sources, depending on the mercury species emitted and other factors, can deposit locally and regionally in U.S. water bodies, as well as contribute to the global pool, where it can be transported and deposited around the world. Each of the mercury forms described below has a different fate in the atmosphere.

Mercury exists in the atmosphere in three forms or species: (1) elemental gaseous mercury, Hg^0 , which is relatively non-reactive; (2) gaseous oxidized mercury, Hg^{+2} , which is highly reactive; and (3) particulate bound mercury, Hg^P , which is attached to particles.

Since it is gaseous and non-reactive, elemental mercury has a long atmospheric residence time on the order of a year and is capable of being transported over very long distances, forming the global background of mercury. This global mercury background circulates around the world, as it is referred to as the global pool, in which Hg^0 dominates the total mercury composition (greater than 95%) (NESCAUM, 2008).

The other two species, gaseous oxidized mercury and particulate bound mercury, levels in air are locally elevated near sources. Due to their shorter atmospheric lifetime on the order of days to weeks, Hg^{+2} and Hg^P are transported over relatively short distances and can deposit via wet (rain) or dry processes within roughly 50 to 500 miles of their source. Total mercury deposition is likely to be dominated by all sources of Hg^0 including global sources, while deposition of Hg^{+2} and Hg^P is primarily from local and regional sources. However, conversion between mercury species may occur during atmospheric transport, which will affect the transport time and distance (Marsik et al., 2007; NESCAUM, 2008).

For the purpose of this TMDL report, Hg^P and Hg^{+2} emissions from NC sources are considered to be the “*deposition-prone mercury*” species given their expected behavior for significant local deposition. Since most Hg^P and Hg^{+2} emissions are expected to deposit within NC boundaries after release, DAQ considers these two deposition-prone mercury species to be more important metrics for TMDL purposes than total mercury emissions.

Similarly to the different atmospheric fate of mercury species, the emission control characteristics of mercury are likewise species dependent. According to a large body of measurements on speciated mercury emission control for coal-fired utility boilers,

- Little-to-no elemental mercury is collected in the EGU electrostatic precipitators or wet scrubbers, as the effective emission control technology for Hg^0 is activated carbon.
- Most (> 90%) Hg^P and Hg^{+2} is collected in EGU ESPs or wet scrubbers.

Since the control and deposition characteristics of mercury emissions are species dependent, it is critical to develop emission inventories for TMDL purposes on mercury species rather

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than on total mercury. Accordingly, DAQ has developed emission inventories to show the relative reductions of mercury species between 2002 and 2016. A focus on the deposition-prone mercury species emissions, rather than on total mercury emissions, more accurately meets the TMDL objectives. Table 5-1 indicates there is an expected 72% reduction in total mercury emissions between 2002 and 2016. But Table 8-1 shows an expected 81% reduction in the deposition-prone mercury species emissions between 2002 and 2016. The mercury speciation estimates for the EGUs are based on numerous emission measurements, while the mercury speciation estimates for the Other Point Sources are based on engineering judgment made by EPA (EPA, 2011). Further information on speciated mercury emissions and on deposition modeling from NC industrial sources will be presented in the 2012 Mercury Report (DAQ, 2012).

Table 8-1. Expected Reductions in NC's Deposition-Prone Mercury (Hg^P and Hg⁺²) Emissions

Source Type	2002 lbs/year	2010 lbs/year	2016* lbs/year	2002-2016 Reduction
Electric Generating	1,645	655	125	92%
Other Point	1,050	440	400	62%
Total	2,695	1,095	525	81%

*2016 projected emission include EPA's Electric Generating Units Maximum Achievable Control Technology (MACT) Rules and planned shutdowns/fuel conversions.

The expected 81% reduction in deposition-prone mercury emissions from North Carolina sources will likely lead to greater reductions in mercury deposition than would be needed to meet reduction targets in this TMDL. Achieving the expected reduction in air emissions of deposition-prone mercury species within NC may introduce another layer of margin of safety towards the goal of this TMDL.

- **Mercury Regulations For Electric Generators** (excerpts):

(<http://www.ncair.org/rules/rules/D2509.pdf> & <http://www.ncair.org/rules/rules/D2511.pdf>)

North Carolina also has two State mercury rules that deserve to be highlighted. They are 15A NCAC 02D .2509, Periodic Review and Reallocations, and 15A NCAC 02D.2511, Mercury Emission Limits (see links above).

Under 15A NCAC 02D .2509, NC Division of Air Quality shall report to the EMC information on the regulation of mercury emissions in 2008 and 2012. Based upon the

upcoming 2012 report, the EMC will review the state of mercury technology and decide if any rule changes are needed.

Under 15A NCAC 02D .2511, Duke Energy and Progress Energy, two of the State's largest mercury emitters, must submit mercury control plans to DAQ by January 1, 2013. Each plan must identify the technology proposed for use at each of their units, the schedule for installation and operation of mercury controls at each unit and the identity of units that will be shut down. Any unit that does not have mercury controls installed by the end of 2017 is required to be shut down by December 31, 2017. The EMC will approve a mercury control plan if it finds that the plan achieves the maximum level of reductions in mercury emissions at each unit that is technically and economically feasible. In addition, each utility will provide DAQ with mercury reduction data collected at four boilers before and after the installation of selective catalytic reductions (SCRs) and scrubbers. All new sources are required to install the best available control technology with an emissions limitation, based upon the maximum degree of reduction of mercury from coal-fired electric steam generating units that is achievable for such units taking into account energy, environmental, and economic impacts, and other costs.

Although the federal Clean Air Mercury Rule no longer exists, significant mercury reductions in North Carolina have already occurred. The controls needed to comply with the Clean Smokestacks Act and the federal Clean Air Interstate Rule provide significant co-benefits in the form of mercury emission reductions.

- **Mercury Programs For Non-electric Generators:** Mercury emissions from non-EGU facilities are declining in North Carolina as well. Emissions from steel mills continue to decline due to implementation of a State law that requires the removal of mercury switches from scrapped vehicles. In addition, the EPA adopted hazardous air pollution standards for industrial boilers in March 2011 that will require industrial boilers to meet certain limits for mercury. EPA agreed to reconsider certain elements of the final boiler rule and is expected to finalize this rulemaking in May 2012.

b) Water Quality

- **Effluent Limit**

Local impacts from wastewater discharges primarily occur where the facility discharges to an effluent dominated stream. In those cases, the NPDES permit requires more stringent controls for BOD removal (monthly average limits are commonly in the range of 5-15 mg/l). TSS limits are not reduced at the same time but, because TSS exerts its own oxygen demand, dischargers must, practically speaking, reduce TSS in order to meet the lower BOD limits. Hence, these restrictive effluent limits to protect receiving waters have the added benefit of

low methylmercury concentrations in the receiving water because most of the mercury is associated with solids and mercury methylation tend to be lower with lower concentrations of organic matter.

In some major coastal plain river basins such as North Carolina's Neuse River and Tar-Pamlico River basins, nutrient TMDLs were developed to control coastal eutrophication. Many facilities in these basins have already converted to biological phosphorus removal, or "Bio-P", processes to meet phosphorus effluent limits. In the Neuse basin, for example, 20 of the 29 wastewater treatment plants with 0.5 MGD capacity or greater employ some form of Bio-P process. This advanced level of treatment reduces effluent solids considerably and in doing so reduces mercury discharge.

As discussed in Section 5.2, seven of the fourteen power plants are being converted to natural gas or retired by the year 2016. The remaining 7 facilities currently have Flue-gas desulfurization (FGD) wastewater treatment systems, which are very efficient in removing mercury from their discharge. Three facilities (Marshall Steam Station, Allen Steam Station, and Belews Steam Station) have mercury concentrations in the discharges typically less than 5 ng/L. Some facilities (Cliffside Steam station, Mayo Steam station, and possibly Roxboro Steam Station) are installing a Zero Liquid Discharge System that would virtually eliminate any mercury discharges to the receiving stream.

All the actions listed above would significantly decrease point source load of mercury to the water bodies in NC.

- **Wastewater Monitoring** (<http://portal.ncdenr.org/web/wq/swp/ps/npdes/guidance> & <http://portal.ncdenr.org/web/wq/swp/ps/pre/mercury>)

Certain facilities are required to use the EPA Method 1631E (or subsequent low-level mercury methods approved by EPA in 40 CFR Part 136) when analyzing for ultra low levels of total mercury, because either the facility has a current total mercury limit in its NPDES permit that is <0.20 ug/l; or the facility has limited instream dilution.

- **Fish Tissue Monitoring** (<http://portal.ncdenr.org/web/wq/ess/bau>)

Division of Water Quality monitors mercury and other contaminants in fish across NC. Special surveys included monitoring near Riegelwood, North Carolina to monitor fish tissue after the removal of a known atmospheric mercury source, a dismantled chlor-alkali plant. Mercury monitoring has also been conducted at selected sites across the State in cooperation with the NC Division of Air Quality as part of clean air rules and as part of a long term effort to monitor mercury trends at specific locations.

Fish tissue mercury concentration, which is the target of this TMDL, will be continually monitored and evaluated. In any cases of locally elevated fish mercury concentrations due to local point sources, DWQ will look to additional permit limitations, and if necessary, develop a site-specific mercury TMDL for those specific waters.

c) Other State Programs

- **Mercury Switch Removal Law**

(<http://portal.ncdenr.org/web/wm/hw/programs/mecuryswitch>):

The Mercury Switch Removal Program is a program created by the North Carolina General Assembly that was signed into law on September 13, 2005. This law requires mercury-containing convenience lighting switches to be removed from all end-of-life vehicles prior to crushing, shredding, or smelting of these vehicles. On August 11, 2006, with the state of North Carolina participating, the EPA, automobile manufacturers, scrap processors, steel makers and auto recyclers, signed a national Memorandum of Understanding (MOU) agreement which, in part, created the National Vehicle Mercury Switch Recovery Program. Following the NC DENR request to amend the existing law so as to align with the National Vehicle Mercury Switch Recovery Program, Governor Easley, on June 29, 2007, signed amendments to the Mercury Switch Removal legislation, which incorporated guidelines from the Memorandum of Understanding and realigned the legislated requirements. For the time period since the law's effective date, July 1, 2007, through June 30, 2010, North Carolina has collected 233,995 of the National Vehicle Mercury Switch Recovery Program's total 2,752,700 mercury switches for that period (8.5% from North Carolina). For the same period the estimated total number of switches available was 11,199,260 nationally and 289,112 in North Carolina. The Program's efficiency (ratio of number of switches collected divided by the number of switches available, expressed as a percentage) nationally was 24.6% and for North Carolina it was 80.9% and for the past two years North Carolina has ranked #1 in the nation in switch removal efficiency. In other terms, North Carolina's Mercury Switch Removal Program, during the three years of operation, has collected 514.79 pounds of elemental mercury that would otherwise have entered the environment through the steelmaking process.

- **Dental Amalgam** (<http://www.p2pays.org/ref/32/31311.pdf>):

Amalgam is regulated as a hazardous waste because some Toxicity Characteristic Leaching Procedure (TCLP) tests have shown that it exhibits characteristic toxicity for mercury (40 CFR 261.24). Facilities that produce less than 220 pounds total of hazardous waste in any one month are classified as "Conditionally Exempt Small Quantity Generators" (CESQGs). Most dental facilities would fall into the CESQG category. As indicated in the name, these CESQGs are generally exempt from most federal Resource Conservation and Recovery Act (RCRA) requirements.

The state of North Carolina prohibits the disposal of hazardous waste, even from CESQGS, into municipal solid waste landfills (15A NCAC 13B .1626). Waste amalgam caught in the traps and screens of the plumbing, as well as other scraps of amalgam from the dental office, must be shipped to a properly permitted facility. Triturating amalgam capsules normally pass the TCLP test and can be disposed as general solid waste. CESQGs are not normally bound by storage time limits but cannot accumulate more than 2,200 lbs total of hazardous waste.

Amalgam in wastewater is regulated either by the sewer use ordinance of the local wastewater authority for discharges to sewer systems or by the local health department for discharges to a septic tank. The sewer discharge limit for all users for mercury recommended in the N.C. Sewer Use Ordinance template is 0.0003 mg/l. Local limits may differ. Dischargers to a septic tank are prohibited from discharging hazardous waste and from contaminating groundwater at the compliance boundary. The standard at which a wastewater is considered to be hazardous for mercury—therefore, prohibited from land disposal—is 0.2 mg/l; the groundwater standard is 0.0011 mg/l ((15A NCAC 2L.0200).

- **Fluorescent Lamps** (<http://p2pays.org/Fluorescent/regstatus.asp>):

Under federal and state regulations, commercial and industrial entities are required to determine whether mercury-containing lamps, including compact fluorescents, which are destined for disposal are classified as a hazardous waste (Households are exempt from these regulations).

A lamp is considered a hazardous waste if it exhibits the characteristic of hazardous waste toxicity. Generators can determine whether a lamp exhibits the toxicity characteristic for mercury by using the TCLP to test the lamp or by receiving documentation from the lamp manufacturer, distributor, vendor or other reliable source.

If a generator cannot demonstrate that waste lamps are non-hazardous, the lamps must be managed as any other hazardous waste, including shipment to a recycler as hazardous waste, or they can be managed as universal waste. The consolidation of hazardous waste lamps via the process of crushing is considered treatment. Generators may crush lamps on-site in closed containers. However, the crushed lamps are then considered to be a fully-regulated hazardous waste.

- **Mercury Pollution Prevention** (<http://www.p2pays.org/mercury/index.asp>):

NC Division of Pollution Prevention and Environmental Assistance provides a wide range of non-regulatory technical assistance on the elimination, reduction, reuse and recycling of wastes such as mercury and the conservation of water and energy. Information is included on

the following sectors and products: Auto, dental, hospital, laboratory, local government, household; thermostats, fluorescent lights, thermometers, and much more.

8.2 National and International Assurances

Due to the long residence time and the long range of mercury (especially elemental mercury) being transported in the atmosphere, reductions in mercury air emissions outside of North Carolina will eventually lead to reduced mercury deposition in North Carolina and reduced contamination of North Carolina fish. A variety of programs, initiatives and regulations exist in the U.S. and internationally to address key mercury issues including data collection and inventory development, source characterization, and best practices for emissions and use reduction. Some major actions and regulations are described below. Additional information can be found at <http://www.epa.gov/mercury/other.htm#Intl>).

- **Mercury Export Ban Act** (http://frwebgate.access.gpo.gov/cgi-bin/getdoc.cgi?dbname=110_cong_public_laws&docid=f:publ414.110.pdf)

The Mercury Export Ban Act was signed into law on October 14, 2008. The Mercury Export Ban Act includes provisions on both mercury exports and long-term mercury management and storage. Because the United States is ranked as one of the world's top exporters of mercury, implementation of the act will remove a significant amount of mercury from the global market. Currently, mercury is exported from the United States to foreign countries where it has various uses, including for use in small-scale gold (artisanal) mining. This use of mercury raises worker safety and environmental emissions issues. To aid in addressing these concerns, EPA has provided expertise to the United Nations Industrial Development Organization's Global Mercury Project's artisanal mining project, which focuses on best management practices to reduce occupational exposure, emissions and mercury use.

- **Mercury-Containing and Rechargeable Battery Management Act** (<http://thomas.loc.gov/cgi-bin/query/z?c104:H.R.2024.ENR>:) (<http://www.epa.gov/osw/hazard/recycling/battery.pdf>)

The Mercury-Containing and Rechargeable Battery Management Act of 1996 phases out the use of mercury in batteries, and provides for the efficient and cost-effective disposal of used nickel cadmium batteries, used small sealed lead-acid batteries, and certain other regulated batteries. The statute applies to battery and product manufacturers, battery waste handlers, and certain battery and product importers and retailers.

- **Resource Conservation and Recovery Act (RCRA)** (<http://www.epa.gov/mercury/regs.htm>)

RCRA requires that EPA manage hazardous wastes, including mercury wastes, from the time they are generated, through storage and transportation, to their ultimate treatment and disposal. EPA has established treatment and recycling standards that must be met before these wastes can be disposed of. Certain mercury wastes -- mercury-containing household hazardous waste and waste generated in very small quantities -- are exempt from some RCRA hazardous waste requirements. RCRA also sets emission limits for mercury-containing hazardous waste that is combusted. States are largely responsible for implementing the RCRA program and their requirements can be more stringent than federal requirements; for example, some states have identified specific mercury-containing wastes, such as dental amalgam, as warranting more stringent treatment and disposal.

- **Clean Air Act** (<http://www.epa.gov/air/caa/>)

The Clean Air Act regulates 188 air toxics, also known as “hazardous air pollutants.” Mercury is one of these air toxics. The Act directs EPA to establish technology-based standards for certain sources that emit these air toxics. Those sources also are required to obtain Clean Air Act operating permits and to comply with all applicable emission standards. The law includes special provisions for dealing with air toxics emitted from utilities, giving EPA the authority to regulate power plant mercury emissions by establishing “performance standards” or “maximum achievable control technology” (MACT), whichever the Agency deems most appropriate. EPA has finalized Mercury and Air Toxics Standards for power plants (<http://www.epa.gov/airquality/powerplanttoxics/actions.html>) (<http://www.gpo.gov/fdsys/pkg/FR-2011-05-03/pdf/2011-7237.pdf>) to limit mercury, acid gases and other toxic pollution from power plants, keeping 91 percent of the mercury in coal from being released to the air. In addition, EPA adopted federal hazardous air pollution standards for industrial boilers in March 2011 that will require industrial boilers to meet certain limits for mercury. The agency agreed to reconsider certain elements of the final boiler MACT rule and is expected to finalize this rulemaking in May 2012.

- **Clean Water Act** (<http://www.epa.gov/regulations/laws/cwa.html>)

Under the Federal Water Pollution Control Act or “Clean Water Act” (CWA), States adopt water quality standards for their rivers, streams, lakes, and wetlands. These standards identify levels for pollutants, including mercury, that must be met in order to protect human health, fish, and wildlife. No person may discharge pollutants, including mercury, into waters unless the person has a permit. Under the Clean Water Act, either EPA or States issue permits, which must include limits that ensure the water quality standards are met. In addition, EPA and States issue information to the public on waters contaminated with mercury and on the harmful effects of mercury, identify the mercury sources and reductions needed to achieve water quality standards. Regional or statewide mercury TMDLs were developed and being implemented in Minnesota, Northeast States and New Jersey.

Under CWA Section 319(g), a state may petition EPA to convene an interstate management conference if the state is not meeting water quality standards in whole or in part as a result of nonpoint source pollution from another state. A Clean Water Act Section 319(g) Management Conference took place on June 22-23, 2010, in Philadelphia, Pennsylvania. The Management Conference was convened by EPA in response to a Clean Water Act Section 319(g) petition from the Northeast States (<http://water.epa.gov/lawsregs/lawsguidance/cwa/tmdl/mercury/319g.cfm>). In the petition, the Northeast States indicated that reductions from outside the Northeast States are needed to meet the Northeast Regional Mercury TMDL goals, and named eleven upwind States as contributing to the deposition in the Northeast. Each of the participating States introduced their successes and lessons learned in their mercury programs. The States also identified key areas for further action at the state and national levels. One outcome of the conference was an agreement among the participants that the dialogue be continued among the participating States in order to advance efforts to reduce mercury emissions.

- **Great Lakes Initiative - Regulation to Ban Mixing Zones in the Great Lakes**

(<http://water.epa.gov/lawsregs/lawsguidance/cwa/criteria/gli/finalfact.cfm>)

EPA took a final action to ban the use of mixing zones that dilute bioaccumulative toxic chemicals discharged into the Great Lakes system in November 2000. This action prohibited new discharges of toxic chemicals into mixing zones and phased out the use of existing mixing zones in the Great Lakes by November 15, 2010. It has been found that toxic discharges into mixing zones build up in the Great Lakes system and threaten human health, aquatic life and wildlife. This regulation will ban up to 700,000 toxic pounds annually of chemicals that are discharged into the lakes and that accumulate in fish and wildlife, including mercury, dioxin, polychlorinated biphenyls and pesticides. Mercury discharges alone will be reduced by up to 90 percent.

EPA includes a limited exception that would allow minimal use of mixing zones for discharges of bioaccumulative chemicals for existing facilities that may suffer unreasonable economic effects.

- **Tennessee Valley Authority (TVA) Coal Plant Air Pollution Settlement**

(<http://www.epa.gov/compliance/resources/cases/civil/caa/tvacoal-fired.html>)

A settlement was reached in April, 2011 in a lawsuit brought by the State of North Carolina against the TVA for operating coal-fired power plants with excessive air pollution. The settlement requires TVA to retire at least 18 of its 59 coal units including its oldest and most polluting, install and continuously operate up-to-date emission-control equipment on most of the remaining units, and pay North Carolina \$11.2 million in mitigation over the next five years for energy efficiency and electricity-demand-reduction programs.

- **United Nations Environmental Programme (UNEP)**

(<http://www.unep.org/hazardoussubstances/Mercury/tabid/434/language/en-US/Default.aspx>)

UNEP has been working to address mercury issues since 2003. Currently, the UNEP mercury programme has two main facets. 1) The Negotiating Process: In February 2009, the Governing Council of UNEP agreed on the need to develop a global legally binding instrument on mercury; 2) UNEP Global Mercury Partnership: Governments initiated partnership activities at Governing Council 23 and have subsequently strengthened the role of partnerships to effectively deliver mercury activities. Governing Council 25/5 specified the UNEP Global Mercury Partnership as one of the main mechanisms for the delivery of immediate actions on mercury during the negotiation of the global mercury convention. The overall goal of the UNEP Global Mercury Partnership is to protect human health and the global environment from the release of mercury and its compounds by minimizing and, where feasible, ultimately eliminating global, anthropogenic mercury releases to air, water and land.

- **The Global Mercury Project (GMP)**

(<http://www.globalmercuryproject.org/about/about.htm>)

The GMP began in August 2002 with a vision to demonstrate ways of overcoming barriers to the adoption of best practices and pollution prevention measures that limit the mercury contamination of international waters from artisanal and small-scale gold mining. Six countries have been formally participating in the GMP: Brazil, Lao PDR, Indonesia, Sudan, Tanzania and Zimbabwe. The GMP aims to introduce cleaner technologies, train miners, develop regulatory mechanisms and capacities within Government, conduct environmental and health assessments and build capacity within participating countries which will continue monitoring Hg pollution after the project.

- **North American Regional Action Plan on Mercury**

(<http://www.cec.org/Page.asp?PageID=924&ContentID=1297>)

The North American Regional Action Plan (NARAP) is one of a number of such regional undertakings that stem from the North American Agreement on Environmental Cooperation between the governments of Canada, the United Mexican States and the United States of America. The Agreement established the Commission for Environmental Cooperation to "facilitate cooperation on the conservation, protection and enhancement of the environment in their territories."

The purpose of the North American Regional Action Plan on Mercury is to provide the governments of Canada, Mexico and the United States, the Parties to the North American Agreement on Environmental Cooperation and to this NARAP, with a path forward in their joint and differentiated efforts to reduce the exposure of North American ecosystems, fish

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and wildlife, and especially humans, to mercury through the prevention and reduction of anthropogenic releases of mercury to the North American environment.

9. IMPLEMENTATION PLAN

An implementation plan is not required for the approval of a TMDL. Action items listed in the Section 8 (Reasonable Assurance) are being implemented. The effect of these action items will be evaluated through continued monitoring efforts by DWQ on fish tissue mercury concentrations, wastewater mercury concentrations and stormwater mercury concentrations. A permitting strategy will be developed by the NPDES unit of DWQ with respect to wastewater dischargers. A further detailed implementation plan may be developed in a later stage as needed.

Fish tissue mercury concentrations will be continually monitored and evaluated by DWQ to investigate potential local impacts from water point sources or air emission sources. If necessary, DWQ will look to additional NPDES permit limitations and/or develop a site-specific mercury TMDL. On the other hand, as reductions occur, if fish tissue data show that target fish concentrations are reached prior to current calculated reductions, reduction goals may be modified accordingly.

Local stakeholder groups, governments, and agencies are encouraged to develop an implementation plan and utilize funding sources for water quality improvement projects targeted at BMP construction and public outreach. Some potential funding sources include the North Carolina Clean Water Management Trust Fund, and Section 319 and 205j funds. Individual land owners may apply for the Community Conservation Assistance Program and Agriculture Cost Share Program to improve the condition of their property.

As noted before, a number of actions taken by the Division of Air Quality under both state and federal air pollution laws have the potential to significantly reduce mercury loading from air emissions. DAQ is also identifying additional measures that could be taken to reduce mercury air emissions and will seek public comment on those possible actions.

10. PUBLIC PARTICIPATION

The development of this TMDL was publicly noticed through various means. A website regarding this TMDL was created and maintained by DWQ during its development phase (<http://portal.ncdenr.org/web/wq/ps/mtu/tmdl/tmdls/mercury>). The TMDL process was reported and presented in a number of conferences and meetings. A list of these outreach activities can be found at Appendix C. Two public meetings regarding this TMDL were held on May 14th and 23rd, 2012.

DWQ electronically distributed the draft TMDL and public comment information to known interested parties. The TMDL was also available from the DWQ's website at <http://portal.ncdenr.org/web/wq/ps/mtu/tmdl/tmdls> during the comment period (Appendix D). The public comment period lasted from April 27th – June 18th, 2012. DWQ received comments from 1700 individuals and organizations. A summary of their comments and DWQ's response is provided in Appendix E.

11. FURTHER INFORMATION

Further information concerning North Carolina's TMDL program can be found on the Internet at the Division of Water Quality website:

<http://portal.ncdenr.org/web/wq/ps/mtu>

Technical questions regarding this TMDL should be directed to the following members of the DWQ Modeling/TMDL Unit:

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Appendix A. List of Abbreviations

AMAP – Arctic Monitoring and Assessment Programme

BAF – Bioaccumulation Factor

BOD – Biological Oxygen Demand

BMPs – Best Management Practices

CMAQ – Community Multi-scale Air Quality (Model)

CWA – Clean Water Act

DAQ – Division of Air Quality, North Carolina Department of Environment and Natural Resources

DMR – Discharge Monitoring Report

DOC – Dissolved Organic Carbon

DWQ – Division of Water Quality, North Carolina Department of Environment and Natural Resources

EGU – Electric Generating Unit

EMC – Environmental Management Commission

EoE – The Encyclopedia of Earth

EPA – United States Environmental Protection Agency

ERMS – The Eastern Regional Mercury Study

ESP – Electrostatic Precipitator

FDA – United States Food and Drug Administration

FGD – Flue-gas Desulfurization

GNPL – Air deposition that is due to global sources

Hg – Mercury

Hg (T) – Total Mercury

INPL – Air deposition that is due to In-state sources

LA – Load Allocation

LMB – Largemouth Bass

MACT – Maximum Achievable Control Technology

MeHg – Methylmercury

MMPs – Mercury Minimization Plans

MOS – Margin of Safety

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MS4s – Municipal Separate Storm Sewer Systems

MSE – Mercury Study Extension

NADP-MDN – National Atmospheric Deposition Program Mercury Deposition Network

NC DENR – North Carolina Department of Environment and Natural Resources

NC DHHS – North Carolina Department of Health and Human Services

NO_x – Nitrogen Oxides

NPDES – National Pollutant Discharge Elimination System

NPL – Nonpoint Source Load

PSL – Point Source Load

PTFE – Polytetrafluoroethylene

RF – The Reduction Factor

RNPL – Air deposition that is due to regional sources

SCRs – Selective Catalytic Reductions

SO₂ – Sulfur Dioxide

TOC – Total Organic Carbon

TSL – Total Source Load

TMDL – Total Maximum Daily Load

TSS – Total Suspended Solids

UNEP – United Nations Environment Programme

USGS – United States Geological Survey

WLA – Wasteload Allocation

WQS – Water Quality Standards

Appendix B. Mercury Photochemical Modeling for Division of Water Quality's Mercury Total Maximum Daily Load (TMDL)

By Nick Witcraft

North Carolina Division of Air Quality

The primary goal of this study is to estimate the amount/percentage of atmospheric mercury deposition in North Carolina that is attributed to sources in North Carolina. A secondary goal was to estimate the percentage of atmospheric mercury deposition that is from the global pool.

Methodology

The North Carolina Division of Air Quality (NCDAQ) conducted a series of modeling runs to generate estimates of atmospheric mercury deposition. The model was a special version of CMAQ (Community Multiscale Air Quality model) version 4.71 obtained from U. S. Environmental Protection Agency (USEPA) that included mercury chemistry. The CMAQ model and associated inputs were used for the base case modeling of the USEPA's Mercury and Air Toxics rule. The USEPA ran a national 36 km CMAQ run with boundary conditions from the GEOS-CHEM model. GEOS-Chem is a global 3-D chemical transport model for atmospheric composition driven by meteorological input from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office. The nested 12 km CMAQ run used boundary conditions from the 36 km run. The USEPA performed a model performance evaluation on the meteorology and air quality modeling. Since NCDAQ used the same version of the CMAQ model with identical inputs for the base run, the NCDAQ relied upon the USEPA's model performance evaluations and did not conduct a separate model performance evaluation for this study.

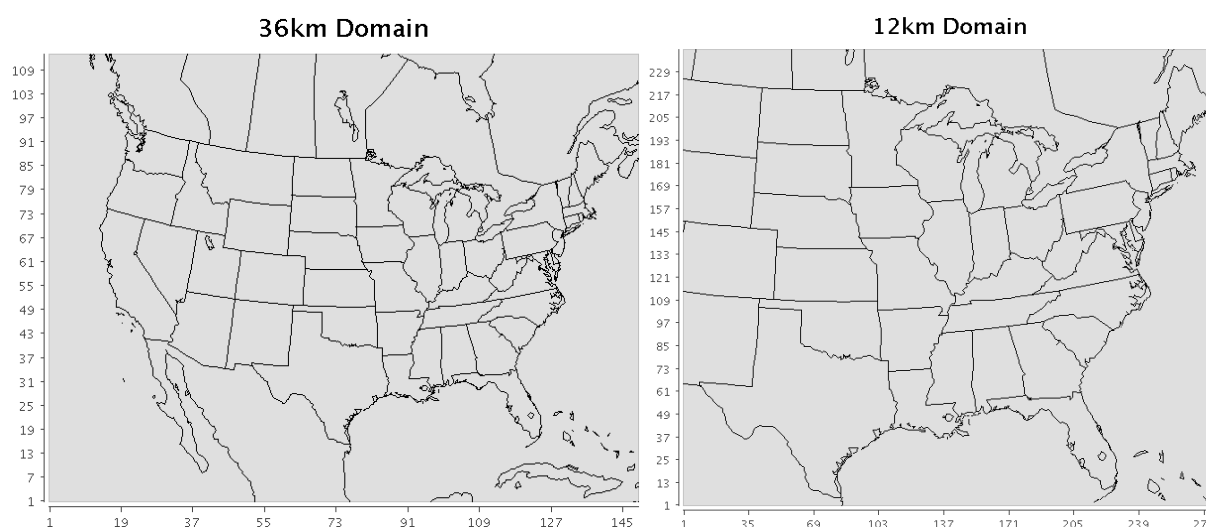


Figure 1: Outer 36km CMAQ modeling domain (left) and nested inner 12km modeling domain (right).

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NCDAQ conducted all model runs at 12 km using the following data obtained from the USEPA:

- 2005 base year emissions,
- boundary conditions from the 36 km USEPA run, and
- meteorology files processed through the Meteorology-Chemistry Interface Processor (MCIP).

NCDAQ first ran a base case CMAQ run with full emissions and boundary conditions for 2005. Next a series of zero-out sensitivities were performed to estimate the contribution of atmospheric mercury deposition in North Carolina.

Table 1. Model runs

Run Name	Emissions	Boundary Conditions
Base	12km USEPA Emissions	36 km USEPA run
Zero NC	12km USEPA Emissions with North Carolina's mercury emissions zeroed out	36 km USEPA run
No BC	12km USEPA Emissions	Boundary conditions with zero mercury emissions

To zero-out the emissions over North Carolina, a multi-step process is required. First a mask grid was created for North Carolina (Figure 2). Then the mask was applied to the "emis_mole_all" files to zero out the low level emissions over North Carolina. The Figure 3 shows the Base emissions (left) and ZeroNC emissions (right) after the mask is applied.

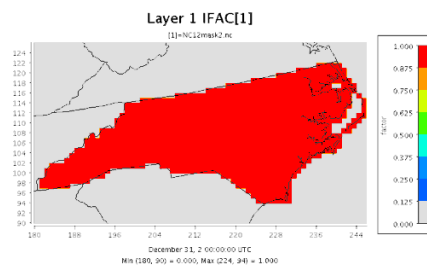


Figure 2: Grid cell mask for North Carolina

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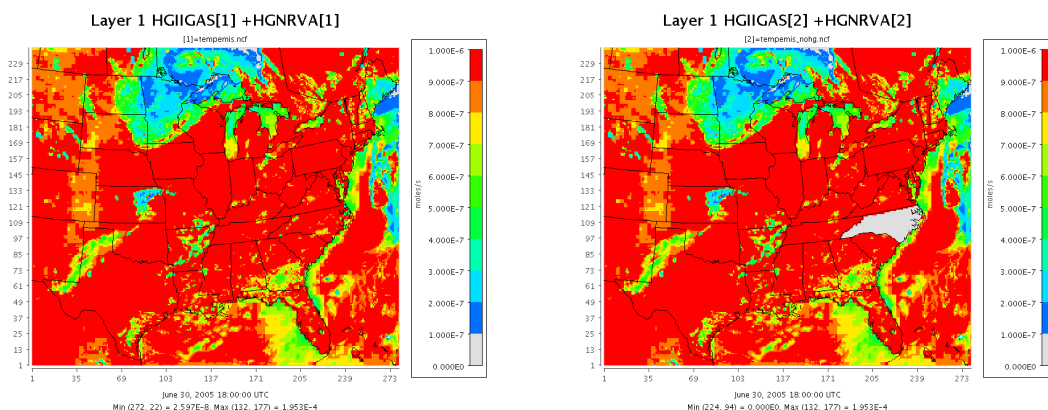


Figure 3: Low level mercury emission inputs at 1800 UTC June 30, 2005. Base emissions (left) and North Carolina zero out (right).

The next step is to zero out all North Carolina emissions within the stack files. The emission files from the USEPA contained 4 different stack files: electric generating units, non-electric generating units, shipping, and other point sources. The stack emissions reference a master file that contains a state's FIPS code, Latitude and Longitude for each source. A simple mask was created for each type of stack file based on the North Carolina FIPS code (37***), and the mask was applied to each stack file type to zero out the emissions over North Carolina.

Results

As a first quality assurance step, the hourly average air concentration (ACONC) files were examined. The image below shows the difference in mercury concentrations, between the base run and the North Carolina zero out run, 5 hours into the first day of the model run. Note that all significant differences in mercury originate from North Carolina.

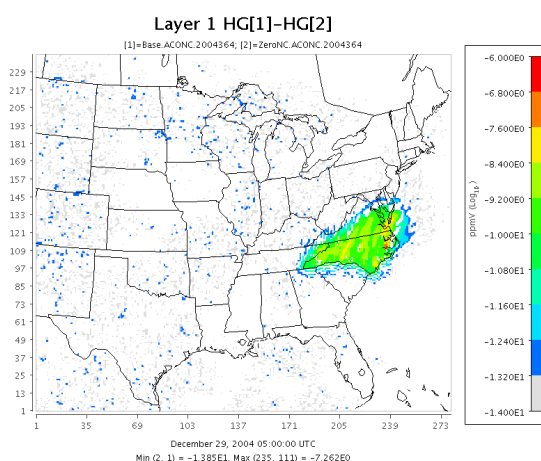


Figure 4: Difference in mercury concentrations between the base and North Carolina zero out runs after 5 hours of model integration.

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To calculate the atmospheric mercury deposition within North Carolina, a mask similar to Figure 2 is applied to the mercury deposition output and the deposition data is extracted. This provides the data in kilograms (kg) per hectare (ha) for each grid cell. The kg/ha for each 12 km grid cell is converted to kilograms deposited through the following equation:

$$(X \text{ kg/ha/cell}) \times (100 \text{ ha/km}^2) \times (144 \text{ km}^2/\text{cell}) = Y \text{ kilograms}$$

Next the grid cells are summed up for the masked area. Finally, the sum is adjusted to account for the actual number of hectares within the state. The mask covered 135,216 hectares and the actual number of hectares in North Carolina is 139,389. Therefore, the summed gridded results were multiplied by 139,389/135,216 or 1.0309.

Table 2 shows the modeled atmospheric mercury deposition in North Carolina for the Base and Zero NC modeling runs. The modeling indicates that approximately 15.8% of the atmospheric mercury deposition in North Carolina is from North Carolina sources. The exact amount of mercury deposition is very uncertain, and NCDAQ has much more confidence in the percent contribution. Figures 5 through 12 show the deposition results. The largest differences in deposition are located across the Piedmont.

Table 2. Mercury Deposition Modeling Results over North Carolina, in kilograms/year

Model Run	Dry Deposition	Wet Deposition	Total Deposition
Base	3651.3	1588.1	5239.4
Zero NC	3054.5	1357.4	4411.9
Difference (NC contribution)	596.8	230.7	827.5
% Contribution of mercury deposition in NC by NC sources	16.3%	14.5%	15.8%

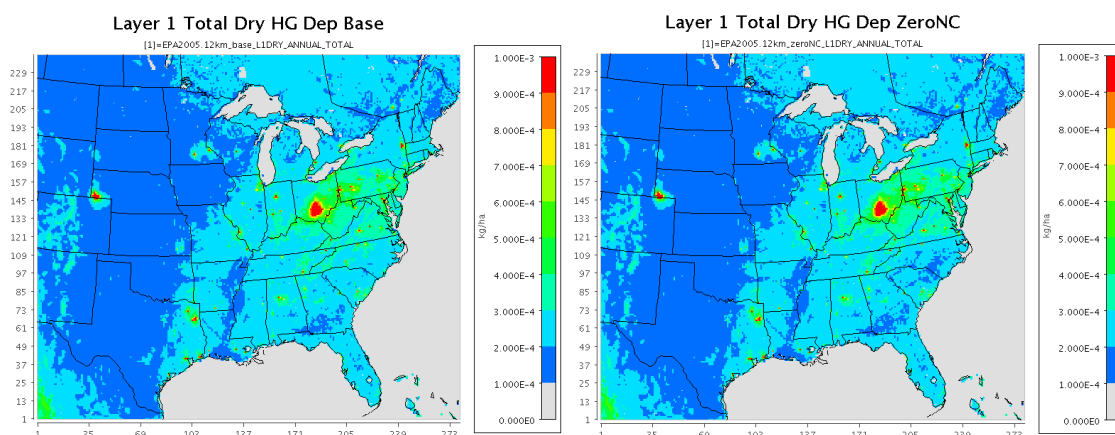


Figure 5: Dry mercury deposition for the Base run (left) and Zero NC run (right).

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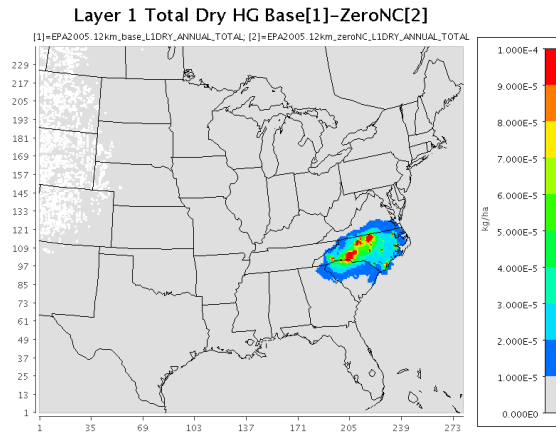


Figure 6: Difference in dry mercury deposition between the Base and Zero NC runs.

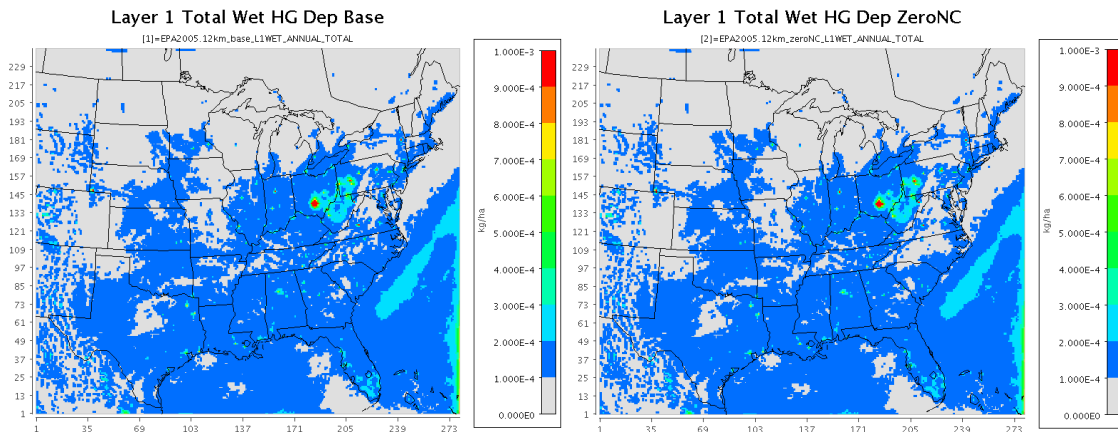


Figure 7: Wet mercury deposition for the Base run (left) and Zero NC run (right).

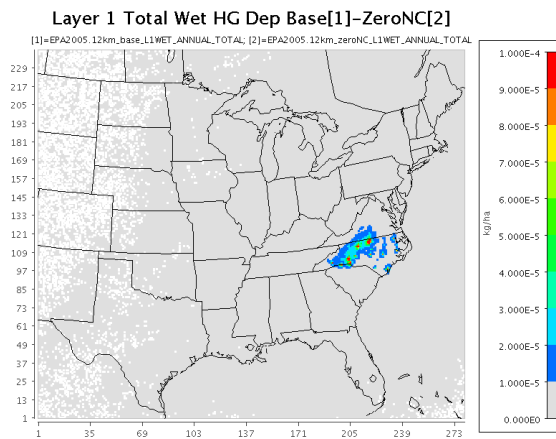


Figure 8: Difference in wet mercury deposition between the Base and Zero NC runs.

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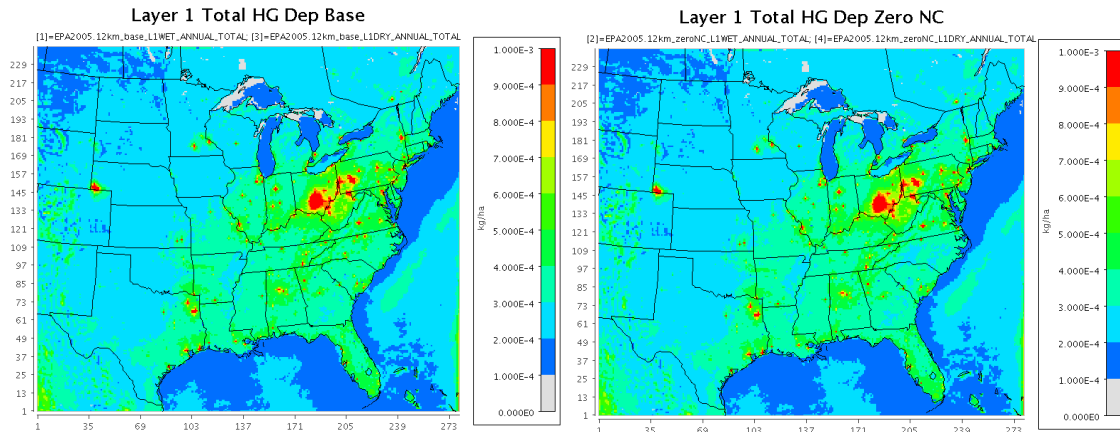


Figure 9: Total mercury deposition for the Base run (left) and Zero NC run (right).

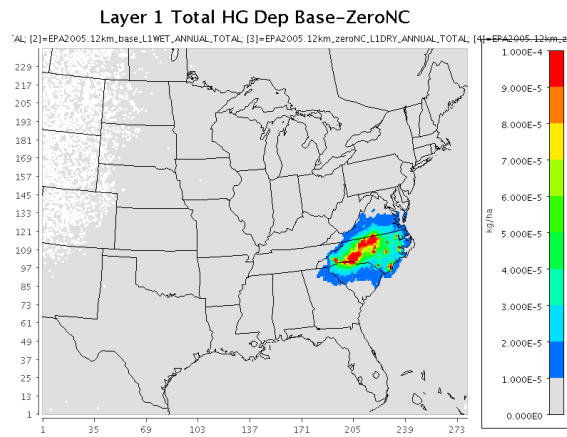


Figure 10: Difference in total mercury deposition between the Base and Zero NC runs.

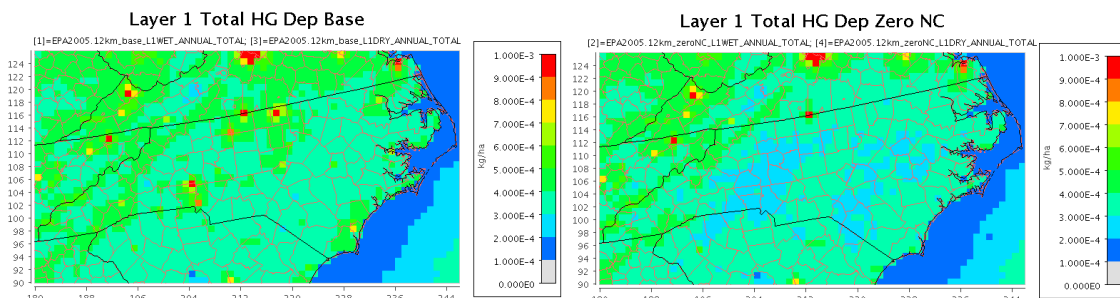


Figure 11: Close up view of total deposition in North Carolina. Base (left) and Zero NC (Right).

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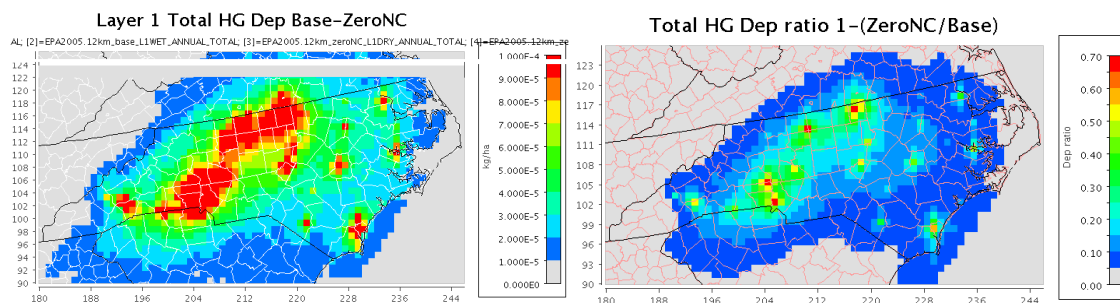


Figure 12: Close up view of North Carolina – difference in total mercury deposition between the Base and Zero NC runs. Left image is on a linear scale, right is the percent difference between the two runs.

Table 3 shows the total kilograms of mercury deposition over North Carolina, Minnesota, and the Northeast. The results for these other areas are provided for comparison to other state’s mercury TMDL efforts.

Table 3. Base Run Mercury Deposition Totals, in kg/year

Area	Dry Deposition	Wet Deposition	Total Deposition
North Carolina	3,651	1,588	5,239
Minnesota	3,985	1,849	5,834
Northeast	7,925	2,995	10,920

No mercury emissions at boundary

A run was made with no mercury emissions at the 12km Domain boundary (No BC). By comparing the Base and No BC runs (Table 4) it is estimated that nearly 70% of the mercury deposition in North Carolina is from sources outside the 12 km domain.

Table 4. Clean Boundary Condition Impact on Mercury Deposition over North Carolina, in kg/year

Run	Dry Deposition	Wet Deposition	Total Deposition
Base	3651.3	1588.1	5239.4
No BC	1198.2	379.4	1577.6
<i>Difference (12 km boundary contribution)</i>	2453.1	1208.7	3661.8
<i>% Contribution of mercury deposition in NC by sources outside the 12km domain</i>	67.2%	76.1%	69.9%

Appendix C. Outreach Activities

DAQ Outreach Activities:

- 1) Unifour Air Quality Conference in Hickory, NC; May 25, 2010; approximately 90 attendees including significant local government participation.
- 2) DAQ Regional Supervisors were updated on June 16-17, 2010; approximately 25 attendees.
- 3) Mercury Total Maximum Daily Load Update, joined DAQ/DWQ information item to Environmental Management Commission; September 2010; approximately 50 people.
- 4) Update on Mercury Total Maximum Daily Load, joined DAQ/DWQ information item to Environmental Management Commission; July 2011; approximately 50 people.
- 5) Outside Involvement Committee Meeting, August 9, 2011, approximately 20 people.
- 6) Outside Involvement Committee Meeting, November 8, 2011, approximately 20 people.
- 7) Update on Air Quality Modeling Results for Mercury Total Maximum Daily Load (TMDL), information item to Air Quality Committee; January 11, 2012; approximately 50 people.
- 8) Outside Involvement Committee Meeting, February 14, 2012, approximately 20 people.

DWQ Outreach Activities:

- 1) North Carolina Statewide Mercury TMDL web page (<http://portal.ncdenr.org/web/wq/ps/mtu/tmdl/tmdls/mercury>) was created and put online on May 24th, 2010.
- 2) North Carolina American Water Works Association and the North Carolina Water Environment Association (NC AWWA-WEA) Water Resources Committee meeting in Raleigh; June 10, 2010; approximately 30 people.
- 3) Regional Stormwater Partnership Meeting in Charlotte; June 11, 2010; approximately 35 people.
- 4) Mercury and pH seminar by Kathy Stecker in Raleigh; August 2, 2010; approximately 45 people.

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- 5) Mercury seminar by Dr. Helen Hsu-Kim from Duke, announcement/discussion of statewide mercury TMDL by Kathy Stecker; Aug. 13th, 2010; approximately 20 people.
- 6) North Carolina Chapter of American Public Work Associations - Water Resources and Technology Conference - Asheville, NC; September, 2010; approximately 200 people
- 7) Mercury “Total Maximum Daily Load” (TMDL) Update, joined DWQ/DAQ information item to Environmental Management Commission; September 2010; approximately 50 people.
- 8) Update on Mercury Total Maximum Daily Load, joined DWQ/DAQ information item to Environmental Management Commission; July 2011; approximately 50 people.
- 9) The 2011 North Carolina Environmental, Energy, Health & Safety School by Manufacturers and Chemical Industry Council of North Carolina (MCIC); August 23, 2011; approximately 50 people.
- 10) Update on Mercury Total Maximum Daily Load (TMDL), information item to Water Quality Committee; January 11, 2012; approximately 50 people.
- 11) Fish Mercury Contamination seminar by Dana Sackett from North Carolina State University, announcement/discussion of statewide mercury TMDL by Kathy Stecker; January 27, 2012; approximately 25 people.
- 12) Western Piedmont Water Resources Committee meeting, February 15, 2012; approximately 30 people.
- 13) Complying with North Carolina’s Water Quality Regulations 2011 – A practical Workshop for Dischargers by MCIC; February 12, 2012; approximately 50 people.

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Appendix D. Public Notification of North Carolina Mercury TMDL on April 27, 2012 (last updated August 8, 2012)

North Carolina Department of Environment and Natural Resources

Division of Water Quality and Division of Air Quality

Now Available for Public Comment

1. DRAFT Statewide Mercury Total Maximum Daily Load (TMDL)
2. DRAFT Mercury Post-TMDL Permitting Strategy (for wastewater)
3. North Carolina's Mercury Reduction Options for Nonpoint Sources

Interested parties are invited to comment on the draft TMDL and the Permitting Strategy by **June 18th, 2012**. We will accept comments to the Mercury Reduction Options for Nonpoint Sources until August 31, 2012. The three documents and information about public meetings can be found at <http://portal.ncdenr.org/web/wq/ps/mtu/tmdl/tmdls/mercury>

The Draft NC Mercury TMDL was developed to meet requirements of Section 303(d) of the Federal Water Pollution Control Act. It is subject to approval by EPA. The other two documents provide supplemental information on how the approved NC Mercury TMDL will be implemented. They are not subject to approval by EPA and will not be included in the TMDL package that will be submitted to EPA.

We invite comments on how to improve the clarity of the Draft NC Mercury TMDL, as well as questions and feedback on the draft wastewater permitting strategy and reduction options for nonpoint sources. The documents may be modified based on the comments received. Comments and responses on the Draft NC Mercury TMDL will be included in the TMDL package to be submitted to EPA.

Comments concerning these three documents only should be directed to Jing Lin at Jing.Lin@ncdenr.gov

Appendix E. Public Comments Response Summary

North Carolina Mercury TMDL

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State of North Carolina
Department of Environment and Natural Resources
Division of Water Quality
Division of Air Quality

Summary of Written Comments and Responses to the Comments Submitted on

Draft NC Mercury Total Maximum Daily Load
Draft Post-TMDL Wastewater Permitting Strategy
Mercury Reduction Options for Nonpoint Sources

July 6, 2012

North Carolina Mercury TMDL

September 13, 2012

Introduction

The Division of Water Quality (DWQ) developed a statewide mercury total maximum daily load (TMDL) consistent with Section 303(d) of the federal Clean Water Act. DWQ also developed a strategy for implementing the wastewater requirements of the TMDL. The Division of Air Quality (DAQ) developed North Carolina's Mercury Reduction Options for Nonpoint Sources in order to get feedback from the public on choices for nonpoint source mercury management.

The NC Department of Health and Human Services issued a statewide consumption advisory for fish high in mercury. The TMDL is needed to address impairment of fish consumption uses in NC waters. It quantifies loads from in-state and out-of-state atmospheric and water-related sources, and provides an aggregate statewide wasteload. The wastewater strategy describes how the wasteload will be allocated in individual NPDES permits.

Three documents were provided to the public on April 27, 2012 for comment:

1. DRAFT Statewide Mercury Total Maximum Daily Load (TMDL)
2. DRAFT Mercury Post-TMDL Permitting Strategy (for wastewater)
3. North Carolina's Mercury Reduction Options for Nonpoint Sources

Comments from the public were accepted April 27, 2012 through June 18, 2012 for the draft TMDL and the permitting strategy. One thousand seven hundred individuals and organizations had submitted comments by the end of June 18th. This document provides a brief summary of comments received regarding the TMDL and the Permitting Strategy, along with responses and any revisions made to the documents. Comments received during the comment period may be viewed from a link on DWQ's mercury web page <http://portal.ncdenr.org/web/wq/ps/mtu/tmdl/tmdls/mercury>. Comments on the Mercury Reduction Options for Nonpoint Sources continue to be accepted and are not included in this Summary.

Ten organizations (or organization combined) submitted their comments. They are:

- Progress Energy (PE)
- Duke Energy (DE)
- North Carolina League of Municipalities (NCLM)
- Town of Valdese (TV)
- The Utility Water Act Group (UWAG)
- US Navy and US Marine Corps with concurrence of the US Army and US Air (USN)
- NC Water Quality Association (NCWQA)
- Waterkeepers Carolina (WC)
- Appalachian Voices • Clean Air Carolina • Environmental Defense Fund • North Carolina Conservation Network • Rocky River Heritage Foundation • Sierra Club • Southern Environmental Law Center (NCCN)
- Catawba Riverkeeper (CR).

Comments and Responses

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(Number of individuals and name of organizations submitting each comment is shown in parentheses).

General Comments (not specifically about TMDL or Strategy)

- Urge the Environmental Management Commission (EMC) to pursue the strongest possible measures to keep mercury pollution out of our air and water (1513 individuals).
- Want mercury reduced to protect children and/or grandchildren from mercury-linked developmental problems (1405 individuals).
- Mercury regulation needs to be strengthened to limit exposure to mercury (1385 individuals).
- Especially concerned about mercury pollution being discharged from Progress Energy's Asheville Coal Plant. Mercury is a potent brain toxin that is particularly dangerous for pregnant women and small children. (1384 individuals).
- I am writing to urge you to use the best scientific information to help reduce mercury pollution from our waterways. Mercury pollution is a serious threat to our waterways and the safety of our fisheries, and we need to make sure any plan the state of North Carolina pursues yields actual reductions in mercury pollution. North Carolina demonstrated true leadership in protecting our air and water in 2002 when we passed the Clean Smokestacks Act. Let's continue to be a leader by implementing a strong TMDL plan that will actually make our water safe for fishing once again (165 individuals).
- High mercury levels in fish and water is a constant concern and want something done about it (25 individuals).
- Tired of company profits first, and regulators for allowing them to do it, at the expense of human health (18 individuals).
- Want coal fired plants held to the most stringent pollution standards and implement the highest pollution reduction technology available (10 individuals).
- Urge the EMC to approve the wastewater implementation plan (8 individuals).
- We know mercury has been a health issue for many years so quit fooling around and do something about it (7 individuals).
- Want general restoration of waters to improve health of North Carolina's environment and quality of life (7 individuals).
- Want more use of renewable (green) energy and energy conservation (4 individuals).
- The cost of preventing mercury contamination is negligible compared to the medical cost of mercury related illnesses (3 individuals).
- Stop Titan (2 individuals).
- New River has contaminated fish and people are still fishing (1 individual).
- EMC needs to look at more mercury being released due to fracking (1 individual).
- North Carolina is a dirty state (1 individual).
- Thank you for your commitment to help reduce mercury (1 individual).
- Applaud the Department's effort to impose reasonable regulatory requirements consistent with the insignificant nature of the point source loadings (NCWQA).
- Appreciate the Department and staff members' hard work that went into the development of the statewide mercury TMDL and for hosting the public meetings in Hickory and New Bern (WC).
- Applaud the Department for its effort to address the mercury impairment issue (USN).
- The permitting strategy should be submitted to EPA along with the mercury TMDL (NCCN).

Response:

We appreciate the interest and feedback from each commenter. Each comment submitted was read and considered, and revisions were made to the documents in response to specific comments (listed in the following sections).

Specific Comments on Draft Statewide Mercury TMDL

A) Statewide Approach

- Suggests alternative of 5m categorization of the state's waters for mercury (NCLM).

Response:

Subcategory 5m was considered. Since 2007, states have had the option of using a "5m" (m for mercury) designation for waters on their 303(d) list that are impaired by mercury predominantly from atmospheric sources. EPA allows state to defer development of mercury TMDLs while they carry out mercury reduction programs; however, 5m does not allow for flexible permitting, and does not remove a state's obligation to develop TMDLs, because the waters remain on the 303(d) list. No state has used subcategory 5m to date, yet 11 states have developed statewide mercury TMDLs in that same time period.

- Concerns with the assessment methodology used to make the determination that all waters in North Carolina are impaired for mercury (PE, DE, NCLM, TV, NCWQA).
- The EMC should lay out a path for reviewing individual watersheds to determine the health of each particular waterway. As mercury concentrations in fish tissue drop to safe levels, these watersheds can then be designated as healthy again, on a case-by-case basis (167 individuals)
- The TMDL should be revised to provide an off-ramp for stream segments where use attainment can be demonstrated (PE, DE, NCLM, UWAG).

Response:

The assessment methodology (AM) is a separate issue from this TMDL. A state's AM is reviewed and revised as needed as part of the two-year 303(d) list cycle. NC's AM for mercury may be revised in the future. A statewide TMDL is an appropriate approach where mercury loading is primarily from atmospheric deposition.

DWQ will continue to assess mercury in fish tissue, applying the AM in place at the time of assessment. The goal of any TMDL is to attain water quality standards.

- Appreciate that States need to act even in the face of uncertainty and change, commend North Carolina for statewide mercury TMDL (UWAG, NCCN).
- Concerns about the statewide approach, suggest site-specific or basin-wide approach (TW, NCWQA).
- Additional studies, using a more sophisticated air model that is readily available and can account for local deposition, are needed to accurately assess hotspots. Scientific studies show that a significant proportion of mercury emitted into the air lands locally (168 individuals).
- Recommend an initial statewide study and plan for reductions, followed by targeted site-specific TMDLs and implementation plans (NCCN).

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- The EMC should approve a modified statewide mercury TMDL now, with a commitment to modeling and follow-on watershed-specific TMDLs (NCCN).

Response:

Given that the mercury problem is widespread, a statewide approach for the development of the TMDL allows us to account for mercury sources that are not confined locally. Section 4.1 of the TMDL document compared a statewide approach with eco-regional and basin-wide approaches and concluded that “A statewide, universal mercury TMDL, which conservatively considers the necessary mercury reduction goal to remove fish consumption advisory across the state, is appropriate for mercury TMDL development in North Carolina.”

We understand the concerns regarding “hot spots.” Some commenters cited CMAQ model results reported in the TMDL document as showing “hot spots” or locally elevated mercury deposition within North Carolina. The “hot spots” of mercury deposition simulated by the CMAQ model (Figure 5-3 in the TMDL document) are mainly due to emissions from sources that are distributed within the entire model grid cell. The CMAQ model is designed to be a regional model and is not meant to be used for local hot spot analyses or to model impacts immediately around individual sources. Pollutant concentrations within the grid cells containing emission sources may be significantly over predicted due to limitations in the CMAQ model. The CMAQ model used in the TMDL study aimed to provide a statewide total deposition and the comparative contribution from different geographical emission sources.

As stated under Section 6.5 of the TMDL document, “No linkages between elevated fish mercury concentrations and local large water and air sources were identified. In order to avoid local impact from individual point sources, a cap of wastewater effluent mercury concentration will be developed by DWQ and included in wastewater permitting strategies. Fish tissue mercury concentrations will be continually monitored and evaluated by DWQ to investigate potential local impacts of point sources in effluent dominated streams. If necessary, DWQ will look to additional permit limitations and/or develop site-specific mercury TMDLs.” Language was added under Section 9 (Implementation Plan) to clarify that the statewide TMDL does not preclude studies, additional modeling, or site-specific TMDLs.

B) Water Quality Target

- The water quality target was devised inappropriately and is inconsistent with the NC Administrative Procedures Act and the 2012 Use Assessment Methodology (PE, DE, UWAG).
- The selected water quality target is inconsistent with EPA guidance and overly conservative (PE, DE, UWAG).
- The TMDL must also include water column mercury standard (back calculated from the fish tissue mercury target) (WC, NCCN).
- The draft mercury TMDL may need a more stringent criterion and a larger reduction factor (NCCN).

Response:

As explained under Section 3 of the TMDL document, North Carolina water quality standards include beneficial use designations (classifications) and numeric levels and narrative statements

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protective of the use designations. The fresh surface water quality standards applicable to the waters covered in this mercury TMDL (15A NCAC 02B .0211) state:

Best Usage of Waters: aquatic life propagation and maintenance of biological integrity (including fishing and fish consumption), wildlife, secondary recreation, agriculture and any other usage except for primary recreation or as a source of water supply for drinking, culinary or food processing purposes.

North Carolina has also adopted water column criteria for mercury of 0.012 µg/l for fresh surface waters. A TMDL is a quantification of the maximum pollutant loading while water quality standards can still be met. In this case, the best usage of waters, which include fishing and fish consumption, need to be met (40 C.F.R. 130.7(c)(1)). TMDLs must identify a numeric TMDL target, a quantitative value used to attain and maintain the applicable WQS, including designated uses, as necessary to calculate the load allocation and wasteload allocation (40 C.F.R. 130.2(i)). No numeric fish tissue water quality criterion for mercury is established in North Carolina; a fish tissue mercury target is therefore needed for this TMDL. The EPA and FDA fish tissue mercury criterion of 0.3 mg methylmercury / kg fish is selected as the target level for this TMDL development. The NC fish consumption advisory action level of 0.4 mg methylmercury / kg fish is not selected, because fish with mercury concentrations at this level will trigger the fish consumption advisory to be in place.

C) Mercury Source and Trend Analyses

- The assessment of mercury sources and trends is sound (PE, DE).
- Pleased to see the significant atmospheric loading reductions (NCWQA).
- Given the international contribution of air deposition of mercury and the minor contribution from NPDES permit holders, it is not clear why the agency is pursuing a TMDL for mercury in North Carolina (DE, TV).
- The draft TMDL should be improved by including a more robust discussion comparing and contrasting the different global sources of anthropogenic mercury emissions. The draft TMDL does a good job of comparing this (from U.S.) relatively small total to the much larger amount of emissions from Asia (on the order of 1,100 metric tonnes). However, the report states that “Together, China, India, and the United States are responsible for 57 percent of the total estimated global anthropogenic emissions of mercury emitted into the air in 2005 (1097 out of 1921 tonnes).” Without the proper context regarding actual U.S. emissions, the statement may give the reader the impression that U.S. emissions are a relatively high proportion to the total, when exactly the opposite is true (PE).

Response:

This TMDL is needed to provide the quantification of total maximum load from known mercury sources, including out-of-state sources. Without this quantification process, it would not be clear that NPDES permit holders contribute 2% of the total mercury loading.

The statement about China, India, and the U.S. was removed from the TMDL document.

D) Model Analysis

- A more sophisticated air model that is readily available and can account for local deposition is needed for the study (171 individuals).
- The TMDL study is flawed or too simplistic; need more studies or a better model (2 individuals).
- DENR did not use a response model to analyze whether reductions will eliminate impairments across the state (WC, NCCN).

Response:

The CMAQ model was used to estimate the total air deposition of mercury within North Carolina and the comparative contributions from in-state and out-of-state air emission sources. CMAQ was developed and is maintained by EPA and regarded as one of the state-of-art air quality models currently available.

A response model that predicts the ultimate results of fish tissue mercury concentrations in North Carolina waters after the reductions are achieved is not currently available. In 2011, EPA's independent Science Advisory Board (SAB), in comments on proposed Mercury and Air Toxics Standards, cited work that supports a linear relationship between mercury loading and accumulation in aquatic biota. The SAB noted that using the CMAQ deposition modeling and proportionality assumption to produce estimates of changes in fish tissue concentrations is considered to be sound.

E) Reduction Needed

- A simple 67% reduction in mercury from 2002 levels is not a sufficient goal for restoring the health of North Carolina's waters (167 individuals).
- 100 % reduction should be the goal (2 individuals).
- Recommend to the Environmental Management Commission that North Carolina require maximum mercury reductions for North Carolina emitters (WC).
- TMDL should include maximum mercury reductions from both point and nonpoint sources (WC).
- The TMDL cannot demonstrate that a 67% reduction from all sources is the correct amount for reducing fish tissue contamination and restoring the health of North Carolina's waters as is required by the Clean Water Act. The EMC should require additional studies and develop an action plan to implement stronger controls if the additional study shows that stronger controls will meet the goal of the TMDL to reduce mercury fish tissue contamination and delist waters from an impaired status (WC).
- The proposed TMDL assumes, without adequate basis, that most of the required reductions in mercury will come from reductions in mercury emissions outside of North Carolina and outside of the United States (CR).

Response:

As explained under section 6.2 of the TMDL document, the reduction factor is the percent reduction needed to achieve the target fish mercury concentration from the existing fish mercury concentration. At this time, neither the mechanisms linking emissions and mercury bioaccumulation nor the effect of a given emissions reduction on fish tissue concentrations are completely understood. Study results and empirical evidence suggest that reductions in fish

tissue mercury are likely to result from reductions in mercury inputs. Therefore we rely on the proportionality assumption regarding the relationships between mercury emissions, deposition, and fish tissue concentrations to conclude that 67% of reduction is needed from all sources of mercury in North Carolina. In 2011, EPA's independent Science Advisory Board, in comments on proposed Mercury and Air Toxics Standards, cited work that supports a linear relationship between mercury loading and accumulation in aquatic biota.

In recognition of mercury source contributions from outside of NC, "North Carolina's Mercury Reduction Options for Nonpoint Sources" was provided for public comment, which includes an option to petition non-NC sources to reduce their mercury release. Under Section 8, Reasonable Assurance, a number of federal and international activities and programs were listed to show reductions are expected from out-of-state mercury sources as well.

F) Load Allocation and Wasteload Allocation

- Supports mercury minimization plans as the mechanism used to address the small contribution of mercury from water point sources (PE, DE, NCLM, UWAG).
- Given the insignificant contribution from the point source, no reduction should be expected from the point source. WLA should be prescribed as baseline loading (UWAG).
- We suggest that DWQ consistently use the term "insignificant" to describe statewide point source loadings in the document (rather than "small" (page 5), "tiny," (page 55) "low," etc.) (NCWQA).
- The TMDL should be revised to clarify that new or expanded point source discharges to surface waters will be addressed pursuant to the permitting strategy. We also suggest that DWQ delete or revise the last paragraph in Section 6.5 (NCWQA).
- Section 6.5 (wasteload allocation) and 6.8 (Final TMDL). These two sections should incorporate by reference the final TMDL Permitting Strategy, as it is amended consistent with our comments (NCWQA).

Response:

This TMDL estimated that point source contributed only 2% of the total mercury loading to the receiving waters. However, this estimated 2% is introduced into receiving waters directly and has an immediate effect. By contrast, the nonpoint sources were estimated as the total air deposition within North Carolina, which include deposition onto water surface as well as deposition onto land and potentially introduced into waters through runoff at a later stage. Therefore, the contribution from NPDES point sources cannot be ignored and a proportional reduction from this source category is needed.

The description of the residual stormwater contribution has been changed from "tiny" to "insignificant" in the text. However, in describing the contribution from NPDES wastewater, for the reasons listed above, we do not believe the effect of total point source loading could be ignored and hence we chose not to use the word "insignificant".

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The last paragraph of Section 6.5 (Wasteload Allocation) was revised to include “New or expanded point source discharges to surface waters will be addressed pursuant to the permitting strategy”.

The wastewater permitting strategy needs to be consistent with the approved TMDL but is not part of the TMDL. An implementation plan is not a required component of a TMDL; therefore, more specific references to the permitting strategy is not made in the TMDL document.

G) Margin of Safety (MOS)

- The implicit margin of safety is based upon four different factors in this section. We believe any of these factors individually would provide an adequate margin of safety and that the combination of the four factors makes the MOS significantly overly protective. Moreover, there are additional implicit margins of safety elsewhere in the document, such as the conservative (overestimation) of point source loadings as described in Section 6.1 (the point source loading “number is likely over-predictive”).
Accordingly, we suggest that the Department acknowledge that the margin of safety may be overly protective and that it can be revisited in the future if a less conservative (yet still legally adequate) MOS becomes desirable. For example, if point source loadings turn out to be slightly above two percent, a small reduction in the large MOS could readily offset any natural variability in the statewide point source mercury loadings (NCWQA).
- Page 57, Section 6.8 Final TMDL. Revise this section as follows: “As described in Section 6.4, a very conservative implicit MOS, based on several factors, is used for this TMDL, and therefore, it is not necessary to include an explicit MOS in the calculations.” (NCWQA)

Response:

Yes, the implicit margin of safety is based upon four conservative assumptions. Unfortunately, these assumptions cannot be quantified exactly, hence there is no line to draw as “overly protective.” Due to the conservative assumptions in the TMDL, the explicit margin of safety is set at zero.

Revisions were made to the TMDL text according to the editing suggested by the commenter.

H) Stormwater

- Page 54, Section 6.5 Wasteload Allocation. Please revise the second sentence in the second paragraph as follows: “~~Although the contribution of stormwater to mercury loading is unknown,~~ ~~the vast majority of mercury from in stormwater that contributes to the impairment of these waters originates from air sources and should~~ will be controlled accordingly.” (NCWQA).
- The TMDL should account for point source contributions from stormwater NPDES phase II permits, which could significantly alter the balance between point and nonpoint contributions (NCCN, CR).
- Supports the decision to not include municipal stormwater NPDES permit-holders in the TMDL permitting strategy (NCLM).
- The effect of stormwater BMP (e.g. pond) on mercury removal needs to be studied (1 individual).

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Response:

Revisions were made to the TMDL text according to the editing suggested by the commenter.

Since the vast majority of mercury in stormwater originates from air sources, this TMDL expects actual reductions in mercury loading in stormwater will be addressed through controls on atmospheric deposition sources that are necessary to meet the load allocation.

This TMDL aims to provide the quantification of a total maximum load from known mercury sources. As described under Section 6.5 of the TMDL document (2nd paragraph of p.55), the best management practices required by stormwater management may also reduce mercury loading, hence a reduction of mercury from NPDES stormwater sources is not proposed in the TMDL. In addition, direct study of the ability of bioretention ponds to remove mercury is still scarce. Instead, sediment was used as surrogate in some reports. The document provided by the commenter is one example.

(http://www.bacwa.org/Portals/0/Committees/Clean%20Estuary%20Partnership/Library/rpt-CEP-SW_Feas-Nov06-09223.pdf). A review of the capabilities of stormwater BMPs in removing mercury is beyond the scope of this document. This type of information may be included in an implementation plan.

I) Daily Load

- Page 57, Section 6.7 Daily Load. We recommend that the Department modify this section as follows:
“Because this TMDL addresses mercury accumulation in fish over long periods of time, annual loads are the only technically ~~more~~ appropriate approach for expressing mercury loading goals. Daily loadings simply cannot be shown to correlate to fish tissue concentrations. There are far too many variables at work to establish such a relationship. Therefore, the calculations and compliance with this TMDL are based on annual loads. However, in order to comply with current EPA guidance, the TMDL ~~is also identifies expressed as a~~ daily load for informational purposes.” (NCWQA)
- Page 58, Table 6-2 TMDL Allocation Summary. We recommend that the Department add an asterisk to the “daily loads” columns in this table noting that the daily loads are shown for “informational purposes” only and that the reader should see the discussion of the appropriateness of implementing the annual loading goals in Section 6.7 (NCWQA).

Response:

Revisions for clarification were made to the TMDL text regarding “Daily Load” and “Annual Load.”

J) Implementation Plan

- The TMDL should be revised to include a detailed, specific adaptive implementation methodology (PE, UWAG).

Response:

We acknowledge that uncertainties are involved in this TMDL study. Language was added to Section 9 (Implementation Plan) to describe an adaptive implementation methodology.

K) Appendix

- Table 2 in Appendix B: Wet deposition number for the zero out run should be 1357.4 instead of 1375.4 (DAQ modeler).

Response:

Corrected as recommended.

Comments regarding the specifics of the Draft Permitting Strategy

A) Level Currently Achieved

- The Level Currently Achieved (LCA) is derived in an unscientific manner, unnecessary and should be eliminated (PE, DE, NCLM, UWAG).
- While we agree the 47 ng/l threshold is reasonably set, we just don't see the need for this exercise and the triggering for MMPs for facilities given the overall insignificant mercury loadings from point sources, particularly where there is no localized water quality issue (NCWQA).
- Instead of evaluation against LCA, evaluate each major point source against the 12ng/l water quality standard for localized impacts in relation to whether a water quality-based limit is necessary (NCWQA).

Response:

As stated in the Permitting Strategy document, 98.5 percent of effluent data from the last 5 years was below the LCA and that 93 % of facilities with mercury monitoring or limits could regularly comply with this limit without the addition of new treatment technology. The LCA is needed in order to avoid local impact and also to stay below the state aggregate wasteload allocation.

B) Mercury minimization plans and limits

- Supports mercury minimization plans as the mechanism used to address the small contribution of mercury from water point sources (PE, DE, NCLM, UWAG).
- We agree that a four year schedule for developing and beginning to implement an MMP is appropriate (NCWQA).
- Major Facilities Receiving New Limits. New limits for facilities should be based only on local water quality concerns and then expressed (annual average) and implemented (through MMPs) (NCWQA).
- New or Expanding Facilities. We disagree with the suggested approach of imposing limits where "there is a potential for mercury to be in the discharge." This will mean mercury limits for every new or expanding POTW. This makes no sense given the larger perspective that the entire point source community is approximately two percent of statewide loadings.

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We believe that new or expanding facilities should be permitted with the sole focus on local water quality standards compliance. If a limit is warranted, it should be expressed (annual average) and implemented (through compliance with an MMP) (NCWQA).

- Modification of Existing Permit Limits. Where the normal reasonable potential analysis shows that there is no longer any reasonable potential to exceed the 12 ng/L water quality-based limit, the limit should be removed and monitoring should revert to the priority pollutant scans (NCWQA).

Response:

The permitting strategy will not necessarily result in mercury limits for every new or expanding POTW. The need for limits will be determined through monitoring and/or wastewater characterization.

C) Stormwater

- Supports the decision to not include municipal stormwater NPDES permit-holders in the TMDL permitting strategy (NCLM)
- We believe that MS4 permits should not impose any mercury-related requirements unless the receiving water has documented water column impairments for mercury. Then an MMP (mercury minimization plan) requirement for the MS4 system may be appropriate. Again, a four year schedule should be provided to develop and begin to implement the approved MMP and compliance with the MMP should establish compliance with any mercury-related effluent limitation or condition in the permit

We agree that the vast majority (if not all) reductions in mercury loadings in stormwater will be achieved through ambient point source controls rather than MS4 best management practices. MS4 BMPs, such as suspended solids and sediment controls, may yield some tangential mercury reduction, yet should not be relied upon to resolve the State's mercury impairment. (NCWQA)

Response:

The suggestion about MMP has been forwarded to the Stormwater Permitting Unit.

D) Monitoring

- Support the monitoring requirement (TV).
- Difficult to support costs of \$150 per sampling event, multiple times annually. Action should be taken in cases where proof supports necessary prudence, such as Reasonable Potential Analysis, on a case-by-case basis (TV).
- DWQ should authorize the use of method 245.7 (1.8 ng/L detection level) as an alternate to method 1631 (NCWQA).
- Given the insignificant overall level of point source mercury loadings, we believe that facilities with local water quality-based effluent limits should monitor for the first two years at a frequency of quarterly. However, after characterizing the mercury loadings over that period, the monitoring should be reduced to either twice-per-year or annual. That would ensure 11-14 samples each permit cycle and that is more than enough in our view to measure an individual facility's insignificant contribution to the insignificant two percent overall point source loadings.

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Quite frankly, after the first permit cycle, the monitoring should be reduced to annually or with the priority pollutant scans (NCWQA).

- For major facilities that do not have mercury limits we believe that monitoring for mercury as part of the priority pollutant scan requirement is appropriate (NCWQA).

Response:

We will evaluate authorizing method 245.7. We believe the monitoring frequencies set for facilities with effluent limits are reasonable.

E) Special Situations

- The TMDL should be revised to provide an off-ramp for stream segments where use attainment can be demonstrated (PE, DE, NCLM, UWAG).

Response:

See p.3 of the Permitting Strategy document for descriptions of Special Situations.

F) Water Quality Standard

- We urge DWQ to revisit the State's water column number for mercury and either remove it altogether in lieu of a whole body fish tissue concentration approach (which would dovetail with the fish consumption advisory approach) or to express the 12 ng/L limit as an annual average concentration. Either approach is scientifically more valid than adopting 12 ng/L as a short-term criterion when we are concerned about long-term fish tissue concentrations (over potentially many years) (NCWQA).

Response:

This suggestion was forwarded to DWQ's Classification and Standards Unit.

G) Reduction Needed

- The implementation plan based on the proposed statewide TMDL should be approved and implemented (NCCN).
- The TMDL won't result in reductions in mercury from 98.5% percent of point sources in the state (WC).

Response:

Mercury in NC wastewater has already been reduced significantly since the TMDL's 2002 baseline. As stated in the Permitting Strategy document, the strategy is designed to maintain mercury loadings from point sources below the wasteload allocation to surface waters as well as prevent localized areas of mercury excursions above the state water quality standard.

H) Clarification Questions

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- P.1 2nd paragraph: How was the baseline calculated? What facilities were used to develop the baseline loading (USN)?
- P.1, 5th paragraph: 7% of the facilities will need additional treatment to comply with mercury limit requirements? What is the compliance timeline/schedule for systems that do not meet discharge limits for mercury (USN)?
- P. 2, Information below 2. Major facilities currently with an Hg limit: If determined that a facility should have a limit on mercury, would monitoring requirements be included in a NPDES permit or will this be enforced by another permit (USN)?
- P. 2, Information below 2. Major facilities currently with an Hg limit: This section does not indicate how long each facility will be given to get in compliance with this requirement? (This information is listed for #3 but not #2) (USN).
- P. 2, New or expanding municipal facilities: Sentence does not make sense – “If there is a potential for mercury to be in the discharge, they will..... (USN)
- This requirement seems to be geared towards the wastewater NPDES program. Are there any plans to monitor mercury through the Stormwater NPDES program (USN)?
- MCB Camp Lejeune currently does not have a Hg limit in its NPDES WW permit or its NPDES Phase 1 Stormwater Permit. WE are not required to analyze for Priority Pollutants on a regular basis; normally these scans are only conducted when renewing the NPDES permit. The NPDES permit would have to be modified to require these scans and their frequency (USN).
- P. 1, 3rd paragraph: Current Priority Pollutant Scan method analyzes mercury utilizing EPA Method 245.2. The current reporting limit for Hg using this method is <0.0002 mg/L or <200 ng/L – which is of no use if trying to compare to a water quality standard of 12 ng/L. If Hg is to be analyzed using EPA Method 1631, either a request to contracting laboratory would have to be made to change the Hg test method or a separate sample would need to be analyzed using just this method. This would make more sense rather than adding a requirement to conduct a Priority Pollutant Scan on a regular frequency; just add the requirement for Hg analysis to the permit (USN).
- Recent process of renewing MCB Camp Lejeune’s NPDES permit required Priority Pollutant Scans to be conducted. Results from sampling in Nov 11 and Feb 12 showed no detections (<0.0002 mg/L or <200 ng/L) of Hg (USN).

Response:

Please see Section 6.1 of the TMDL, Baseline Mercury Load for 2002. Compliance schedules are determined on a case-by-case basis. Please contact DWQ permitting staff for questions about specific permits.