

Modeling Mercury in the Northeast United States

**Prepared by
NESCAUM**

October 2007

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Executive Summary

The northeast states, along with U.S. federal and Canadian partners, documented the state of knowledge of mercury in the environment in a comprehensive report published in 1998 (NESCAUM, 1998). This report covered topics including background information on mercury, how it cycles in the environment, which and in what quantity were the primary emission sources in the Northeast in 1996 (i.e., a 1996 mercury emissions inventory), and how local, regional, and global sources affected the Northeast. Following this publication, the New England Governors and Eastern Canadian Premiers (NEG-ECP) released a Mercury Action Plan (MAP). This plan and revisions thereof outlined the region's goal for virtual elimination of regional mercury emissions, with interim emission reduction goals of 50 percent by 2003 and 75 percent by 2010 (Conference of New England Governors-Eastern Canadian Premiers, 1998; 2001).

In support of the MAP, NESCAUM and the New England Regional Office of the United States Environmental Protection Agency undertook a project to document mercury levels in the NESCAUM states. The intent was to build upon the 1996 inventory to create a 2002 update for tracking progress toward the NEG-ECP mercury reduction goals. In addition, NESCAUM performed updated deposition modeling using an improved model with both the 1996 and newly generated 2002 inventories. NESCAUM conducted the model runs using the Regional Modeling System for Aerosols and Deposition (REMSAD) v7.13 with two specific goals: (1) to determine mercury deposition in the NESCAUM region and apportion the contribution to deposition according to source region and major source category, highlighting differences attributable to emission reductions, and (2) to provide input to aquatic and ecological models that can inform regulatory and policy decisions.

This report documents the modeling exercise and includes a description of the model and inputs generated. Model results were evaluated against a limited set of measurements for validation purposes. This comparison revealed reasonable model performance and suggests that the existing wet deposition network of monitors are not sited in areas most affected by mercury emission reductions in the Northeast. Variability in ambient data indicates the strong influence of year-to-year meteorological changes, which may mask deposition decreases attributable to emission declines.

NESCAUM modeled substantial regional emission reductions totaling over 60 percent from point sources in the Northeast, including Municipal Waste Combustors (MWCs), Medical Waste Incinerators (MWIs), and Sewage Sludge Incinerators (SSIs). Total deposition averaged across the NESCAUM states declined by 37 percent as a direct result. Smaller geographic regions near sources showed even greater deposition declines (over 80 percent reduction) from local emission changes. The analysis demonstrates that substantial local benefits can be achieved from local and regional scale emission reductions. The results also indicate the growing importance of global emissions relative to emissions from within the Northeast, although substantial impacts remain from emissions across the United States.

1. INTRODUCTION

Mercury deposition emerged as an important environmental concern in the northeastern United States in the late 1990s (NESCAUM, 1998). Mercury is a persistent, bioaccumulative, neurotoxic pollutant. When released into the environment and deposited or carried into water bodies, mercury is easily converted to methylmercury, a particularly toxic form of mercury. Methylmercury readily passes up the food chain, accumulating in the tissues of fish and other animals. Ingestion of methylmercury can cause numerous adverse effects in plants, birds, and mammals, including humans.

A major route of exposure to mercury in humans is through the eating of fish. Women of child bearing age are of special concern as methylmercury ingested by a mother can transport across the placenta into the brain of a developing fetus. In young children and fetuses, methylmercury inhibits the normal development of the nervous system, an effect that may occur even at low exposure levels. This damage frequently is not apparent until later in the developmental process, when motor and verbal skills are found to be delayed or abnormal. Developmental effects have been found in children exposed *in utero*, even though their mothers did not experience any symptoms of adult toxicity.

Given recent measurements showing elevated mercury levels in freshwater fish in the region, the eight NESCAUM states (Connecticut, Maine, Massachusetts, New Hampshire, New Jersey, New York, Rhode Island, and Vermont) issued health advisories that recommended limiting the consumption of fish from state water bodies. This is the best immediate approach for limiting exposure to mercury that is already present in the environment. Over the longer term, because most mercury in the Northeast is believed to reach watersheds through atmospheric deposition, decreasing its introduction into the environment by limiting mercury emissions to the atmosphere should permit an eventual lifting of the fish consumption warnings.

A first step to address mercury in the environment was taken in 1998 by the northeast states (through air, water, and waste interstate agencies), along with U.S. federal and Canadian partners, by documenting the state of knowledge of mercury in the environment (NESCAUM, 1998). The report covered a wide range of topics, including: background information on mercury; how it cycles in the environment; what were the primary emission sources in the Northeast in 1996 and in what quantity; and how local, regional and global sources affected the Northeast. Following this publication, the New England Governors and Eastern Canadian Premiers (NEG-ECP) released their Mercury Action Plan (MAP). This plan, and revisions thereof, outlined the region's goal for virtual elimination of regional mercury emissions, with interim emission reduction goals of 50 percent by 2003 and 75 percent by 2010 (Conference of New England Governors-Eastern Canadian Premiers, 1998; 2001).

In support of the MAP, NESCAUM and the New England Regional Office of the United States Environmental Protection Agency (US EPA) undertook a project to

document mercury emission levels in the NESCAUM states. The intent was to build upon the 1996 mercury emissions inventory to create a 2002 update for tracking progress toward the NEG-ECP mercury reduction goals. In addition, NESCAUM performed updated deposition modeling using an improved model with both the 1996 and newly generated 2002 inventories. NESCAUM conducted the model runs using the Regional Modeling System for Aerosols and Deposition (REMSAD) v7.13 with two specific goals. The first goal was to determine mercury deposition in the NESCAUM region and apportion the contribution to deposition according to source region and major source category, highlighting differences attributable to emission reductions. The second goal was to provide input (i.e., loading) values to aquatic and ecological models that can inform regulatory and policy decisions. One example of this is the setting of total maximum daily loads (TMDLs) for mercury in water bodies under the federal Clean Water Act.

This report documents the modeling exercise and includes a description of the model and inputs generated for this work. We compare modeled deposition and atmospheric mercury concentrations where possible to measurements, and discuss differences between the two modeled years of 1996 and 2002. The tagged source results demonstrate the multi-scale impact of mercury emissions on deposition, revealing local, regional and global influences.

2. MERCURY IN THE ENVIRONMENT

Mercury (elemental symbol Hg) exists naturally in the earth's crust at trace levels. This metal can enter the environment through natural (e.g., volcanic eruptions, diffusion from water and land) and man-made processes (e.g., combustion of mercury-containing fuels), after which it may cycle through land, air, and water while undergoing chemical and physical transformations. From the perspective of public health, the concern rests primarily with a toxic organic form, methylmercury, which bioaccumulates in fish, thus exposing people who eat the fish to mercury's toxic effects. Although this report focuses on anthropogenic emissions and their eventual deposition, this section provides a brief overview of the mercury cycle. The context here provides a basis for understanding the importance of tracking the human impact in the global cycling of this pollutant.

2.1. Chemical properties

Mercury is present in several forms in the environment. In the gas phase, two forms dominate: elemental mercury (Hg^0) and its oxidized divalent form (Hg^{2+}). Divalent mercury often binds with other elements (sulfur, oxygen, halogens) as mercuric salts, and may exist in different phases (e.g., gas, particle, or aqueous). Atmospheric particulate mercury is a third species of mercury that is operationally defined as mercury collected in particulate measurement devices (e.g., filters) (Cohen et al., 2004).

Elemental mercury does not readily dissolve in water and has a relatively high volatility. As a result of these characteristics, it exists primarily in the gas phase as only small amounts will dissolve in atmospheric droplets or remain adsorbed onto the surfaces of aerosol particles. Therefore, elemental mercury is removed relatively slowly from the atmosphere, and has an atmospheric lifetime on the order of a year (Cohen et al., 2004, Seigneur et al., 2003; Poissant et al., 2005).

The divalent form of mercury (Hg^{2+}) in the gas phase is often termed reactive gaseous mercury (RGM). RGM is highly soluble, less volatile than Hg^0 , and adheres readily to surfaces. The divalent form of mercury as well as other oxidized states can also exist in the atmosphere as particulate-bound mercury ($\text{Hg}(\text{p})$). Particulate-bound mercury is relatively insoluble and less volatile than elemental mercury. Oxidized mercury in either of these two phases is prone to removal from the atmosphere by wet and dry deposition, and has a considerably shorter atmospheric lifetime (days to weeks) than the elemental form (Cohen et al., 2004).

2.2. Atmospheric processes

Each of the mercury forms described above has a different fate in the atmosphere. Although mercury cycles between its elemental (reduced) and oxidized forms, most of the mercury in the atmosphere (the "global pool") exists in the elemental state (generally >95 percent). This is a direct result of the limited solubility and high volatility of Hg^0 , such that it remains in the atmosphere with a lifetime on the order of one year, free from deposition processes associated with aqueous or particle bound states.

Notably, both Lindberg et al. (2002) and Sprovieri et al. (2005) report an exception to the general preponderance for elemental mercury in the atmosphere that occurs in the Arctic during polar sunrise. The observed behavior in the Arctic reveals rapid conversion of elemental mercury to both reactive gaseous and particle-bound mercury.

With its relatively long lifetime, gaseous elemental mercury can be transported over very long distances, even globally. Thus, emissions in any continent can contribute to deposition in other continents (UNEP, 2002). As noted above, the global pool of mercury is almost entirely elemental mercury. By contrast, reactive gaseous mercury and particle-bound mercury are more readily deposited, thus they have shorter lifetimes of days to weeks and typically deposit within 50 to 500 miles of their source. These forms of mercury tend to have a more local and regional impact.

2.3. Ecological and health effects

Mercury deposited to water bodies can form complexes with organic molecules through a process known as methylation. A number of factors influence the rate of methylation in the water, including the acidity of the surrounding water, dissolved sulfate, and dissolved organic carbon (DOC) levels (Wiener et al., 2006). Acidity and DOC appear to be particularly important parameters, with more acidified conditions and higher levels of DOC frequently associated with higher levels of methylmercury (Kamman, 1998). Methylated mercury in the aquatic food chain can bioaccumulate in fish tissue to concentrations markedly higher than in the surrounding water. Because methylmercury is more toxic than inorganic mercury, and fish consumption contributes to exposure for human beings and other animals, the formation and bioaccumulation of methylmercury in aquatic ecosystems is of particular concern to environmental and public health officials.

Given the various factors that affect the methylation of mercury, different water bodies will contain varied concentrations of methylmercury. Additionally, not all methylated mercury accumulates in fish. It can be de-methylated (i.e., converted back to an inorganic form) or volatilized back into the atmosphere. The spatial and temporal differences in deposition of mercury will also contribute to wide variations in levels of mercury found in fish. Nonetheless, researchers have developed models to help understand these variations and can use the deposition results from the atmospheric modeling reported here as inputs for their ecosystem models.

3. REMSAD

3.1. General description

The Regional Modeling System for Aerosols and Deposition (REMSAD version 7.13) is a three-dimensional Eulerian grid model developed by Systems Applications International, Inc. The US EPA and others have used the model to simulate the physical and chemical atmospheric processes relevant to atmospheric pollutants, including fine particles and air toxics. The model relies on the continuity equation, which represents the mass balance of each species by mathematically tracking emissions, advection, diffusion, chemical reactions, and removal processes.

Model users specify grid spacing and dimensions. Input requirements for the model include meteorological parameters, emission fields, and boundary conditions. Using these inputs, the model solves the continuity equation in a stepwise fashion. For each time step, fresh emissions are added, followed by horizontal and then vertical transport by advection, diffusion and deposition. Chemical reactions are performed, and then transport processes are again performed.

After the model has been run, gridded output is available for analysis. The output is user-specified and generally includes concentration fields for the surface layer and deposition results. Post-processing programs are used to reformat the output for comparison to monitored results in assessing model performance, often summarizing results by relevant time intervals, such as daily or annual average values.

3.1.1. Model framework

REMSAD relies on a three-dimensional grid system, which overlays the region of interest for atmospheric modeling. The model permits grid nesting that enables calculations to be conducted on a finer scale than the overall coarse grid would allow. This capability offers a balance between model extent and resolution, such that a large domain can be modeled, with a refined analysis conducted in specific regions of concern.

Several options exist for the horizontal coordinate system employed in REMSAD. The application described here relied on a latitude/longitude (geodetic) definition, with grid spacing in constant degree definitions. Figure 3-1 displays the map of the domain. The dimensions are 120 by 84 with grid spacing of $\frac{1}{2}$ degree longitude and $\frac{1}{3}$ degree latitude, nominally 36 km grids. The domain ranges from 66-126° West longitude and 24-52° North latitude.

Figure 3-1 Representation of continental 36-km gridded modeling domain

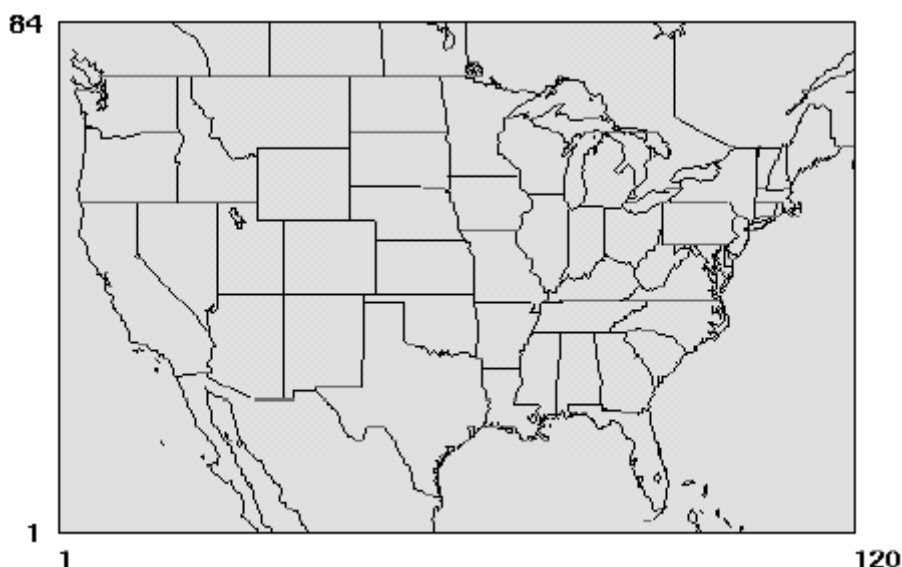


Table 3-1 Sigma pressure layers for US EPA and NESCAUM modeling platforms

14 layer	US EPA Sigma	Height (m)	Pressure (mb)	12 Layer	NESCAUM's Sigma
0	1.00	0	1000	0	1.000
1	0.995	38	995.5	1	0.995
2	0.990	77	991	2	0.988
3	0.980	154	982	3	0.970
4	0.960	310	964		
5	0.940	469	946	4	0.938
6	0.910	712	919	5	0.893
7	0.860	1130	874	6	0.839
8	0.800	1657	820	7	0.777
9	0.740	2212	766	8	0.702
10	0.650	3108	685		
11	0.550	4212	595	9	0.582
12	0.400	6154	460	10	0.400
13	0.200	9626	280	11	0.200
14	0.000	16000	100	12	0.000

The vertical structure of the model is expressed in sigma-pressure coordinates. The two reference pressures are pressure at the surface and at the top of the domain. Any number of model layers can be specified and are generally matched with available output from meteorological models. Here, a twelve layer model was used, with sigma levels

shown in the right-hand side of Table 3-1 ranging from the surface to 16,000 meters (100 mb).

Given the effort required to develop model applications, NESCAUM relied on the US EPA's proposed Clear Skies Act (CSA) modeling platform for its mercury modeling. The US EPA defined the grid system described in the previous paragraphs and created the necessary meteorological and emission fields to drive the model, along with other necessary input files for land surface definitions.

3.1.2. Chemistry

Criteria pollutants

REMSAD v7.13 uses a simplified version of the Carbon Bond Mechanism – version 4 (CB-IV) (Gery et al., 1989) to simulate atmospheric photochemistry. This mechanism (called “micro-CB-IV,” or μ CB-IV) reduces the number of organic species to three, which are grouped as volatile organic species (representing most anthropogenic species), carbonyl species (both as direct anthropogenic emissions and as products of reactions), and biogenic species (kinetically representative of isoprene). The inorganic and radical parts of μ CB-IV remain the same as in the full CB-IV mechanism. Important for purposes of calculating secondary particulate matter, REMSAD includes aqueous-phase (or in-cloud) chemistry that contributes to sulfate formation. For nitrate and sulfate aerosols, REMSAD uses the methodology of Saxena et al. (1986) and Kim et al. (1993) as the basis of an algorithm (known as MARS-A) to account for the equilibrium among sulfuric acid, nitric acid, ammonium nitrate, and ammonium sulfate under local meteorological conditions. While previous REMSAD versions treated secondary organic aerosols as directly emitted species from the emissions inventory, REMSAD version 7 added a new module with a methodology to calculate these aerosols from anthropogenic and biogenic organic precursors (Pankow, 1994; Odum et al., 1997; Griffin et al., 1999).

Mercury

REMSAD incorporates 17 chemical reactions involving mercury species in the gas and aqueous phases based on the review by Lin and Pehkonen (1999). In undergoing chemical transformations, REMSAD tracks the oxidation states and phases (gas or particulate) of mercury. It does not track specific mercury compounds. The species tracked by REMSAD are Hg^0 (gas phase), Hg^{2+} (gas phase), and $\text{Hg}(p)$ (as divalent mercury compounds in particulate phase). The REMSAD chemical reactions result in the transfer of mercury mass from one of these states to another. The REMSAD chemistry assumes some of the Hg^{2+} is adsorbed onto soot particles using primary elemental carbon as an indicator of the amount of soot present. REMSAD contains mercury chemistry involving chlorine that is only active at night. Because REMSAD does not internally estimate chlorine concentrations, it requires an input file to specify these. Chlorine concentrations decrease linearly with altitude, and are also set at different concentrations

at the surface according to whether over land or ocean. Reduction of Hg^{2+} to Hg^0 includes a pathway involving the aqueous formation of sulfide (SAI, 2002).

3.1.3. Transport and diffusion

Air pollution transport occurs primarily through the bulk motion of air (advection). REMSAD depends on the accurate representation of the magnitude and variability of winds to model transport processes (SAI, 2002). Diffusion processes influence the redistribution of air pollutants within the REMSAD domain, and are driven by turbulent eddies that form due to atmospheric gradients. REMSAD handles horizontal transport through an advection scheme developed by Smolarkiewicz (1983). It assumes horizontal diffusion is proportional to the horizontal concentration gradient. Vertical advection occurs through the vertical component of the wind field, which REMSAD calculates through conservation of mass. Vertical winds are typically much smaller than horizontal, and are greatest under convective conditions (e.g., within convective clouds) and terrain- and sea-breeze-induced convergence zones. As with horizontal diffusion, REMSAD assumes vertical diffusion is proportional to the vertical concentration gradient (SAI, 2002).

3.1.4. Deposition

REMSAD simulates wet and dry deposition of air pollutants to the surface. Wet deposition is the removal of gas phase and particulate pollutants from the atmosphere in aqueous form (e.g., in rain, snow, or mist). Dry deposition is the direct transfer of a gas or particulate to the Earth's surface without the aid of precipitation. Mechanisms for dry deposition may include gravitational settling, diffusion, and even surface adsorption by plant-leaf uptake.

REMSAD has two separate treatments for wet deposition of gases and particulates. For gases, it uses a wet scavenging algorithm based on Henry's law and the work of Hales and Sutter (1973). Particulate wet deposition comes from relationships established by Scott (1978) connecting rainfall rate and cloud type with the fraction of ambient sulfate in rain reaching the ground. REMSAD extends this to other aerosol species as an assumed constant fraction of the sulfate rate, depending on the properties of the aerosol species (SAI, 2002).

REMSAD handles dry deposition according to a scheme described by Wesley (1989). For a given species, the dry deposition to the surface is directly proportional to its concentration in the lowest model layer. The proportionality factor is the deposition velocity, which is the inverse sum of a series of resistance terms (e.g., aerodynamic, boundary layer, and surface resistances). Larger resistances for a species result in a slower deposition velocity. The various resistance terms are calculated from parameters related to meteorological conditions and physical traits of the lowest model layer and the underlying surface boundary. These include temperature and pressure, wind speed, moisture stress on vegetation, differences due to water surfaces, and surface moisture (SAI, 2002).

3.1.5. Re-emissions

Current atmospheric chemistry and transport models struggle to accurately parameterize re-emission of mercury from the earth's surface (Lin et al., 2005). Until very recently, models often assumed no net mercury flux occurred from the surface, such that dry deposition rates of elemental mercury were zero and re-emission was not considered.

The REMSAD developers recognized the importance of understanding surface fluxes and introduced a methodology for estimating and tracking these emissions. Their approach calculates the rate of change of available mercury at the surface as the sum of wet and dry deposition minus the re-emission and retention (where retention represents mercury "fixed" to the surface and therefore not labile). The rate of re-emission is two to three orders of magnitude faster than the retention rate and is 0.5 percent of the available deposited mercury. This approach has not been fully evaluated but the current modeling did account for these emissions through the available tagging feature, as described next.

3.1.6. Emissions source tagging

REMSAD employs an attractive feature termed "emission source tagging." The model permits users to track emissions from a specific source, source category, source region, or combination of these by assigning a "tag" to the emissions. The tagging scheme is an accounting system that follows species through space and time in the model without disturbing the physical or chemical processes affecting that species. With careful consideration, the user can establish a model run to assess the impact and influence of particular modeled sources, including boundary conditions.

For mercury, three model species (RGM, gaseous elemental mercury, and particulate mercury) are followed for each tag. The model can track twenty-four different tags, although the first tag is generally reserved for all mercury in the system and the 24th tag is used for the re-emission term. By tagging all sources of mercury in the system, the user can confirm that the model works correctly by comparing the sum of all tags to that of the first tag. Minor differences are the result of numerical diffusion in the model, whereas significant differences would indicate a problem in the implementation of the tagging (e.g., that mercury is not the limiting reagent for chemistry).

3.2. Model inputs

3.2.1. Meteorology

REMSAD requires a number of meteorological fields as input to adequately represent the three dimensional motions of the atmosphere. Six separate files provide hourly values for key parameters (horizontal winds, temperature, surface pressure, specific humidity, vertical diffusion, and cloud/rainfall). This modeling relied on the US EPA input files developed for its modeling of the proposed CSA (US EPA, 2003). As part of that work, the US EPA ran the Fifth-Generation NCAR/Penn State Mesoscale Model (MM5), which is a numerical meteorological model that solves the full set of

physical and thermodynamic equations governing atmospheric motions. They modeled the 1996 calendar year, using 23 vertical layers. These results were then collapsed into the twelve modeling layers used in REMSAD by a post-processor developed to convert MM5 output to REMSAD input files (MM5toREMSAD). Further details on the MM5 options used by the US EPA to develop this meteorology are available in the US EPA's technical support document (US EPA, 2003).

3.2.2. Emissions

Modeling inventory preparation

In 1998, the northeast states worked with the US EPA to model mercury deposition based on a 1996 emission inventory of stationary sources in the region. The study was designed to provide a better understanding of the dispersion and deposition of mercury emitted by sources within the region, outside the region, and the relative contribution of the global reservoir (NESCAUM et al., 1998). During 2003 and 2004, the northeast states updated the mercury inventory for the northeast region used in the 1998 report by including new sources and improving emission estimates for existing sources (NESCAUM, 2005). NESCAUM integrated both the 1996 inventory and the newly developed emissions inventory (EI) with the US EPA's CSA EI, and then processed them into a REMSAD-ready format for two model simulations. The model results were used to assess impact for watersheds and ultimately, fish populations. Emission inventories used and their sources are:

Mercury emissions:

- For emissions outside the Northeast for both scenarios:
 - US EPA's 1999 mercury inventory from proposed CSA of 2003
 - 2000 Canadian mercury emissions (inventory provided by the US EPA)
- For emissions within the Northeast:
 - Scenario 1: NESCAUM's 1996 inventory with pre-1996 NJ emissions from Municipal Waste Combustors (MWCs)
 - Scenario 2: NESCAUM's updated mercury inventory for 2002

Criteria pollutants emissions:

- 2001 "proxy" surface and point emission files for criteria pollutants provided by the US EPA (proposed CSA of 2003)

NESCAUM initially pulled annual mercury emissions from the US EPA's National Emissions Inventory Input Format (NIF) 3.0 emission tables for each source sector and state into one "base" table to prepare summary charts and maps because the parent NIF3.0 files are difficult to manipulate. The files, which include all necessary fields, were exported to MS Excel for easier quality assurance processing, updating, and faster conversion into the Sparse Matrix Operator Kernel Emissions (SMOKE)/Inventory Data Analyzer (IDA) text format supported by air quality modeling.

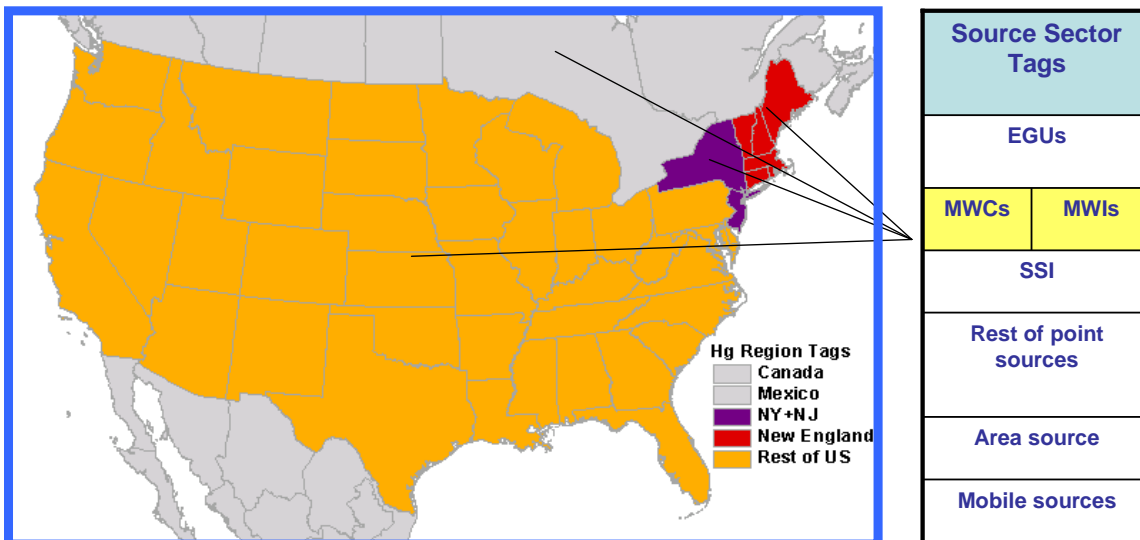
Emissions tagging

REMSAD offers a key feature in its source tagging capability. To take advantage of source tagging, NESCAUM developed in-house emissions tagging techniques for application in air quality impact analyses, effectively adding an additional step to emissions processing. In general, these emissions tagging schemes can be used to assess source contributions in various ways, including by:

- (1) size and susceptibility to transport (e.g., allowing comparisons between large elevated sources vs. small, low-level sources);
- (2) sectors/types (e.g., by source classification codes (SCCs) or by point, area, or mobile source categories);
- (3) regions (e.g., by country/state/county); or
- (4) combinations (e.g., largest electricity generating unit (EGU) in a specific state).

For this research, all combustion and industrial process emissions sources in the modeling domain were tagged by source types/sectors (i.e., EGUs, MWCs, MWIs, SSIs, Rest of Point sources, area sources, mobile sources) and by regions (six New England states, New York/New Jersey, rest of US, Canada). Moreover, boundary conditions were tagged to assess out-of-domain impact. Figure 3-2 shows the tagging scheme employed for this analysis and Table 3-2 displays the resulting emissions. These are shown geographically in Figure 3-3.

Figure 3-2 Illustration of source tagging regions and sectors



Note:

- 1. All Canadian point sources are treated as one tag
- 2. Mobile sources = On-road + Non-road
- 3. The boundary conditions are tagged. MWCs and MWIs are combined for the Rest of the US, while they are tagged separately in the NESCAUM region.

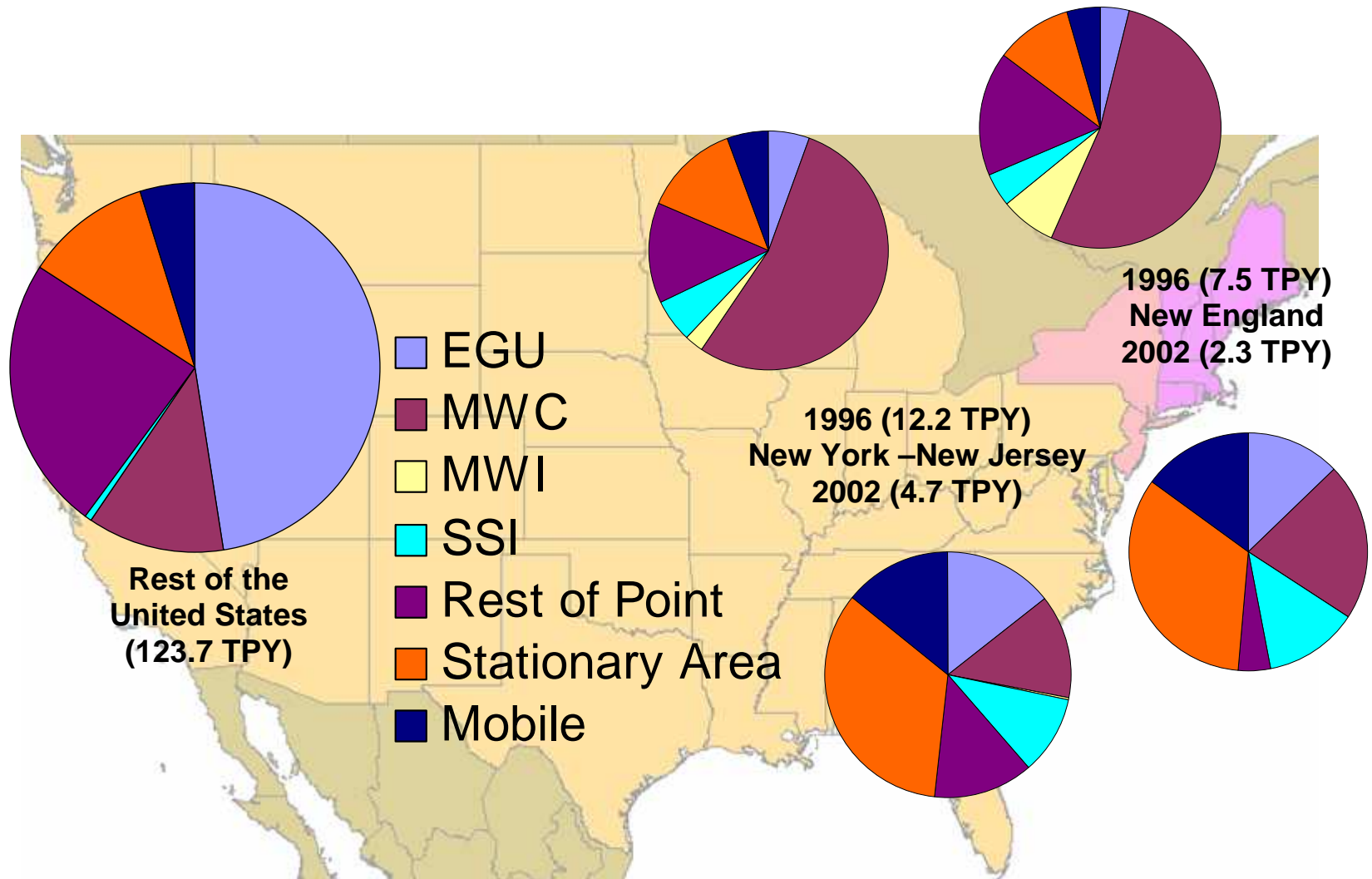
EGU	Electric Generating Unit
MWC/MWI	Municipal Waste Combustor/Medical Waste Incinerator
SSI	Sewage Sludge Incinerator
Other	Rest of point sources (other than above three classes)
	Area sources
	Mobile sources

Table 3-2 Summary of tagged emissions inventory for NESCAUM modeling in tons per year

Source Sectors	Regions							Emission Total (TPY)		
	Tag No.	New England		Tag No.	New York - New Jersey		Tag No.	Rest of US	96 US Total	02 US Total
		96 NESCAUM	02 NESCAUM		96 NESCAUM	02 NESCAUM		CSA		
EGU	1	0.288	0.288	7	0.677	0.677	13	58.835	59.800	59.800
MWC+MWI	2 / 21	3.962 / 0.563	0.482 / 0.003	8 / 22	6.563 / 0.321	0.631 / 0.014	14	14.674	26.084	15.804
SSI	3	0.323	0.288	9	0.728	0.485	15	0.837	1.888	1.610
Rest Point	4	1.255	0.099	10	1.661	0.605	16	29.824	32.740	30.527
Stationary Area	5	0.757	0.757	11	1.590	1.590	17	13.786	16.133	16.133
NonRoad	6	0.332	0.332	12	0.653	0.653	18	5.617	6.603	6.603
OnRoad		0.009	0.009		0.014	0.014		0.162	0.184	0.184
Sum		7.490	2.259		12.207	4.668		123.735	143.432	130.662

Note the **bold red text** highlights substantial emission reductions from specific source categories. NESCAUM region emissions for both model simulations are shown. "Rest of the US emissions" (ROUS) is based on US EPA inventories developed for the proposed CSA and was held constant for both runs. Tag 14 combines emissions from MWC and MWI for ROUS while separate specific tags for each of these source types are applied in the New England and New York – New Jersey regions.

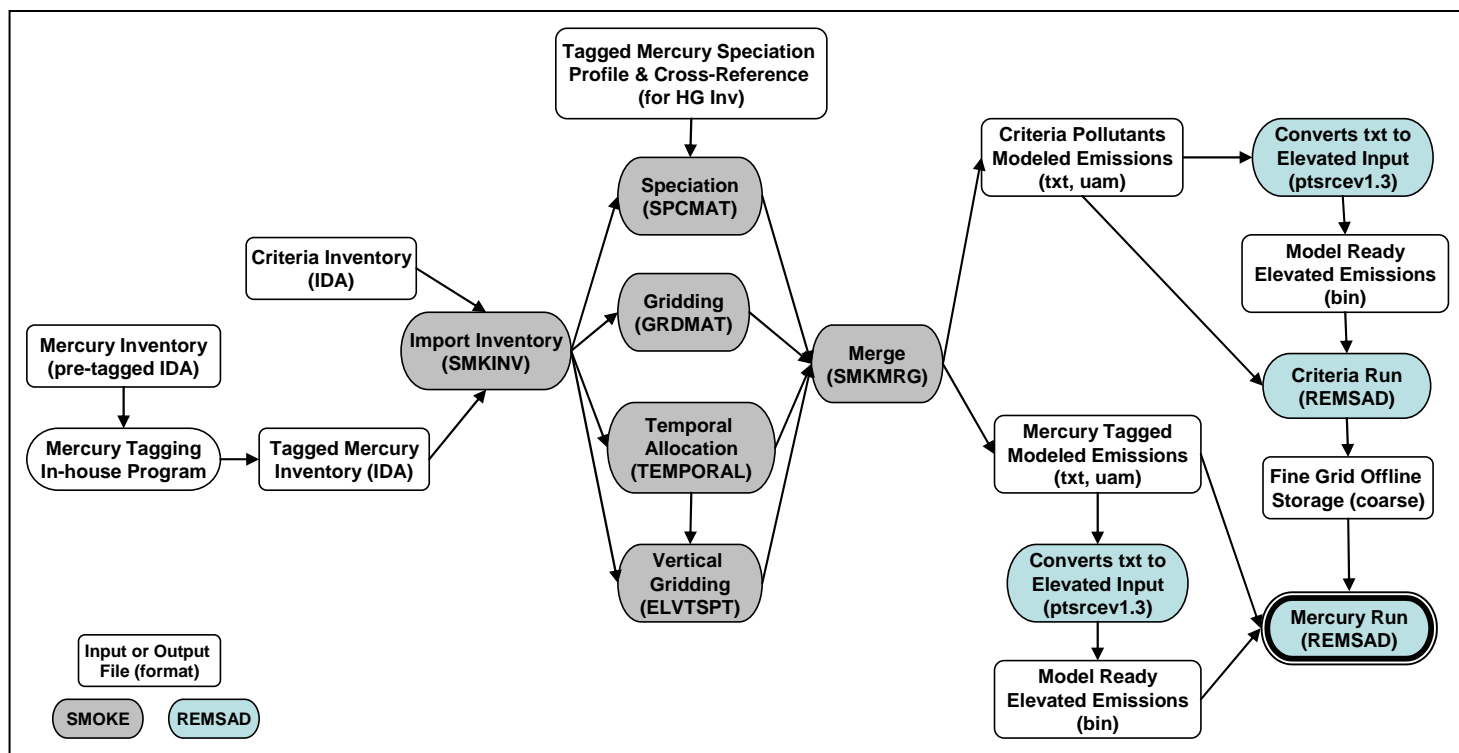
Figure 3-3 Mercury emissions by tagged source category and region



Emissions processing

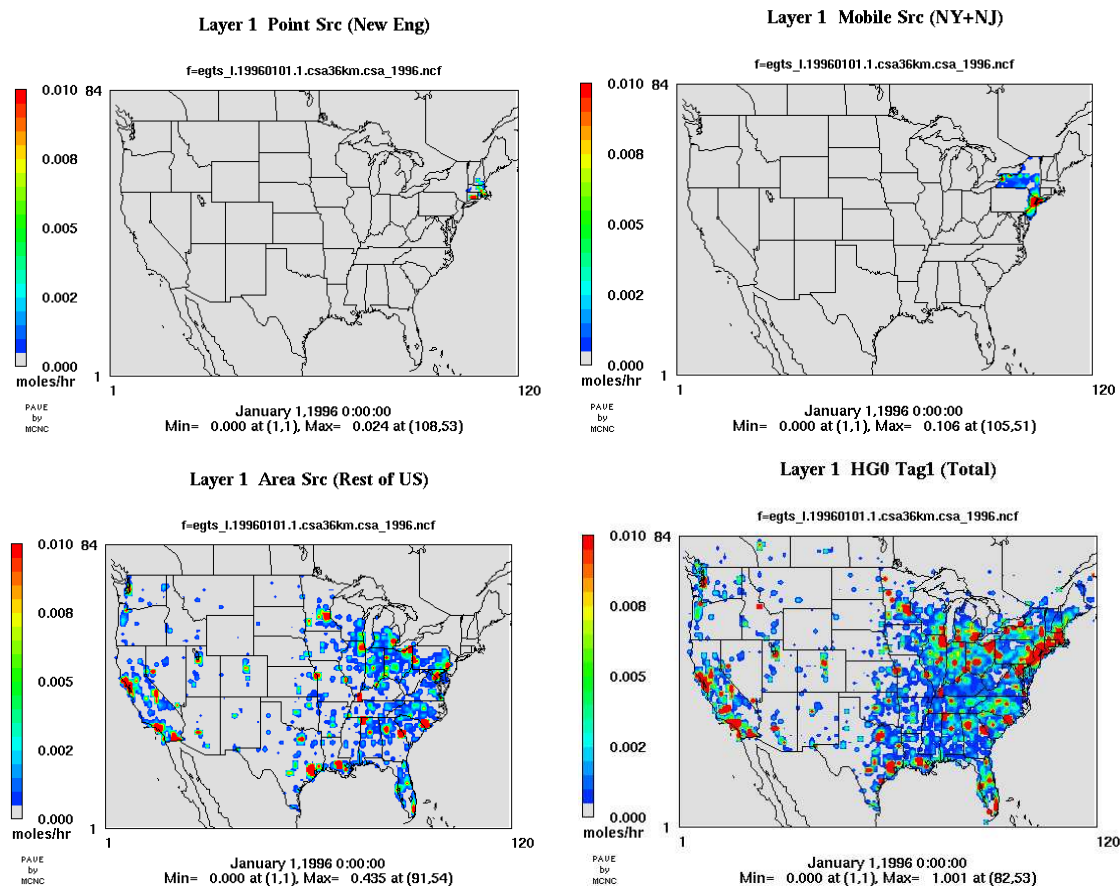
To simulate source emissions, NESCAUM used the Sparse Matrix Operator Kernel Emissions (SMOKE) Version 2.1 Modeling System compiled on a Red Hat 9.0 Linux operating system with the Portland Group FORTRAN compiler version 5.1. SMOKE is primarily an emissions processing system designed to create gridded, speciated, hourly emission input files for a variety of air quality models such as CMAQ, REMSAD, the Comprehensive Air quality Model with extensions (CAM_x), and the Urban Airshed Model (UAM). SMOKE supports area, biogenic, mobile (both onroad and nonroad), and point source emissions processing for criteria, particulate, and toxic pollutants. SMOKE is also integrated with the on-road emissions model MOBILE6.

Figure 3-4 Modeling processing flow chart



To process tagged emissions, NESCAUM updated the chemical speciation related files and the species list to permit SMOKE to process tagged emissions. Other than updating those files, the rest of the processes are the same as conventional SMOKE processing, e.g., gridding, speciation, and temporal allocation. For on-road mobile sources, NESCAUM used pre-calculated emissions supplied with the CSA emissions dataset. Biogenic emissions were not included in NESCAUM’s emissions inventory or emissions modeling/processing. Figure 3-4 shows the entire emissions tagging and processing flows. After processing, SMOKE produces tagged, speciated, and spatially/temporally allocated emissions, as illustrated in Figure 3-5.

Figure 3-5 Four example maps of SMOKE-processed emissions (New England point sources, NY/NJ mobile sources, rest of US area sources, and all sources)



3.2.3. Mercury emission species profiles

As discussed in Section 2.1, mercury can be emitted in several forms and can be transformed among those different forms. Due to significant differences in atmospheric residence time among those species, chemical speciation of emissions directly affects the atmospheric transport and deposition patterns.

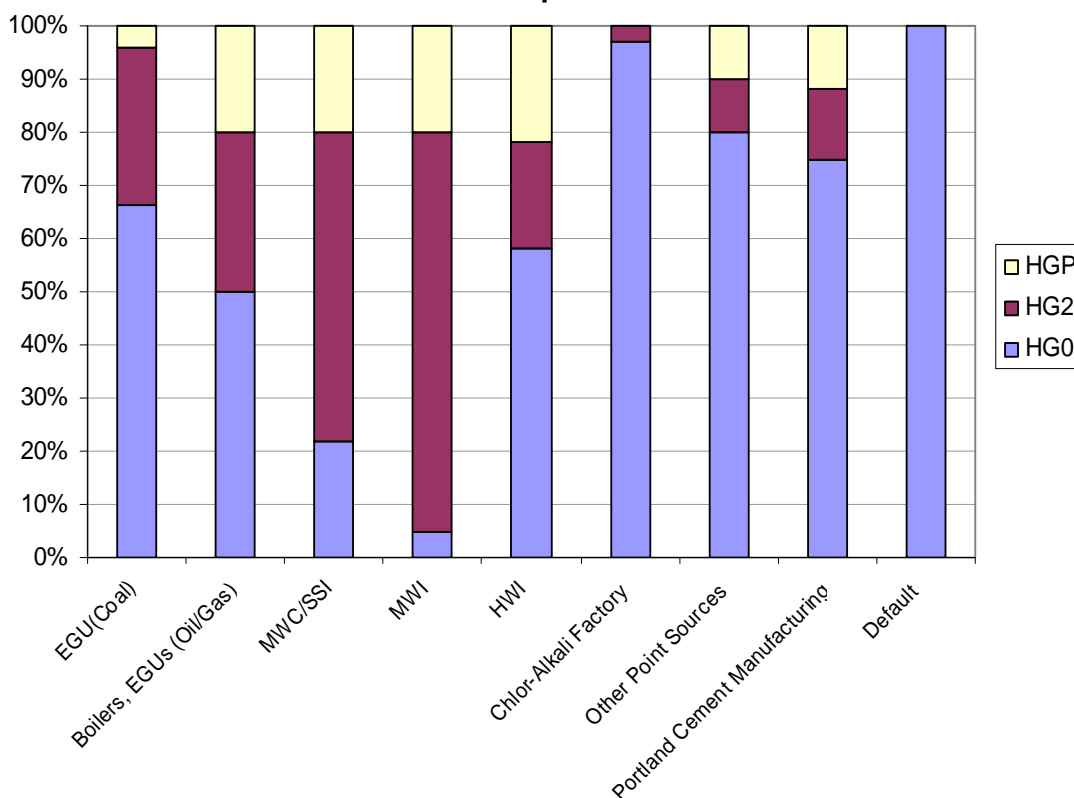
During emissions modeling using SMOKE, NESCAUM modified two major ancillary data sets to incorporate speciation of mercury tagging. One is the inventory table, which is used to read the pollutant codes from the US EPA's National Emissions Inventory and convert them to SMOKE inventory pollutants, and the other is the chemical speciation data used to convert the raw mercury emissions into the species needed by the REMSAD tagging model. NESCAUM added the tagged mercury species to the SMOKE inventory table, speciation profile, and speciation cross reference file so that the SMOKE programs could understand and speciate tagged mercury species. The speciation profiles used were MACT-based speciation data that supports REMSAD version 7 with Micro-CB4 speciation plus mercury. The dataset was provided as part of the SMOKE version 2.1 download package (CMAS, 2005). The speciation profile data

file provided with SMOKE version 2.1 was corrected to use a molecular weight of 200.59 for Hg^{2+} .

NESCAUM ultimately performed emissions speciation outside of SMOKE using in-house software due to the complexity of the combined tagging and speciation processes. This speciation was confined to the eight northeast states because the US EPA's proposed CSA mercury modeling inventory for the rest of the United States was already speciated. Figure 3-6 presents the speciation profiles along with their source categories that are used in NESCAUM's research.

For inter-comparison purposes, NESCAUM compared its speciation factors against the US EPA's most updated modeling inventory from the Clean Air Mercury Rule (CAMR) (US EPA, 2005). The speciation profiles NESCAUM used for NESCAUM region emissions are mainly the same as that of the US EPA for the important source categories, including MWCs, MWIs, and SSIs. The speciation profiles NESCAUM used for coal EGUs (4.2 : 29.3 : 66.5 for $\text{Hg}(\text{p})$: Hg^{2+} : Hg^0) differ, however, from what the US EPA used for its modeling. NESCAUM used the speciation profile in SMOKE, while the US EPA mercury speciation profiles for coal EGUs varied according to coal type and control technology, with averaged speciation profiles ranging from 0.99-9.26 for $\text{Hg}(\text{p})$, 20.29-47.25 for Hg^{2+} , and 43.50-78.72 for Hg^0 across EGUs grouped according to coal type and control technology (US EPA, 2004). Therefore, the REMSAD speciation profile in SMOKE provides one aggregated speciation profile for coal-fired EGUs whereas the US EPA modeling draws upon a larger set of speciated profiles. Because the profile used in CAMR speciates more mercury into the reactive portion, it may model more mercury as being deposited near the EGU source. This difference, however, would not be significant for EGUs in the Northeast because they do not dominate the region's sources (~16 percent of total mercury emissions).

Figure 3-6 REMSAD mercury speciation by source category



3.2.4. Boundary conditions

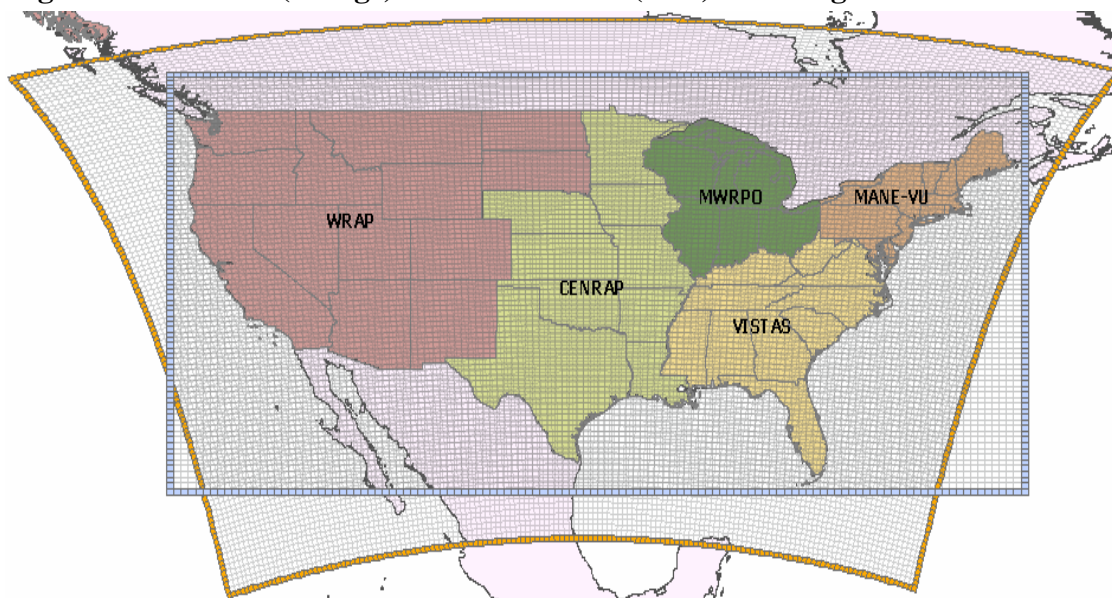
Preliminary modeling used static boundary conditions for the mercury simulation. As done by the US EPA, NESCAUM decided to adopt a dynamic boundary condition to better represent the changes in mercury concentrations in time and space. For its 2001 modeling efforts, the US EPA had relied on output from a global three-dimensional atmospheric transport and chemistry model, GEOS-Chem, driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office. The global model provided boundary conditions for fourteen atmospheric layers every third hour for an entire year on a 36-km Lambert Conformal domain.

The NESCAUM model platform described in 3.1.1 did not exactly match these dimensions. In addition, inputs varying every third hour were deemed excessive and not likely to be more accurate than monthly averaged diurnal profiles. The GEOS-Chem results, therefore, were reduced to 96 distinct vertical profiles per boundary grid column, eight per day (every three hours), one set of eight per month. These averaged conditions permitted model inputs that varied on a diurnal and monthly basis, reflecting changes on these timescales.

In addition to the temporal averaging, spatial adjustments were required to use the GEOS-Chem output. Vertically, two pairs of layers in the GEOS-Chem output were collapsed to their corresponding layers for REMSAD, converting the fourteen layer values into a twelve layer domain (Table 3-1). Horizontally, the REMSAD geodetic domain was matched to the Lambert Conformal domain by averaging at each vertical layer the three cells nearest in horizontal distance (Figure 3-7).

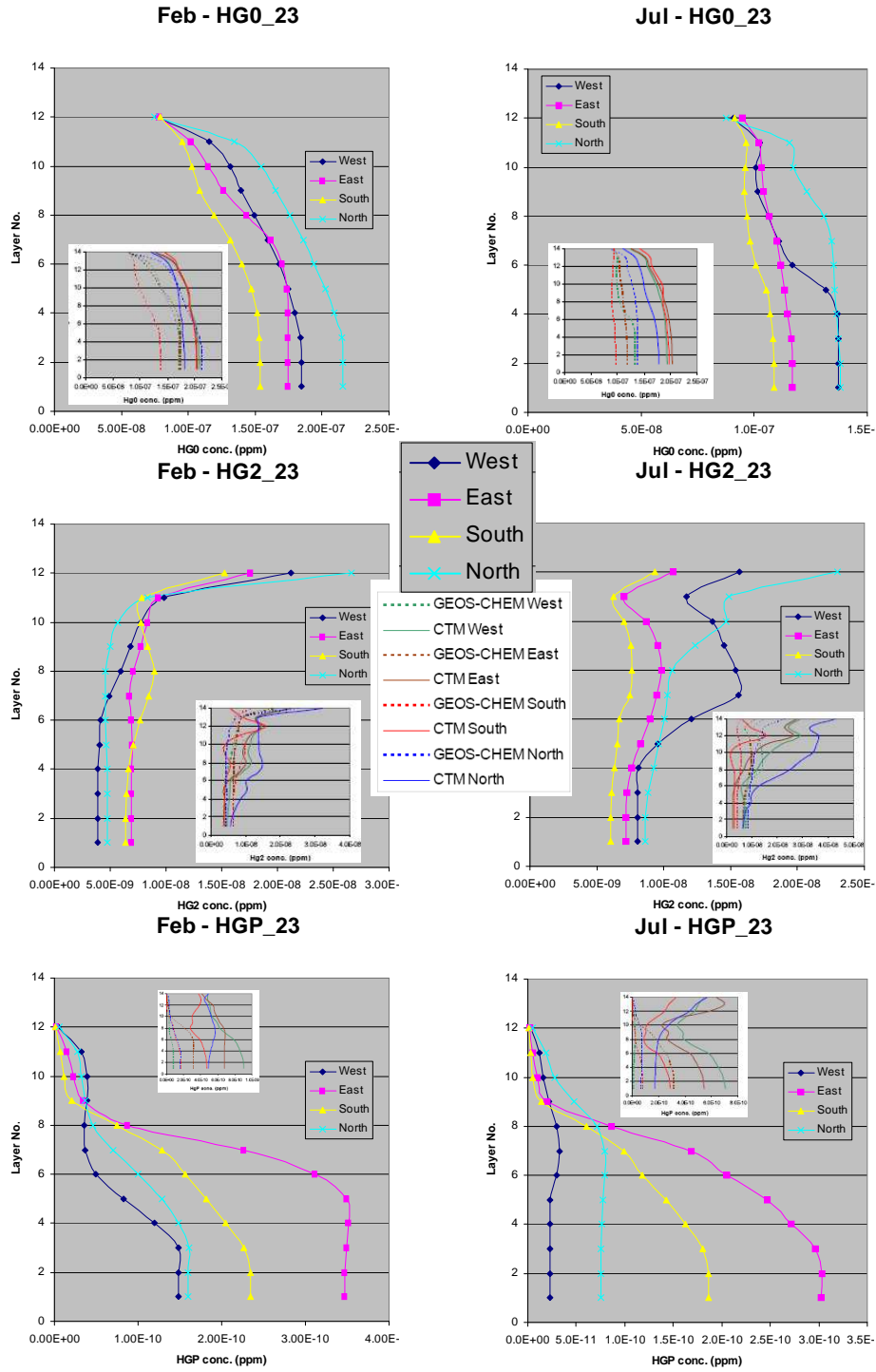
Quality assurance plots were created for two different seasons for all three mercury species to confirm the accuracy of the NESCAUM averaging procedure for boundary conditions. These results are shown in Figure 3-8. NESCAUM profiles are plotted in the main graphic, with the US EPA profiles provided in the inset figure. The inset figure also shows differences between the GEOS-Chem profile and profiles created by a different global model. Based on these plots, the NESCAUM-averaged boundary conditions were deemed equivalent to the parent GEOS-Chem results.

Figure 3-7 US EPA (orange) and NESCAUM (blue) modeling domain boundaries



The two insets show fairly different profiles based on different models and inputs. The associated variations in global model results can drastically influence the impact of boundary conditions in continental-scale modeling. Subsequent to NESCAUM's modeling, GEOS-Chem developers adjusted emission inventories and model chemistry to improve model performance relative to limited available measurements. A comparison of mercury speciation and levels between these two model runs was conducted and showed that the older model run had lower total mercury levels but higher RGM values.

Figure 3-8 Comparison of global model-generated speciated Hg boundary conditions (original US EPA GEOS-Chem boundary conditions shown in insets)



4. MODEL EVALUATION

4.1. Mercury monitoring in the Northeast

For evaluating model results, it is possible to compare the REMSAD outputs to readily available mercury monitoring datasets, keeping several important caveats in mind. There are several ongoing and historical data records of mercury levels in the Northeast. Most of the measurements are focused on wet deposition, while some more limited data are available for ambient levels in the surface layer of the atmosphere.

As with any model to monitored data comparison, a number of caveats exist. For this work, significant items of note include:

- model meteorology is for 1996, while modeled emission inventories are a mixture of years (details available in 3.2.2);
- monitored data cover a number of years, none as old as 1996; and
- grid size represents roughly 36 km by 36 km, which is very large compared to point measurements.

4.1.1. Wet deposition monitoring data in the Northeast

The primary source of monitored mercury wet deposition for the Northeast is through the Mercury Deposition Network (MDN), which functions as a sub-network of the National Acid Deposition Program (NADP). The data record varies by year and location. These samples are collected over a week-long period and are intended to provide information on the spatial and temporal deposition patterns of mercury, including trend tracking. Other data sources are available in the region, including event-based samples analyzed by University of Michigan.

Because direct model to monitor comparisons cannot be conducted given the limitations of this study, we provide a general review of the monitored data, along with corresponding model results. First, annual wet deposition data are plotted to show behavior over time for a select set of monitors in the Northeast's model domain (Figure 4-1). As seen in the figure, and documented previously (VanArsdale et al., 2005), we do not observe the anticipated deposition decreases due to regional emission reductions. A number of factors could explain this lack of correlation, although most are not tested by this modeling study. For example, a significant decrease in Northeast mercury emissions occurred in the late 1990s.¹ The MDN data available for comparison with this study, however, extend back in time only to 1997, and then only for a few monitoring sites. As a result, a representative "baseline" period does not exist in the deposition record. Additionally, the MDN monitors deposition in remote areas, generally far from local

¹ In October 1995, the US EPA issued final regulations for MWCs designed to reduce emissions by 90 percent from a 1990 baseline by December 2000. Where applicable, the northeast states adopted MWC mercury limits almost three times more stringent than the federal limits. In August 1997, EPA issued emission standards for MWIs with compliance by September 2002. These were designed to reduce emissions from these sources by 94 percent relative to 1990 levels.

source contributions. Model results summarized in Figure 4-2 show that decreases in wet deposition in the Northeast due to emission reductions are less than 20 percent for 60 percent of the region. The cells with substantial modeled deposition reductions do not contain MDN ambient monitors.

Figure 4-1 Time series of wet deposition from Mercury Deposition Network (MDN)

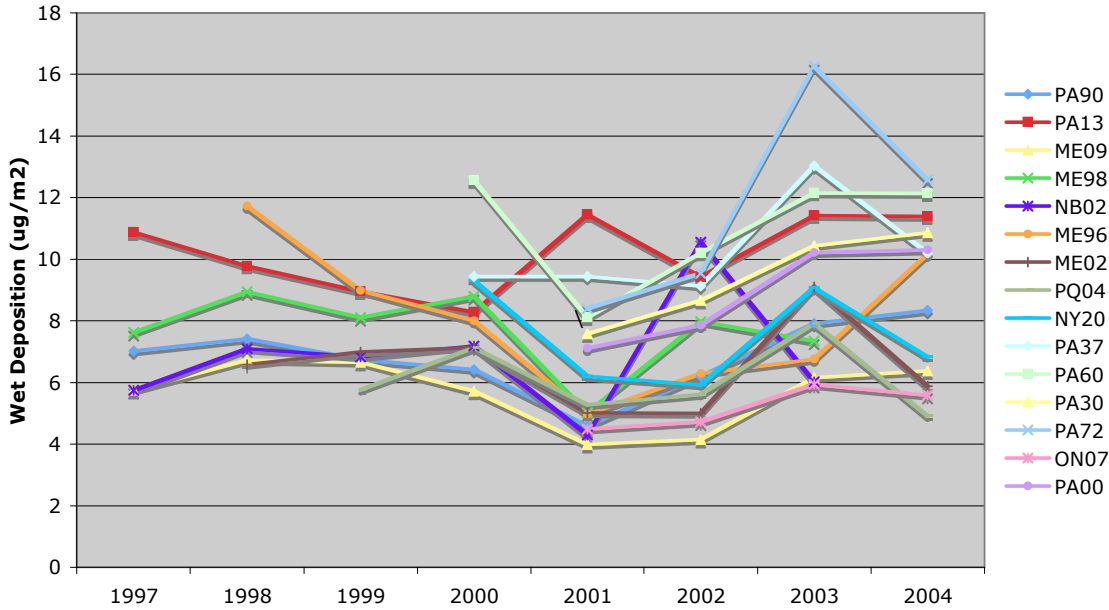
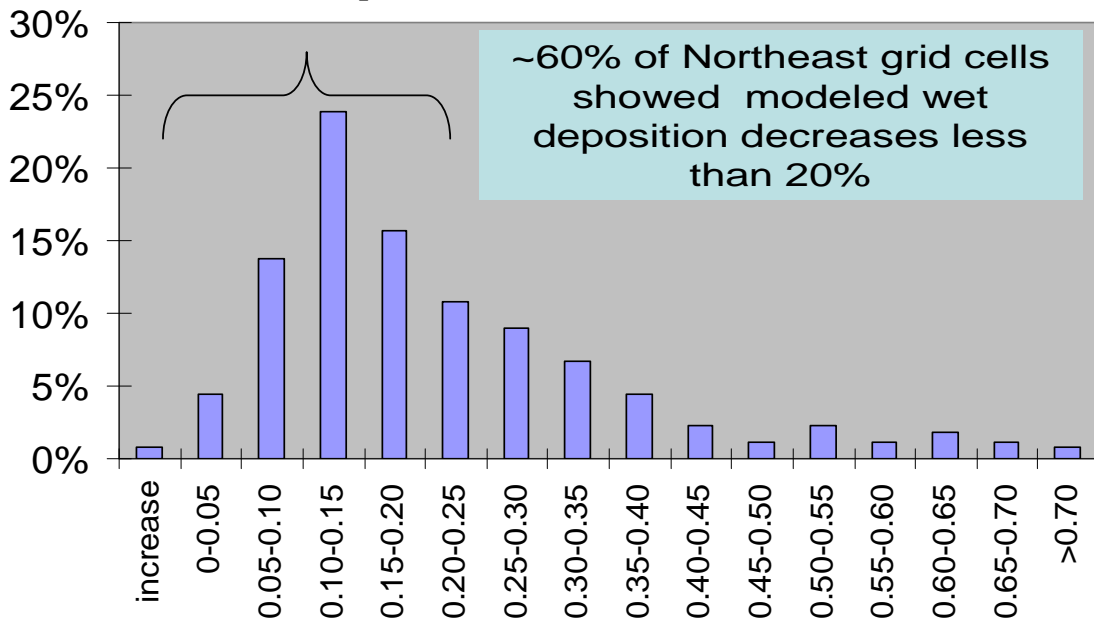


Figure 4-2 Percentage of monitors (y-axis) with modeled percent change in wet deposition from 1996 to 2002 (x-axis)



Since no 1996 deposition results are available for comparison, an alternative evaluation of the data is presented in Table 4-1. Eight MDN sites with data covering most years from 1997 to 2004 are compared to model results from the corresponding grid cell deposition output. In the first two columns, four-year average wet deposition was calculated from the monitoring dataset. A few sites had fewer than four complete years of data, with the number of years noted in parentheses. The next pair of columns shows the modeled deposition for the 1996 and 2002 emissions scenarios. Percent deposition change is tabulated in the third column pairing. The relative monitored and modeling changes are reasonably similar, although in two instances the monitored averages show an increase in deposition (PA13 and NB02). The model predicts much greater deposition decreases in two Maine locales (ME02 and ME51), which reflects substantial modeled emission reductions. The final two columns compare the absolute modeled and monitored deposition, expressed as the ratio of the modeled to monitored deposition. The model captures the average deposition within a factor of two.

Table 4-1 MDN data compared to modeled data at select MDN sites. Skill reflects the ability of the model to capture the monitored average wet deposition ($\mu\text{g}/\text{m}^2$).

MDN Site	Average 97-00	Average 01-04	1996 Model	2002 Model	Monitor	Model	Skill 1996	Skill 2002
PA13	9.47	10.92	17.46	17.36	15%	-1%	1.84	1.59
PA90	6.90	6.76	6.49	6.01	-2%	-7%	0.94	0.89
PQ04	6.44 (2)	5.93	3.68	3.24	-8%	-12%	0.57	0.55
ME02	6.90 (3)	6.25	9.38	6.34	-9%	-32%	1.36	1.01
ME96	9.57 (3)	7.05	12.97	6.36	-26%	-51%	1.36	0.90
ME09	6.21	5.17	3.56	2.94	-17%	-17%	0.57	0.57
ME98	8.37	6.77 (3)	4.75	4.14	-19%	-13%	0.57	0.61
NB02	6.71	6.96 (3)	7.06	6.38	4%	-10%	1.05	0.92

Figure 4-1 does highlight the inter-annual variability that exists, both within a site and between sites. This likely reflects changes in wet deposition patterns from year to year. Broad regional precipitation patterns could drastically affect wet deposition patterns. For example, drier than normal conditions upwind of the Northeast might allow for a greater transport fraction of mercury than wetter conditions. Figure 4-3 shows statewide average annual rainfall from 1995 through 2004 (National Climate Data Center, 2005). The 2001 figure reveals a much dryer than normal year, which is reflected in the time series of mercury wet deposition in Figure 4-1. Years 2003 and 2004 show above normal precipitation for much of the Northeast, and may be associated with the relatively high mercury wet deposition observed in those years.

The same data plotted as a time-series in Figure 4-1 are replotted in Figure 4-5 as ranges of site-specific results. In addition, the two model results are shown as hollow symbols for comparison. An obvious trend does not exist, with the model showing results that are lower, higher, or in the range of the monitored values. In some instances, for example, the model under predicts in Ontario, Canada, which may be due to missing emission sources in Canada or to under-representation in the model of true northern boundary conditions. However, the model generally over predicts the results in

Pennsylvania. This could be a direct result of the meteorology of 1996, which was the wettest year on record.

Figure 4-3 Annual rainfall patterns in the eastern US 1995-2004 (20-year trend for Northeast shown at bottom)

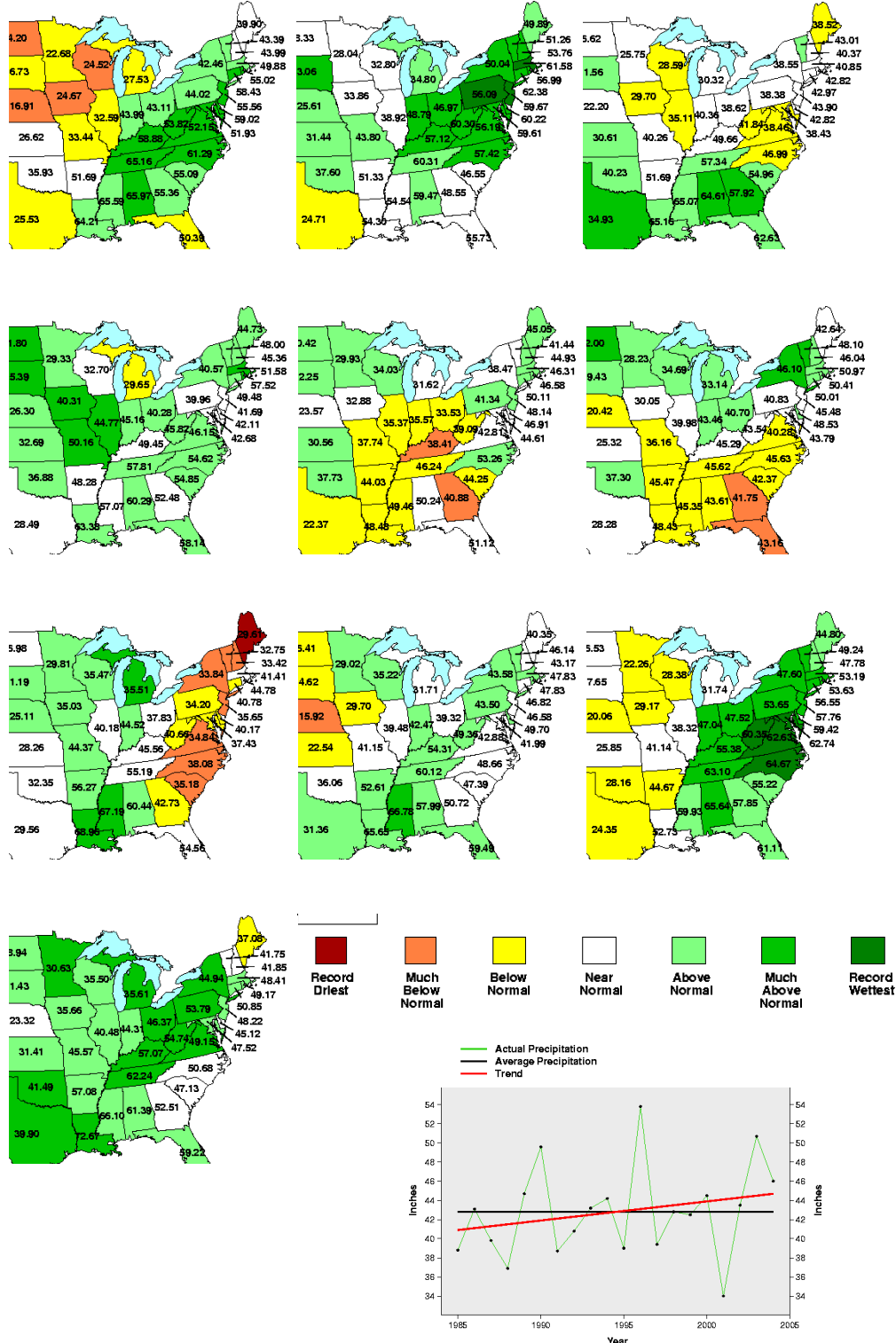
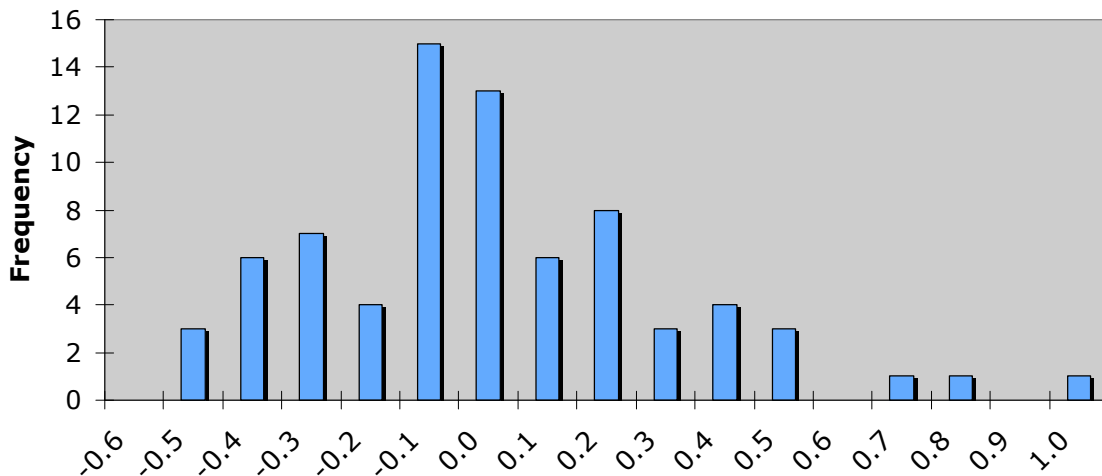


Figure 4-4 shows the monitored year-to-year variation for fifteen MDN sites as a percentage change, expressed as the difference between the current and prior year deposition divided by the prior year deposition. Variability ranges from a decrease of 50 percent to a doubling of deposition. These monitored changes, since they occur from year to year, are not likely to reflect emission changes directly, as those would occur gradually over time as controls come into place. Presumably, these changes directly result from interannual meteorological differences. These measured changes are in most cases comparable to the deposition changes modeled at the monitor grid-cells attributable to emission reductions.

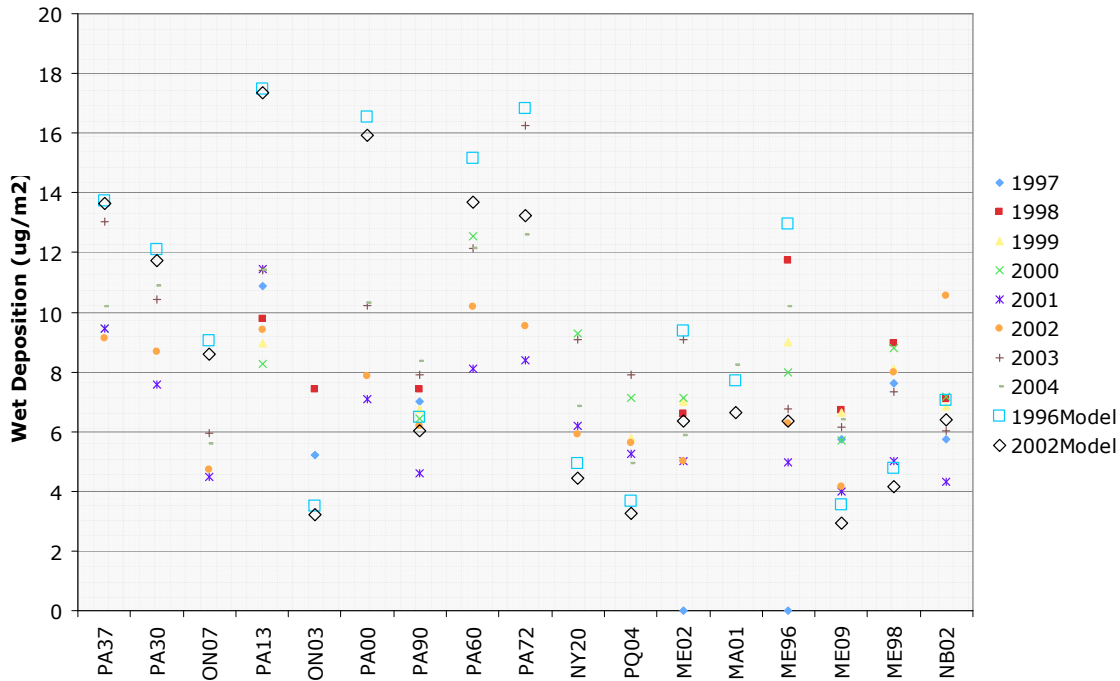
Figure 4-4 Monitored year-to-year percent change for 15 sites in Northeast



Results for two nearby MDN locations in Maine were evaluated versus model results for the two corresponding grid cells. These two cells provide an important contrast because one has several point sources and reasonably substantial area source emissions within the cell (ME96 monitor) while the other does not (ME02 monitor). Figure 4-5 shows that the model captures the relative differences between these two sites. Substantial emission reductions near the ME96 monitor led to the large decline in modeled deposition for 2002. Generally, the two modeled results reflect the range of monitored concentrations quite well.

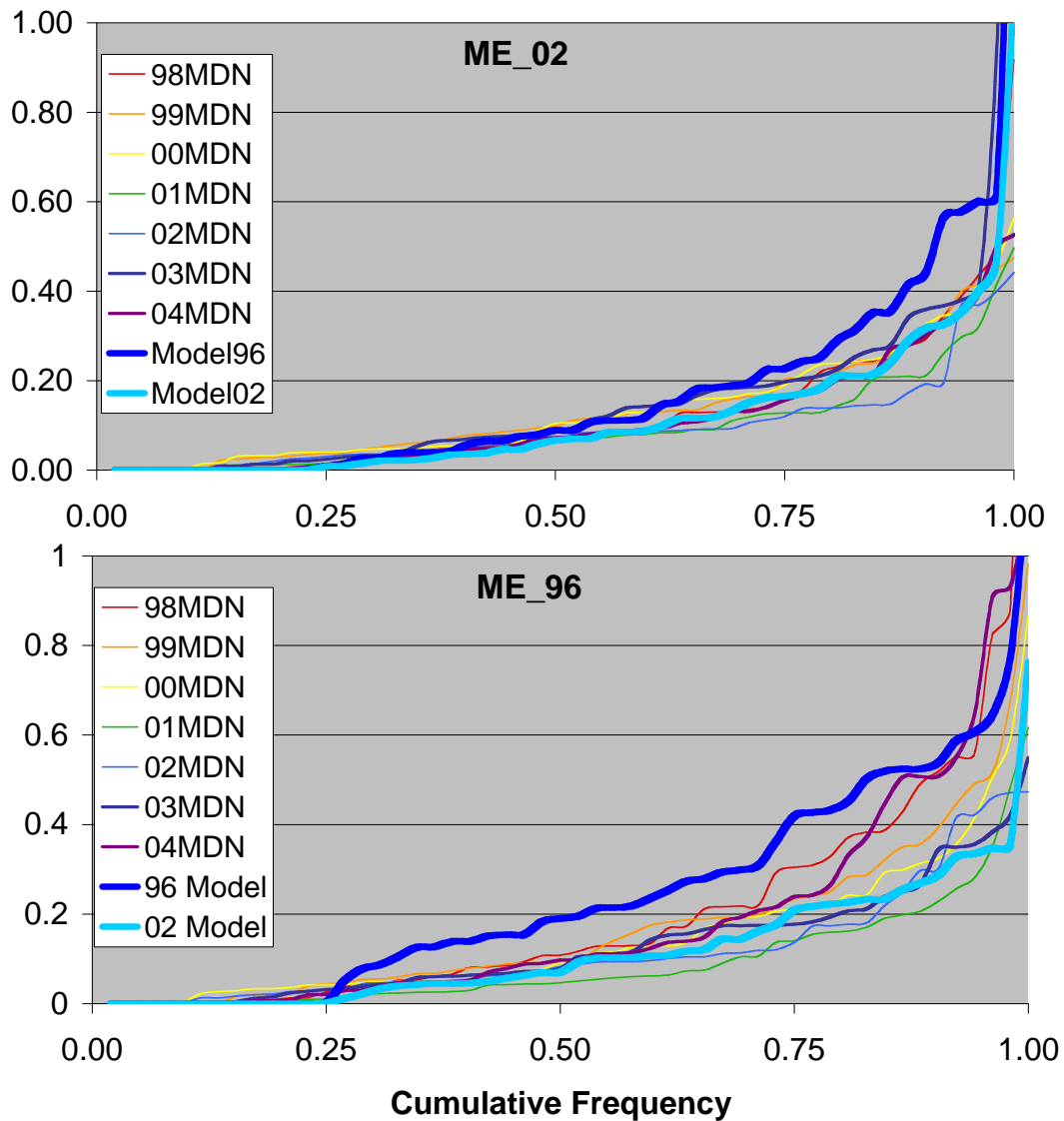
To analyze the model performance for this grid cell pair further, plots were generated to compare the weekly monitored results to the modeled weekly deposition averages. The results are not paired, which would not make sense given the different meteorology from year to year, but instead are ranked from low to high. Figure 4-6 shows that the model generally reflects the week-to-week differences in deposition. The results are especially good for the 2002 run, which falls within the observed year-to-year variability from monitored wet deposition.

Figure 4-5 MDN deposition ranges compared to modeled wet deposition



In addition to monitor specific comparisons with individual grid cell results, spatial patterns can be assessed. Figure 4-7 plots monitored wet deposition results from 1996 and 2003 next to the modeled deposition fields. Both model and monitor results show a general spatial trend of higher deposition in the mid-Atlantic region with lower values in northern Maine. Both also show a localized deposition hotspot on the coast of southern Maine, especially apparent in the 1996 maps. These maps also display one of the advantages of deposition modeling versus a sparse observation network. The model reveals high deposition in New York around the Great Lakes. Since no monitors are located there, spatial interpolation of monitored values fails to show this. As noted before, the model over-predicts wet deposition in Pennsylvania.

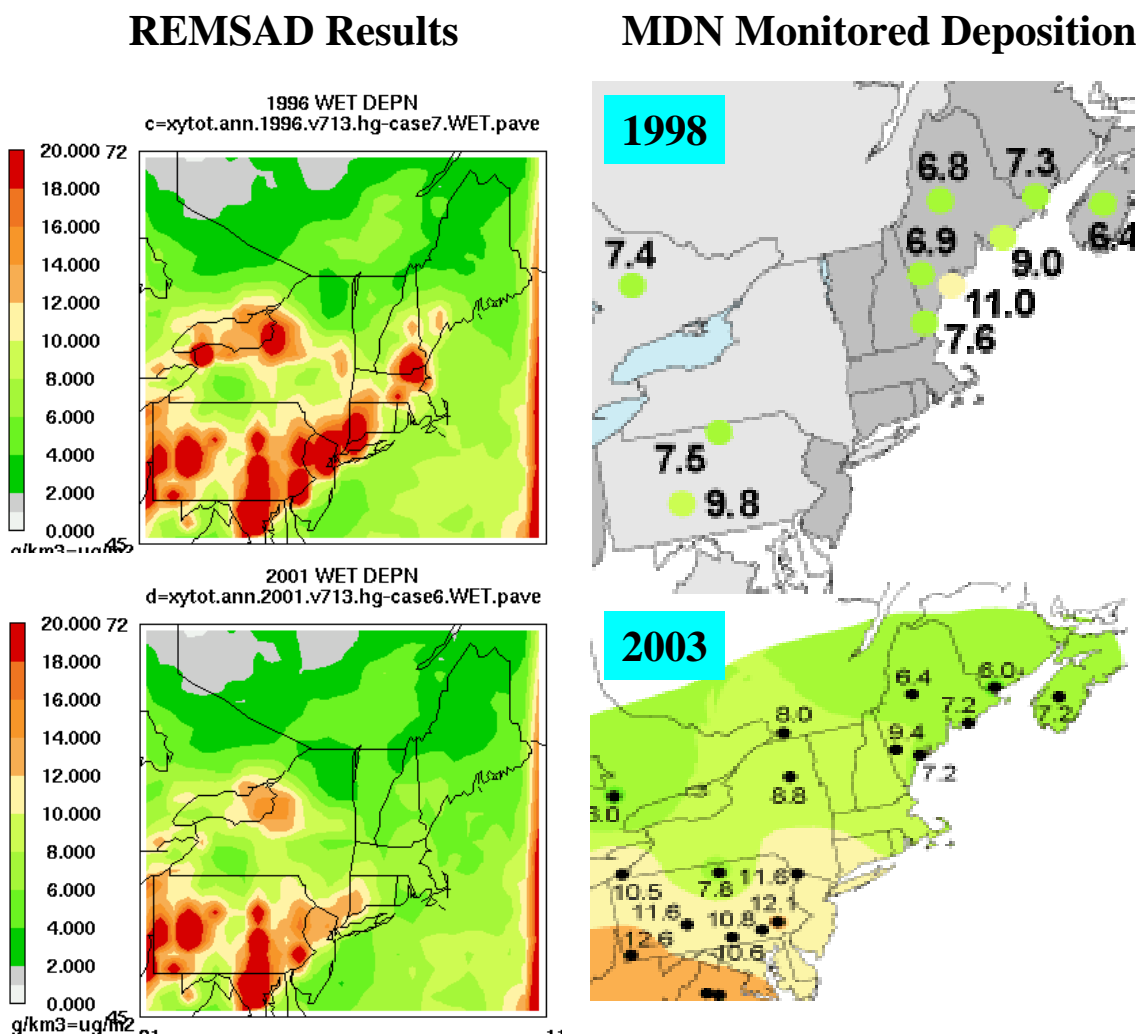
Figure 4-6 Cumulative frequency plot of monitored and modeled weekly deposition



4.1.2. Dry deposition

Although a substantial fraction of the total modeled mercury deposition is attributed to dry deposition, there are no corresponding measurements for evaluation purposes. Recently published studies of dry deposition from Nevada (Lyman et al., 2007) and Florida (Marsik et al., 2007) discuss the limitations and challenges of different approaches for making dry deposition measurements. The studies also note substantial variability of dry deposition in time and space, based on their measurements.

Figure 4-7 Spatial comparison of modeled and monitored results for two years



4.1.3. Measurements of elemental mercury vapor in the atmosphere

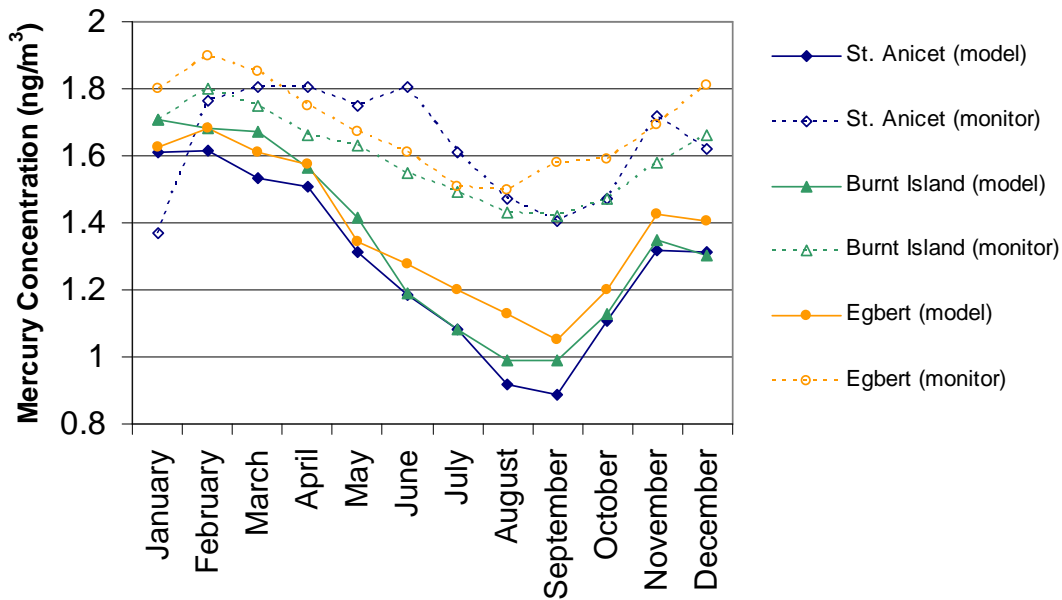
In addition to deposition measurements, some researchers measure ambient levels of mercury at the surface. Currently there is no national ambient mercury monitoring network. However, several research groups have published measurements conducted around the Great Lakes, including measurements in Canada (Poissant et al., 2004; Kim et al., 2005) and in New York (Han et al., 2004). Annual-averaged measurements were compared to surface concentrations predicted by REMSAD, as shown in Table 4-2. In general, modeled elemental mercury levels are 15-50 percent lower than those measured. The model substantially overestimates levels of Hg^{2+} at the surface by an order of magnitude. Given the current high level of uncertainty in both measured and modeled values, assessing the importance of these differences is difficult.

Table 4-2 Annual average Hg concentrations at the surface

Location	Modeled Values (ng/m ³)		Monitored Values (ng/m ³)	
	Total Hg	Hg ²⁺	Total Hg	Hg ²⁺
Stockton, NY ^(a)	1.23	0.056	1.83	0.006
Sterling, NY ^(a)	1.30	0.066	2.59	0.006
Potsdam, NY ^(a)	1.25	0.035	1.84	0.004
St. Anicet, Canada ^(b)	1.29	0.033	1.68	0.003
Egbert, Canada ^(c)	1.41	0.070	1.69	
Point Petre, Canada ^(c)	1.32	0.049	1.93	
Burnt Island, Canada ^(c)	1.34	0.032	1.58	
Great Mountain, CT ^(d)	1.29	0.051	1.60	

^(a) Han et al., 2004; ^(b) Poissant et al., 2004; ^(c) Kim et al., 2005; Sigler et al. (2006)

Figure 4-8 Comparison of monthly average total mercury concentrations



Comparisons of monthly averaged total mercury were conducted for three sites, as shown in Figure 4-8. The model results are plotted with solid lines, while the corresponding monitored results are plotted with dashed lines, matching in color with the appropriate modeled result. The model captures the monthly trends reasonably well, showing minima in late summer and fall with maximum levels in the springtime. The magnitude of the month-to-month changes is greater in the modeled results.

5. MODEL RESULTS

5.1.1. Inventory analysis vs. modeled results

One of the simplest approaches for understanding the sources of mercury in the environment comes from emission inventory estimates. Based on the inventories used for this work as detailed in Figure 3-3, both MWCs from within the NESCAUM region and EGUs from outside the region appear as the dominant sources of concern for 1996. After controls were put in place in the NESCAUM region, local stationary and area sources of mercury appear to rise in importance. This simple analysis neglects the form of mercury in the emissions, unlike air quality modeling of mercury.

The model incorporates information on the various forms of emitted mercury. Equally important are chemical and meteorological processes dealt with by the model. Combined with the tagging capability in REMSAD, the results provide a better estimate of which source types and source regions contribute significantly to the mercury levels observed in the Northeast.

5.1.2. Contribution by source region

As an area impacted by mercury deposition, NESCAUM states want to understand the extent to which emissions within the region account for observed mercury deposition. The Mercury Action Plan of virtual elimination of in-region mercury sources was motivated by the belief that local sources contribute substantially to the local mercury problem. Based on these modeling results, the local reductions in mercury emissions have a profound effect on the magnitude of local deposition.

Analysis of deposition across the entire NESCAUM region is shown in Figure 5-1 and Figure 5-2. The stacked bar chart shows annual deposition results for the two model runs, split both regionally and by deposition type (wet or dry). Since the boundary conditions and emissions from the area outside the Northeast were held constant, no changes were observed in their modeled deposition totals. Substantial decreases in both wet and dry deposition are modeled as a direct result of reduced mercury emissions in NESCAUM states. Wet and dry deposition contribute equally in the 1996 run, whereas modeled wet deposition in 2002 exceeds that from dry deposition by about 20 percent. The pie charts of Figure 5-2 show that the in-region contribution to deposition declines from nearly half of the modeled deposition in 1996 to about one-sixth of the total in 2002. This highlights the relative increase in importance of emissions outside the Northeast to deposition within the Northeast as a result of local emission reductions.

Figure 5-1 Regional contribution to wet and dry deposition in NESCAUM states

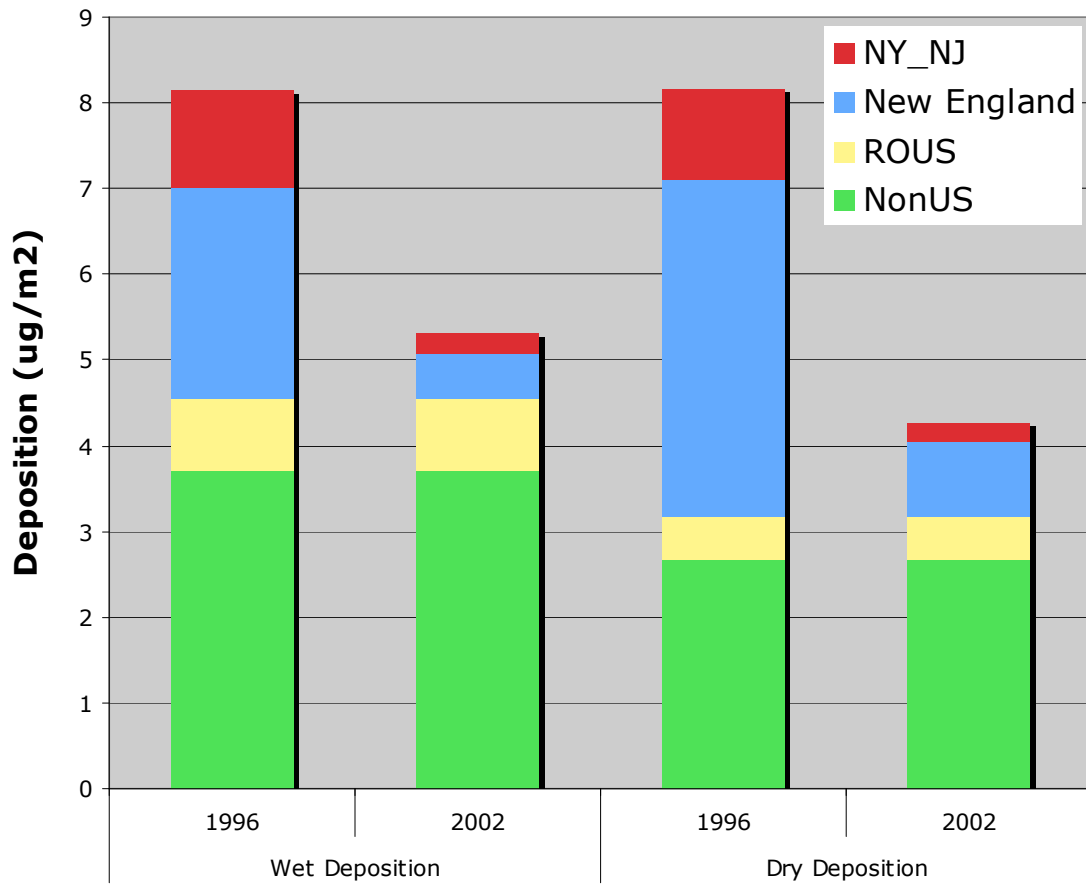


Figure 5-2 Relative contribution to total deposition in NESCAUM by region

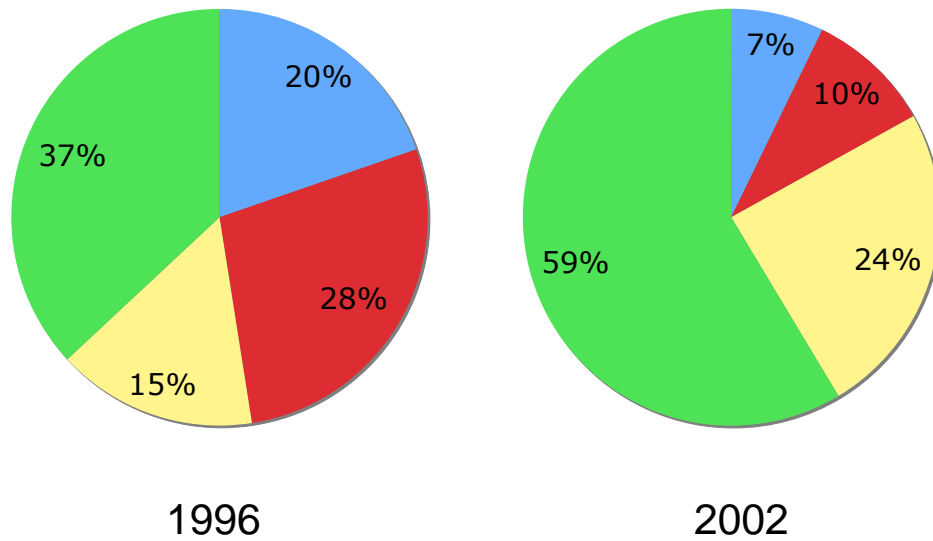
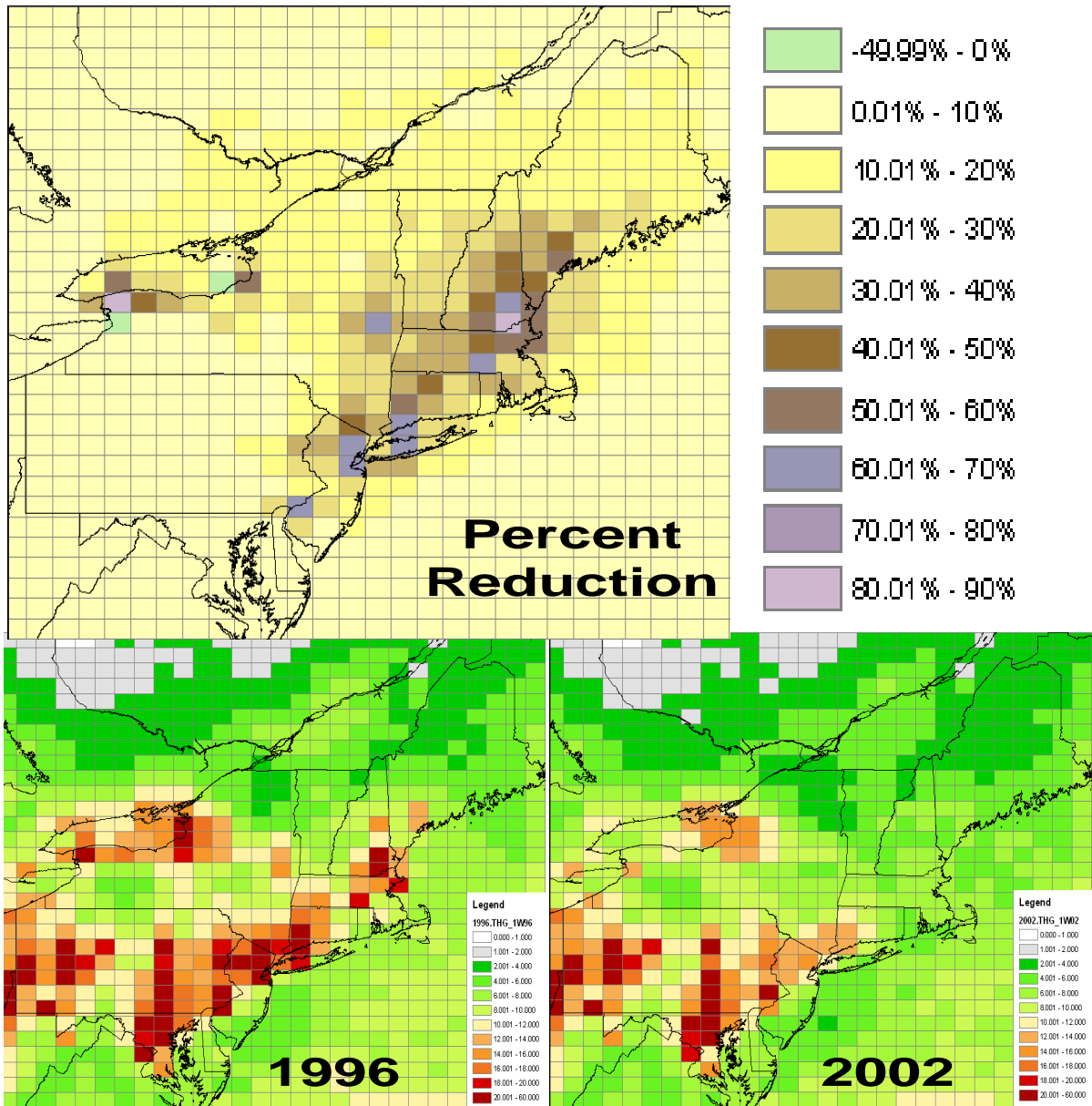


Figure 5-3 Wet deposition percent changes between 1996 and 2002 (top panel) based on modeled deposition in 1996 and 2002 (bottom panels)



Given the potential localized nature of mercury deposition, the geographic area chosen to summarize results can substantially affect the interpretation. Figure 5-3 displays wet deposition results across the Northeast for both model scenarios in the bottom panels. Substantial spatial gradients exist in the deposition fields. The location of modeled emission reductions clearly affects these gradients, with substantial grid-specific differences in deposition percent reduction. Table 5-1 illustrates the variation in modeled deposition, both in absolute deposition totals and relative contributions from source region. The first two columns are the data plotted to generate Figure 5-1 and Figure 5-2.

The next pair of columns provides the deposition source breakdown to two regional receptors (New York-New Jersey and New England). Total deposition to these two regions show large differences, with deposition averaged over New York and New Jersey 50 percent greater than average deposition modeled in New England for 2002. The differences in apportionment indicate the rest of US contributes more than twice the percentage of total deposition to the New York-New Jersey region than New England, and is nearly four times greater in absolute terms. The final three columns provide state-specific deposition allocations. Note that non-US sources dominate the modeled deposition in Maine, due in part to Maine’s location near the edge of the domain. This highlights one limitation of the tagging, as the mercury at the model boundary can reasonably be attributed to re-circulated US emissions out of and back into the domain, rather than arising from purely outside the domain. However, the exact proportion and allocation to region and source type can only be estimated by the contribution of these tags near the edge of the boundary. In other words, air masses that pass out of the model domain may get re-circulated back as part of the boundary, but no longer retain information on the origin of the mercury.

Table 5-1 Modeled mercury deposition averaged over different geographic regions

	1996	Total Mercury Deposition 2002						
	Deposition	to NESCAUM	to NESCAUM	to NY/NJ	to New England	to NY	to MA	to ME
Average (µg/m ²)								
In Region Sources	8.89	2.01	2.17	1.86	1.86	4.47	0.52	
Other US Sources	2.89	2.89	4.71	1.32	4.02	2.07	0.67	
Non-US Sources	6.94	6.94	7.62	6.35	7.50	7.40	5.85	
Total	18.72	11.83	14.50	9.53	13.38	13.94	7.04	
Percent Contribution								
In Region Sources	47%	17%	15%	20%	14%	32%	7%	
Other US Sources	15%	24%	32%	14%	30%	15%	10%	
Non-US Sources	37%	59%	53%	67%	56%	53%	83%	

5.1.3. Contribution by source type

The model scenarios relied on source-specific tags in addition to the regional tags to provide direct impacts of emission reductions from source classes. In addition to modeling reductions in MWCs, MWIs, and SSIs, other major source category tags, including EGU, area, and mobile sources, provide information on their relative contribution to overall deposition.

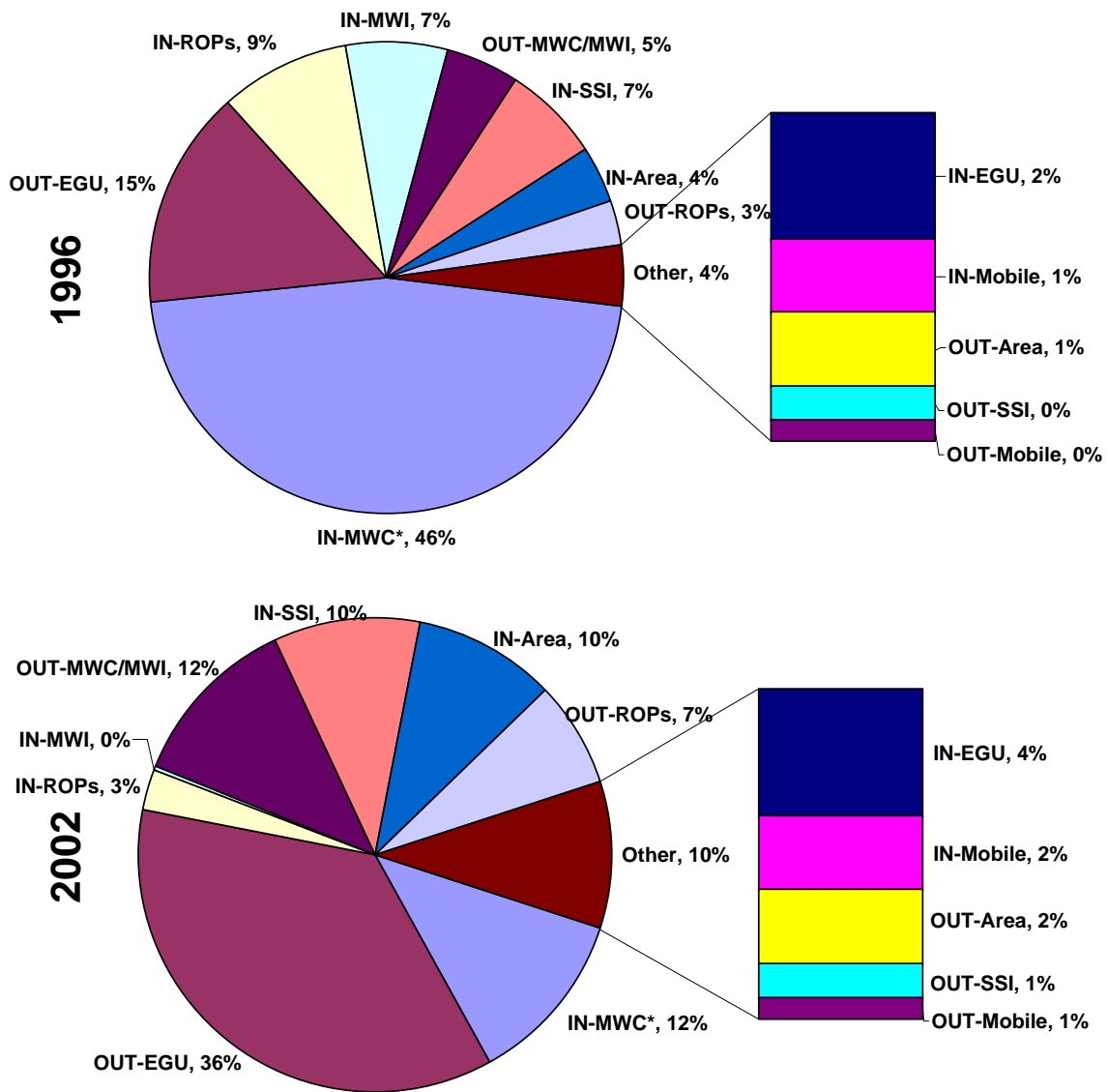
A summary of source-specific total deposition results for both model scenarios are shown in Table 5-2 and. Based on the table, in 1996 the predominant anthropogenic source of mercury deposition in the region came from MWC emissions (note that the emissions total modeled in 1996 include pre-1996 emissions for New Jersey MWCs). The substantial emission reductions modeled for this source category resulted in nearly a

90 percent decrease in associated mercury deposition. In 2002, the MWC contribution to regional deposition fell to 5 percent of the total from 29 percent in 1996. (Note that the higher percentages of Figure 5-4 are only for percentage source contributions from US sources and do not include the contribution from non-US sources). Other source categories showed substantial deposition reductions as well. For the “rest of point sources” category, chloralkali facilities that closed between 1996 and 2002 accounted for a large part of the decrease.

Table 5-2 Total average mercury deposition in NESCAUM states

		Wet+Dry Deposition ($\mu\text{g}/\text{m}^2$)		
		1996	2002	Percent Change
NESCAUM Sources	EGU	0.19	0.19	0%
	MWC*	5.49	0.59	-89%
	MWI	0.83	0.01	-99%
	SSI	0.78	0.49	-37%
	Rest of Points (ROPs)	1.02	0.14	-86%
	Area	0.48	0.48	0%
	Mobile	0.11	0.11	0%
NESCAUM Total		8.89	2.01	-77%
Rest of US Sources	EGU	1.77	1.77	0%
	MWC/MWI	0.59	0.59	0%
	SSI	0.05	0.05	0%
	Rest of Points (ROPs)	0.35	0.35	0%
	Area	0.11	0.11	0%
	Mobile	0.03	0.03	0%
Rest of the US Total		2.89	2.89	0%
Total Non-US Sources		6.94	6.94	0%
Grand Total		18.73	11.82	-37%

Figure 5-4 Relative contribution to total mercury deposition in NESCAUM region from sources in the US



The tagged information can also be plotted spatially to demonstrate the area of influence of the sources. Figure 5-5 shows the total, wet, and dry deposition contributions from MWCs and MWIs in the NESCAUM region for the 1996 (top) and 2002 (bottom) simulations. These plots clearly demonstrate the local impact of emissions from these facilities in 1996, and reveal the drastic reduction in mercury deposited in the region. In particular, note the high deposition levels in northeastern Massachusetts that virtually disappear as a direct result of local emission reductions. Modeled emissions from MWC and MWI sources outside the NESCAUM region have limited impact on deposition in New England, with the exception of New Jersey (Figure 5-6).

Figure 5-5 1996 and 2002 deposition results for MWC and MWI tags from NESCAUM sources

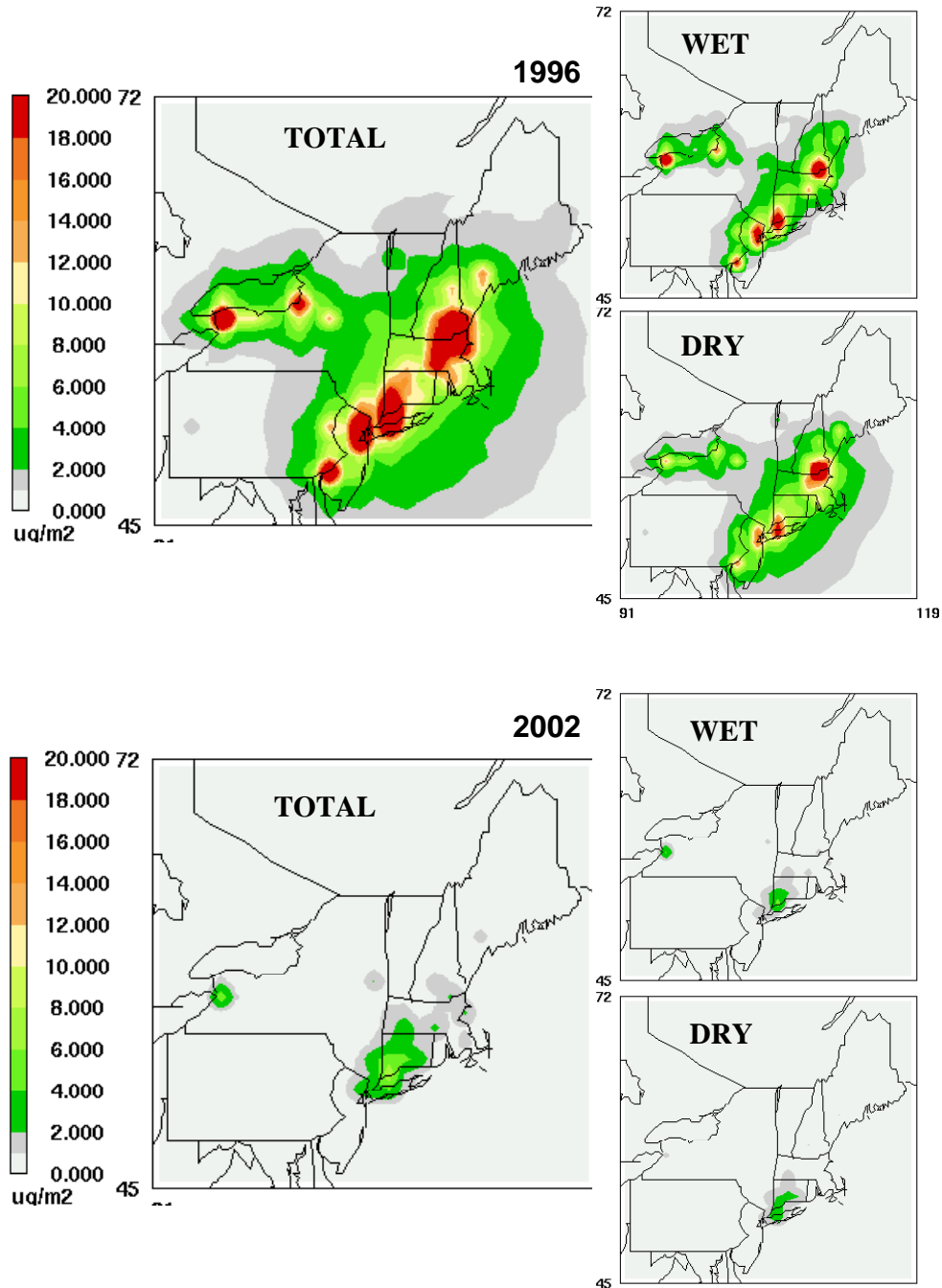
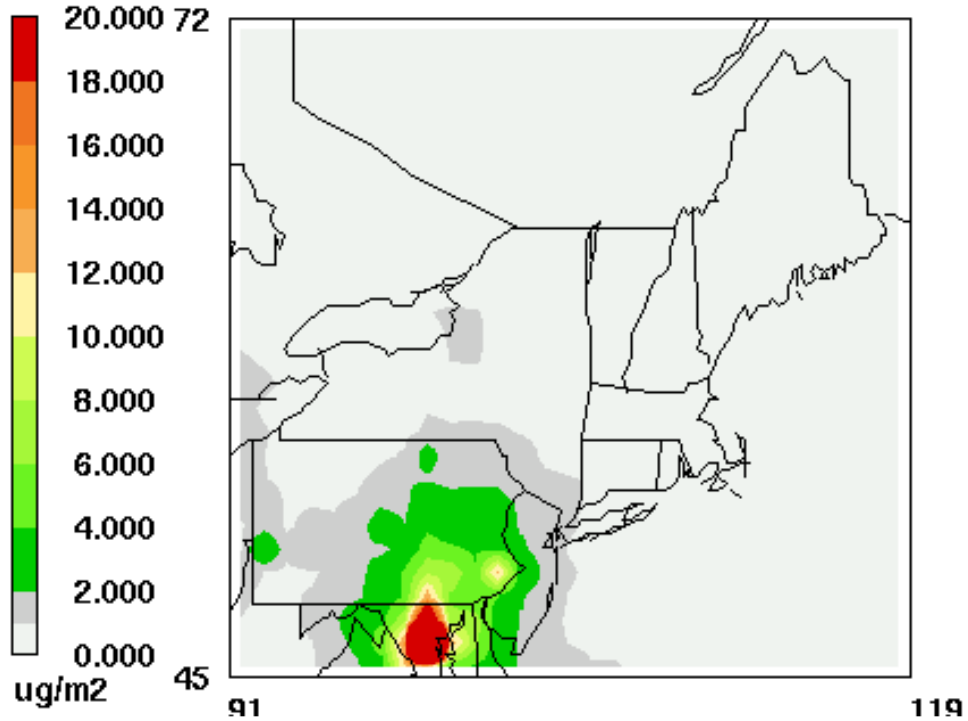


Figure 5-6 Total deposition from MWCs and MWIs outside of NESCAUM region



5.1.4. Comparison with RELMAP

In the late 1990s, modeling for the Northeast was conducted using the mercury version of the Regional Lagrangian Model of Air Pollution (RELMAP). Like the current study, the model study was designed to aid regional efforts in understanding the contributions of different source types and regions to deposition in the Northeast. The current REMSAD 1996 simulation results are compared with the earlier RELMAP results to demonstrate the consistency between the two modeling exercises.

Table 5-3 Comparison of RELMAP and REMSAD

	RELMAP	REMSAD
Type	Lagrangian (Puff)	Eulerian (Grid)
Background Conc.	Static (1.6 ng/m3)	None (use IC/BC)
Vertical Structure	4 (up to 1.5km above ground)	12 (up to 16km)
Emissions	1996 (Some from 1992)	1996/2002
Meteorology	1989	1996

Table 5-3 lists notable differences between the two modeling platforms. RELMAP is a Lagrangian or puff model, while REMSAD is an Eulerian or grid model. While RELMAP assumed a static background concentration that did not dry deposit, REMSAD relied on the use of initial and boundary conditions. The vertical structure in REMSAD included 12 layers extending 16 km into the atmosphere, while RELMAP was constrained to four layers with a top much closer to the surface. The meteorology driving

the models was from different years, which was shown previously to have a strong influence on wet deposition patterns. Finally, the emissions were not exactly the same for the simulations, and differences are detailed in Figure 5-4. Total emissions modeled with RELMAP were about 20 percent greater than in the 1996 REMSAD simulation, although only a 10 percent difference when considering emissions from the Northeast.

Table 5-4 Emissions modeled in three Northeast scenarios

Emission /Region	1996 RELMAP(TPY)	1996 REMSAD(TPY)	2002 REMSAD (TPY)	% difference (REMSAD96 /RELMAP)
Northeast	22.0	19.7	7.0	-10.6
ROUS	160.1	124.3	124.3	-22.3
Total	182.1	144.0	131.3	-20.9

Figure 5-7 plots wet deposition attributed to EGUs from outside the Northeast for both RELMAP and REMSAD. A strong resemblance in the magnitude and spatial patterns of the two model results is observed. Similarly, the absolute wet deposition magnitude and spatial patterns are consistent for deposition attributed to MWCs in the northeast region (Figure 5-8). This correspondence in part results from the fact that the point source locations are the same for both scenarios. Although the absolute emission totals and even speciation of those emissions may not be the same, they are sufficiently similar, as is the underlying predominance of Hg²⁺ deposition. Presumably, the deposition fields of both models are dominated by direct emission of Hg²⁺ and its propensity to deposit near its source.

Table 5-5 compares apportionment results from the two models, showing three receptor regions (Massachusetts, New England, and the NESCAUM states). The RELMAP results were reanalyzed and summarized to correspond more closely to the REMSAD spatial definitions (since the original 1998 report was summarized for a broader northeast region). The total deposition and its breakout into NESCAUM, Rest of the US, and ‘natural deposition’ are provided, where the last category includes any non-US anthropogenic source. The REMSAD results predict a somewhat higher contribution from this ‘global pool,’ which is consistent with RELMAP not modeling dry deposition from its background source. In addition, since REMSAD modeled lower direct emissions, lower contributions from US sources should be expected. The combined effect of REMSAD deposition from the boundary conditions and lower US anthropogenic emissions yields a greater relative contribution of the global pool in the REMSAD modeling relative to the previous RELMAP results.

Table 5-5 Comparison of RELMAP and REMSAD apportionments

	Massachusetts				New England				NESCAUM			
	Total	NESC	ROUS	natdep	Total	NESC	ROUS	natdep	Total	NESC	ROUS	natdep
ug/m2	30.4	22.6	2.5	5.3	19.2	11.8	2.9	4.4	22.4	12.1	5.5	4.8
RELMAP	100%	74.4%	8.3%	17.3%	100%	61.6%	15.3%	23.1%	100%	54.2%	24.4%	21.4%
ug/m2	27.1	17.7	2.1	7.3	16.1	8.4	1.3	6.3	18.6	8.9	2.9	6.9
REMSAD	100%	65.3%	7.6%	27.1%	100%	52.5%	8.1%	39.4%	100%	47.5%	15.3%	37.1%

Figure 5-7 Comparison of RELMAP and REMSAD wet deposition from EGUs outside the NESCAUM region for 1996

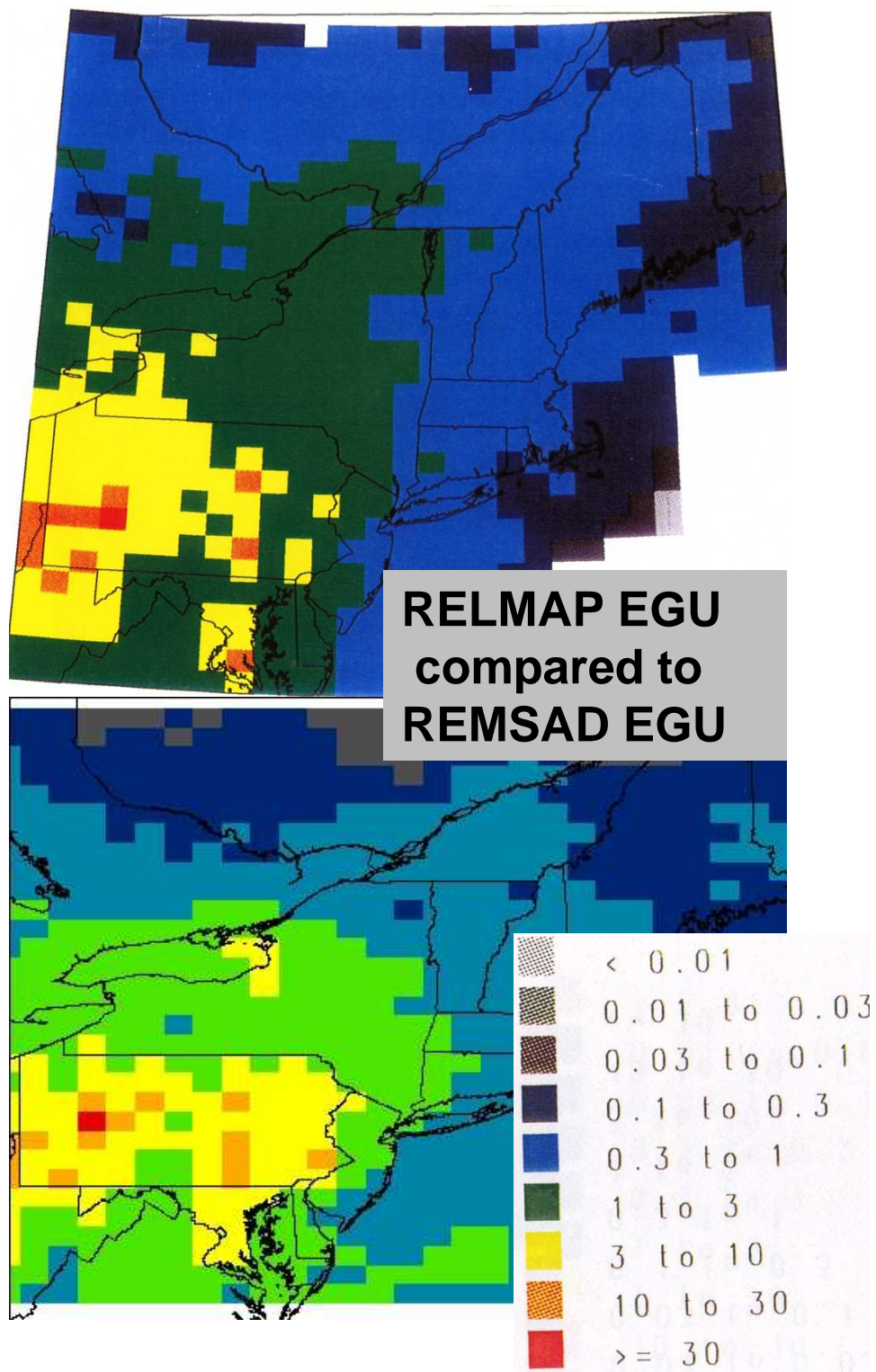
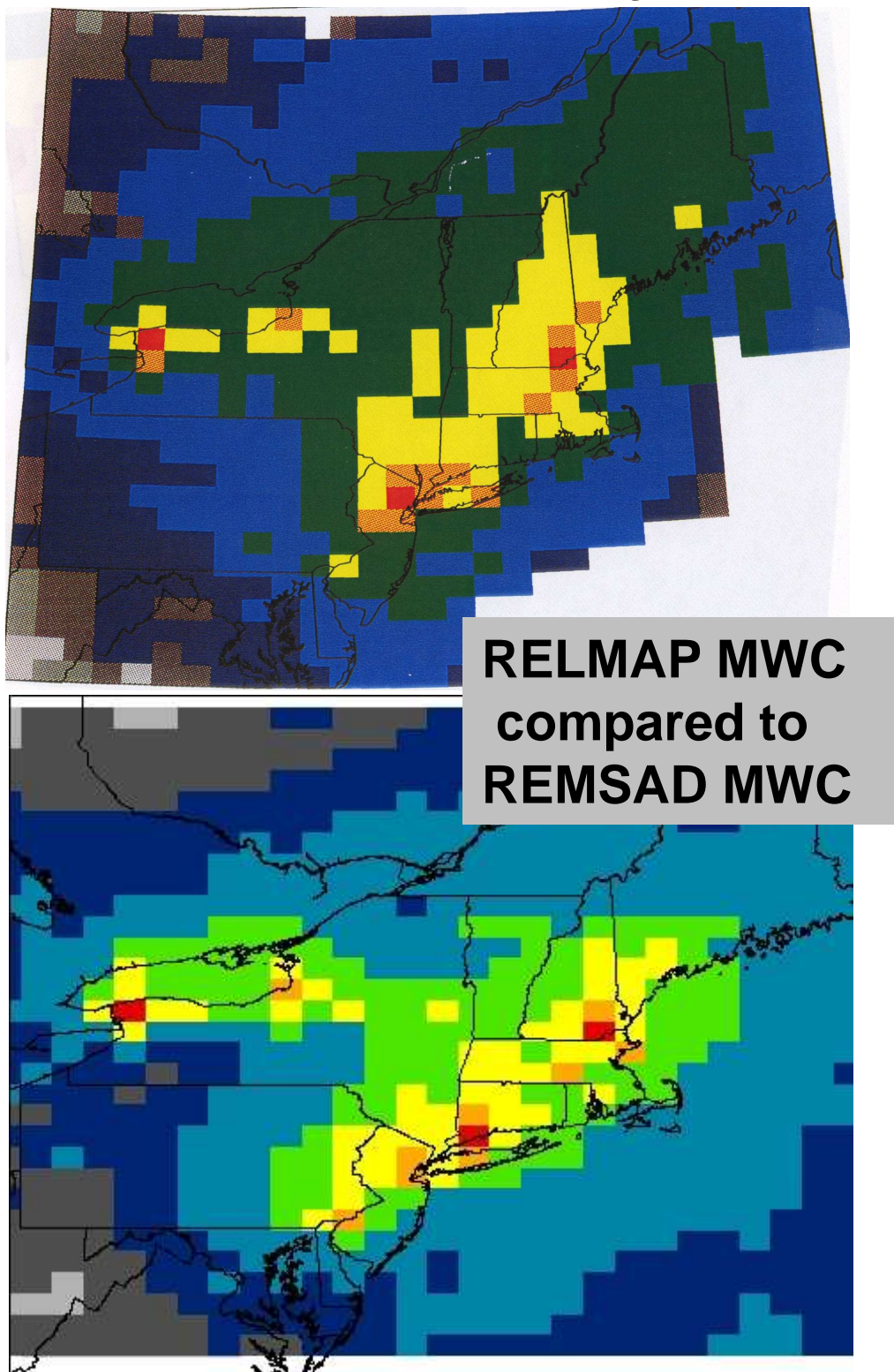


Figure 5-8 Comparison of RELMAP and REMSAD wet deposition from MWC emissions within the NESCAUM region for 1996



5.1.5. Comparison with other modeling and monitoring results

New York State Global Modeling of Mercury Deposition

A number of studies have presented mercury deposition modeling results relevant to the Northeast. One study reports the global and regional contributions to mercury deposition in New York State (Seigneur et al., 2003). Using a global model with a nested continental scale model, Seigneur et al. conducted a sensitivity study of deposition to three sites in New York, assuming different transport tendencies of mercury (e.g., difference in transport scale or area of deposition influence).

Results of this modeling exercise are summarized broadly and compared with the results of the REMSAD 1996 and 2002 modeling (Table 5-6). The upper part of the table gives the ranges of dry and wet deposition modeled for this study as compared to that reported at the three New York receptors by Seigneur et al. The deposition results shown in the table are very consistent between the two studies.

The lower part of Table 5-6 provides the source apportionment results from the modeling studies. Note that NESCAUM's modeling results for 2002/1996 uses a tagging scheme that combines emissions from New Jersey and New York. The results for (1) the Adirondacks, (2) Finger Lakes and (3) the Catskills are given, as reported in tables 5, 6 and 7, respectively, in Seigneur et al. (2003). Again, the results agree that New York mercury sources account for a non-trivial amount of the deposition within the State, along with a substantial contribution from the rest of the US and global sources.

Table 5-6 Comparison of REMSAD results and Seigneur et al. (2003) attribution of mercury deposition to New York State

	NESCAUM Modeling	AER-Seigneur
Dry Deposition (ug/m ³)	5.5-8.8	3.9-10.2
Wet Deposition (ug/m ³)	7.9-10.5	9.3-12.7
Total Deposition (ug/m ³)	13.4-19.3	13.2-22.9

Source	NESCAUM	Seigneur (1)	Seigneur (2)	Seigneur (3)
New York (NJ)	12/37	10-14	19-25	9-13
Rest of US	32/24	25-28	30-31	47-50
Global / Natural	56/39	28-65	45-50	37-44

Massachusetts Mercury Monitoring and Source Apportionment

Massachusetts Department of Environmental Protection (MA DEP) has supported the collection and analysis of wet deposition at several sites across the State, including North Andover and Quabbin Reservoir. Unlike the MDN week-long integrated samples, the Massachusetts work relies on event-based collection, collection wet deposition for much shorter periods (i.e. a discrete rain event). The samples were analyzed for mercury

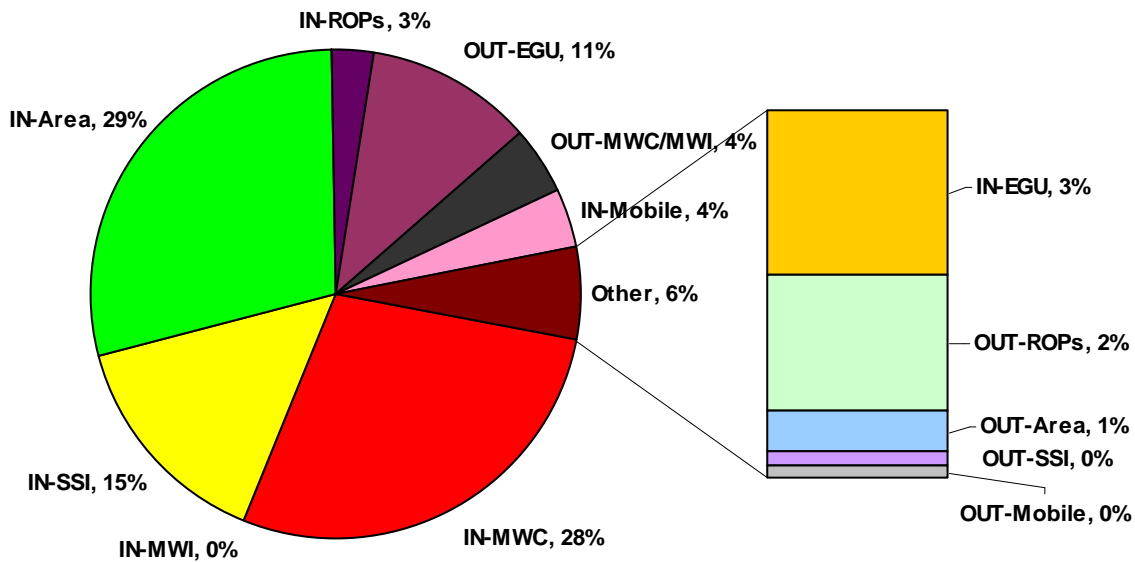
and trace elements (by ICP-MS) by Keeler at University of Michigan. Mercury results are tabulated (Table 5-7) and compared to modeled results from this study. The concentrations observed in North Andover were ~20% higher than at Quabbin, which is comparable to the ~15% difference from the model results. The model does predict higher deposition than the measurements, although this could be due to the use of 1996 meteorology and its associated wetter than average character.

Keeler conducted PMF source apportionment modeling using the elemental data obtained from the deposition measurement (Keeler 2004). The levels of trace elements suggested that more than one incinerator was influencing mercury deposition in North Andover. Based on the analysis, two different waste incinerator signatures were associated with 37 and 24 percent of the monitored deposition at North Andover. Fossil fuel combustion contributed another 24 percent to the mercury deposition at that site. Figure 5-9 shows the apportionment determined by the 2002 model results at North Andover which shows that 47 percent of the emissions are from incinerators (e.g. MWC, MWI, SSI) and 23 percent from fossil fuel sources (e.g. EGU, ROP, Mobile).

Table 5-7 Comparison of event based measurements to modeled wet deposition

Wet Hg Deposition ($\mu\text{g}/\text{m}^2$)	1996 modeled	2002 modeled	7/02-6/03 measured
Quabbin	22.2	12.7	8.8
North Andover	134.9	14.5	10.7

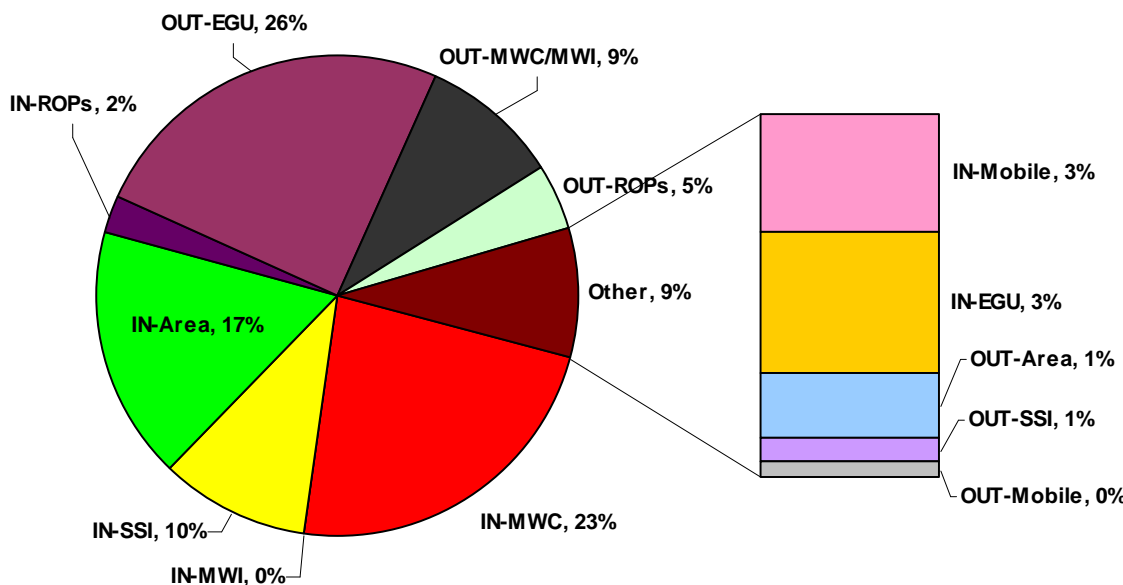
Figure 5-9 Relative Contribution to Total Modeled Deposition in North Andover, MA from Sources in the US



The corresponding analysis for the Quabbin site showed a lower relative contribution from incinerators, totaling 49 percent and relatively greater 41 percent from fossil fuels, as compared to the North Andover site. These results seem reasonable given the nearby Mt. Tom facility to Quabbin. The modeled deposition at this site also predicted a greater fossil fuel contribution of 39 percent, with 43 percent contribution from incineration sources (Figure 5-10).

Results from both the grid model and receptor model show that fossil fuel combustion and incineration contribute substantially to mercury deposition in Massachusetts. It should be noted, however, that the model results used for the comparison did not include the deposition attributed to the boundary conditions, as these cannot be associated with specific source types. Likewise, the receptor model fails to attribute any of the deposition to global sources, per se. PMF attempts to apportion all of the input to sources based on the variability within the data. Some portion of the mercury mass associated with each of the PMF identified sources is likely derived from background environmental levels. The consistency of the results derived from two very disparate modeling approaches

Figure 5-10 Relative Contribution to Total Modeled Deposition at Quabbin from Sources in the US



Massachusetts: Fish Tissue Sampling

A recent study in Massachusetts observed significant declines in mercury appearing in fish in Massachusetts lakes that coincided with the steep decline in mercury air emissions from incinerators. MA DEP has established a network of lakes for the long-term monitoring of mercury concentrations in the edible tissue of two fish species. This network has provided the state with an indication of changes in fish tissue mercury concentrations over six years from 1999 to 2004 in seventeen lakes (MA DEP, 2006).

Over this period, statistically significant decreases in mercury concentrations in the sampled fish in a number of the network lakes located in the northeastern part of the state. This decline of mercury in fish tissue coincided with a significant decrease, ~87 percent, in mercury air emissions in the same local region due to new pollution controls on and closures of municipal and medical waste incinerators. This part of the state was known as a “hot spot” of mercury air emissions due to the relatively high number of incinerators in the area. The fish tissue samples in northeast Massachusetts were also higher than the state on average, indicating it was a mercury deposition hotspot as well as an emissions hotspot.

The mercury trends in the fish tissue samples from northeast Massachusetts are consistent with mercury deposition modeling presented here, as well as with modeling work by Evers et al. (2007) for this location. The REMSAD 1996 modeling indicates an area of high total mercury deposition in northeast Massachusetts that corresponds to a high density of municipal and medical waste incinerators in the same locale. The mercury emissions in this area greatly decreased since 1996 due to more stringent mercury emission controls on incinerators, mercury reductions in waste streams, and incinerator shutdowns. The REMSAD results for 2002 (after the decrease in local mercury emissions) show a corresponding decrease in mercury deposition from these sources, consistent with the findings from the MA DEP fish tissue sampling study and Evers et al. (2007).

6. SUMMARY

Since 1998, the Northeast states and eastern Canadian provinces have worked towards goal of virtually eliminating all in-region anthropogenic sources of mercury releases to the environment. A near-term step was to cut mercury releases by 50 percent by 2003, with an intermediate goal of 75 percent by 2010. To assist this effort, NESCAUM and the New England Regional Office of the United States Environmental Protection Agency (US EPA) undertook the task of creating a 2002 progress report for the region.

The work described in this report had two objectives: 1) to determine mercury deposition in the NESCAUM region and apportion the contribution to deposition according to source region and major source category, and 2) to provide input (i.e., loading) values to aquatic and ecological models that can inform regulatory and policy decisions. This report documents the mercury modeling platform employed, with a description of the model and inputs generated for this work. It compares modeled deposition and atmospheric mercury concentrations where possible to measurements, and discusses differences between the two modeled years of 1996 and 2002.

This effort used the Regional Modeling System for Aerosols and Deposition (REMSAD version 7.13), a three-dimensional Eulerian grid model. The model includes wet and dry deposition and re-emission of mercury in three forms – reactive gaseous mercury (RGM), gaseous elemental mercury, and particulate mercury. REMSAD employs an attractive feature termed “emission source tagging.” The model permits users to track emissions from a specific source, source category, source region, or combination of these by assigning a “tag” to the emissions. The tagging scheme is an accounting system that follows species through space and time in the model without disturbing the physical or chemical processes affecting that species. With careful consideration, the user can establish a model run to assess the impact and influence of specific modeled sources, including boundary conditions. For mercury, REMSAD can follow the three model species (reactive gaseous mercury, gaseous elemental mercury, and particulate mercury) with their own tags.

Model results were evaluated against a limited set of measurements for validation purposes. This comparison revealed reasonable model performance and suggests that existing mercury wet deposition monitors are not in locations most affected by mercury emission reductions in the Northeast.

Variability in ambient data indicates the strong influence of year-to-year meteorological changes, which may mask deposition decreases attributable to emission declines. The REMSAD modeling only used meteorological data from 1996—the wettest year on record in a few states—to represent all years. The amount of rainfall has a significant effect on the wet deposition of mercury and how far it travels from the source; often the model gives higher wet deposition levels than were monitored. A lack of

monitoring data prevents a comprehensive comparison of model results with dry deposition measurements.

Published measurements of ambient elemental mercury indicate that the model estimates at the surface generally tend to be 15-50 percent lower than annual-averaged ambient measurements collected in New York, Canada, and around the Great Lakes. Given the current high level of uncertainty in both measured and modeled values, assessing the importance of these differences is difficult.

The model captures the monthly trends in total mercury deposition reasonably well, showing minima in late summer and fall with maximum levels in the springtime. The magnitude of the month-to-month changes is greater in the modeled results than in the monitored data.

Substantial decreases in both wet and dry deposition are modeled as a direct result of reduced mercury emissions in the NESCAUM states. Mercury emission reductions totaling over 60 percent occurred from point sources in the Northeast, including Municipal Waste Combustors (MWCs), Medical Waste Incinerators (MWIs), and Sewage Sludge Incinerators (SSIs). Total deposition averaged across the NESCAUM states declined by 37 percent as a direct result. Smaller geographic regions near sources showed even greater deposition declines (over 80 percent reduction) from local emission changes. The analysis demonstrates that substantial local benefits can be achieved from local and regional scale emission reductions.

The deposition results also indicate the growing importance of emissions outside the Northeast relative to the declining emissions within the Northeast. The model estimated the share of total mercury deposition from sources in the NESCAUM region decreased from 47 percent in 1996 to 17 percent in 2002, while the share from US sources outside the NESCAUM region increased from 15 percent to 24 percent, and non-US sources increased from 37 percent to 59 percent. The REMSAD modeling, however, cannot distinguish between US and non-US mercury in air masses that pass out of the model domain but may get re-circulated back in, thus losing tagged model information at the domain boundary on the origin of the mercury. For example, “non-US sources” dominate the modeled deposition in Maine, due in part to Maine’s location near the boundary of the modeling domain. However, the re-circulation of mercury out of and back into the domain would lose track of US mercury sources that may contribute to deposition at this near-boundary location.

In 1996, the predominant anthropogenic source of mercury deposition in the NESCAUM region came from MWC emissions. The REMSAD model estimated almost a 90 percent decrease in associated mercury deposition from MWCs since 1996 as a result of the substantial emission reductions from this source category. In 2002, the modeled MWC contribution to regional mercury deposition fell to 5 percent of the total from 29 percent in 1996. Other source categories showed substantial deposition reductions as well. For example, closures of chloralkali facilities between 1996 and 2002

accounted for a large part of the decrease in mercury emissions from the “rest of point sources” category.

The 1996 REMSAD modeling results were consistent with earlier modeling of the Northeast using the mercury version of the Regional Lagrangian Model of Air Pollution (RELMAP). While the mercury emissions inventories differed by about 20 percent (with RELMAP being greater), the magnitude and spatial patterns of modeled mercury wet deposition displayed a strong resemblance between the two models. Because the point source locations are the same in both models, the correspondence in the deposition patterns suggests that the direct emission of Hg^{2+} with its propensity to deposit near its source dominates the deposition fields of both models. The REMSAD results predict a somewhat higher contribution from the non-US “global pool” background source than RELMAP, which is consistent with RELMAP not modeling dry deposition from its background source. The combined effect of dry deposition of the global pool and decreased anthropogenic emission levels modeled with REMSAD yields a relatively greater contribution of the global pool to the REMSAD results relative those from RELMAP.

The REMSAD results presented in this report can also be compared to other modeling and monitoring results in the Northeast in addition to the RELMAP study. In a sensitivity modeling study of deposition to three sites in New York State, Seigneur et al. (2003) found, in agreement with the REMSAD results, that New York mercury sources account for a non-trivial amount of the deposition within the State, in addition to a substantial contribution from the rest of the US and global sources. Sampling of mercury in fish tissue between 1999 and 2004 by MA DEP found a spatial correspondence between an area of high mercury emissions and high fish tissue mercury samples in northeastern Massachusetts. The level of mercury in the sampled fish tissue decreased over the sampled years at the same time mercury emissions from local sources decreased significantly. The 1996 REMSAD modeling indicated this area of northeastern Massachusetts was a local “hot spot” for mercury emissions and deposition, which was largely eliminated by 2002 due to the reduction in mercury emissions from the local sources. A study by Evers et al. (2007) also found the same “hot spot” in this portion of northeastern Massachusetts.

The deposition results from this modeling effort show reasonable agreement with previous deposition modeling, ambient measurements and receptor modeling. In addition, the conclusion that local emission reduction efforts can have a profound effect on local deposition is supported by this modeling and corroborated by fish tissue measurements in Massachusetts. The added benefit of source tagging from this modeling should prove helpful as the northeast states move forward toward their goal of virtual elimination of mercury emissions in the region.

7. REFERENCES

- Cohen, M., Artz, R., Draxler, R., Miller, P., Poissant, L., Niemi, D., Ratte, D., Deslauriers, M., Duval, R., Laurin, R., Slotnick, J., Nettesheim, T., McDonald, J., 2004. Modeling the atmospheric transport and deposition of mercury to the Great Lakes. *Environ. Res.* 95, 247-265.
- Conference of New England Governors–Eastern Canadian Premiers (1998). New England Governors and Eastern Canadian Premiers Mercury Action Plan. June 1998 (available on-line at <http://www.state.ma.us/dep/ors/files/negecp.pdf>).
- Conference of New England Governors–Eastern Canadian Premiers (2001). Resolution 26–3, adopted at the 26th annual conference, Westbrook, CT (available on-line at http://www.negc.org/documents/NEG-ECP_Overview07_03.pdf, see pp. 47–48).
- Evers, D.C., Han, Y.-J., Driscoll, C.T., Kamman, N.C., Goodale, M.W., Lambert, K.F., Holsen, T.M., Chen, C.Y., Clair, T.A., Butler, T., 2007. Biological mercury hotspots in the northeastern United States and southeastern Canada. *BioScience* 57, 29-43.
- Gery, M.W., Whitten, G.Z., Killus, J.P., Dodge, M.C., 1989. A photochemical kinetics mechanism for urban and regional computer modeling. *J. Geophys. Res.* 94, 12,925-12,956.
- Griffin, R.J., Cocker III, D.R., Flagan, R.C., Seinfeld, J.H., 1999. Organic aerosol formation from the oxidation of biogenic hydrocarbons. *J. Geophys. Res.* 104, 3555-3567.
- Hales, J.M., Sutter, S.L., 1973. Solubility of sulfur dioxide in water at low concentrations. *Atmos. Environ.* 7, 997-1001.
- Han, Y.-J., Holsen, T.M., Lai, S.-O., Hopke, P.K., Yi, S.-M., Liu, W., Pagano, J., Falanga, L., Milligan, M., Andolina, C. 2004. Atmospheric Gaseous Mercury Concentrations in New York State: Relationships with Meteorological Data and Other Pollutants. *Atmos. Environ.* 38, 6431-6446.
- Kamman, N.C. 1998. Mercury Inputs, Transport and Cycling in Northern Lakes. Vermont Department of Environmental Conservation. 1-35.
- Keeler, G.J., 2004. Mercury Deposition Monitoring and Source Apportionment in Massachusetts.
- Kim, Y.P., Seinfeld, J.H., Saxena, P., 1993. Atmospheric gas-aerosol equilibrium I. thermodynamic model. *Aerosol Sci. Technol.* 19, 157-181.
- Kim, K.-H., Ebinghaus, R., Schroeder, W. H., Blanchard, P., Kock, H.H., Steffen, A., Froude, F.A., Kim, M.-Y., Hong, S., Kim, J.H. 2004. Atmospheric Mercury Concentrations from Several Observatory Sites in the Northern Hemisphere. *J. Atmos. Chem.* 50, 1–24.

- Lin, C.A., Pehkonen, S.O., 1999. The chemistry of atmospheric mercury: a review. *Atmos. Evt.* 33, 2067-2079.
- Lin, C.-J., Lindberg, S.E., Ho, T.C., Jang, C. 2005. Development of a Processor in BEIS3 for Estimating Vegetative Mercury Emission in the Continental United States. *Atmos. Evt.* 39, 7529-7540.
- Lindberg, S., Brooks, S., Lin, C., Scott, K., Landis, M., Stevens, R., Goodsite, M., Richter, A., 2002. Dynamic oxidation of gaseous mercury in the Arctic troposphere at polar sunrise. *Environ. Sci. Technol.* 36, 1245-1256.
- Lyman, S.N., Gustin, M.S., Prestbo, E.M., Marsik, F.J. 2007. Estimation of Dry Deposition of Atmospheric Mercury in Nevada by Direct and Indirect Methods. *Environ. Sci. Technol.* 41, 1970-1976.
- MA DEP (Massachusetts Department of Environmental Protection), 2006. Massachusetts fish tissue mercury studies: Long-term monitoring results, 1999-2004. MA DEP Office of Research and Standards, Boston, MA and Wall Experiment Station, Lawrence, MA.
- Marsik, F.J., Keeler, G.J., Landis, M.S. 2007. The dry-deposition of speciated mercury to the Florida Everglades: Measurements and modeling. *Atmos. Evt.* 41, 136-149.
- National Climate Data Center. Accessed January 2005.
<http://www.ncdc.noaa.gov/oa/climate/research/cag3/cag3.html>
- NESCAUM, NEWMOA, NEIWPC, EMAN, 1998. Northeast States and Eastern Canadian Provinces Mercury Study: A Framework for Action.
- NESCAUM, 2005. [Inventory of Anthropogenic Mercury Emissions in the Northeast](#), NESCAUM, Boston, MA (available on-line at:
<http://www.nescaum.org/documents/inventory-of-anthropogenic-mercury-emissions-in-the-northeast>).
- Odum, J.R., Jungkamp, T.P.W., Griffin, R.J., Flagan, R.C., Seinfeld, J.H., 1997. The atmospheric aerosol-forming potential of whole gasoline vapor. *Science*, 276, 96-99; *see also* Odum, J.R., Jungkamp, T.P.W., Griffin, R.J., Forstner, H.J.L., Flagan, R.C., Seinfeld, J.H., 1997. Aromatics, reformulated gasoline, and atmospheric organic aerosol formation. *Environ. Sci. Technol.* 31, 1890-1897.
- Pacyna, J.M., Pacyna, E.G., Steenhuisen, F., Wilson, S., 2003. Mapping 1995 global anthropogenic emissions of mercury. *Atmos. Evt.* 37(S1), S109-S117.
- Pacyna, J.M., Pacyna, E.G., Steenhuisen, F., Wilson, S., 2006 (accepted). Global anthropogenic mercury emission inventory for 2000. *Atmos. Evt.*
- Pankow, J.F., 1994. An absorption model of the gas/aerosol partitioning involved in the formation of secondary organic aerosol. *Atmos. Evt.* 28, 185-193.

- Poissant, L., Pilote, M., Beauvais, C., Constant, P., Zhang, H.H., 2005. A year of continuous measurements of three atmospheric mercury species (GEM, RGM and Hg_p) in southern Québec, Canada. *Atmos. Eno. 39*, 1275-1287.
- SAI, 2002. User's Guide to the Regional Modeling System for Aerosols and Deposition (REMSAD), Version 7, ICF Consulting/SAI, San Francisco, CA.
- Saxena, P., Hudischewskyj, A.B., Seigneur, C., Seinfeld, J., 1986. A comparative study of equilibrium approaches to the chemical characterization of secondary aerosols. *Atmos. Eno. 20*, 1471-1483.
- Scott, B.C., 1978. Parameterization of sulfate removal by precipitation. *J. Appl. Meteor. 17*, 1375-1389.
- Seigneur, C., Lohman, K., Vijayaraghavan, K., Shia, R.-L., 2003. Contributions of global and regional sources to mercury deposition in New York State. *Environmental Pollution 123*, 365-373.
- Selin, N.E., Jacob, D.J., Park, R.J., Yantosca, R.M., Strode, S., Jaegle, L., Jaffe, D., 2007. Chemical cycling and deposition of atmospheric mercury: Global constraints from observations. *J. Geophys. Res. 112*, D02308, doi:10.1029/2006JD007450.
- Sigler, J.M., Lee, X. 2006. Recent Trends in Anthropogenic Mercury Emission in the Northeast United States. *J. Geophys. Res. 111*, D14316, doi:10.1029/2005JD006814.
- Smolarkiewicz, P.K., 1983. A simple positive definite advection scheme with small implicit diffusion, *Mon. Wea. Rev. 111*, 479-486.
- Sprovieri, F., Pirrone, N., Landis, M.S., Stevens, R.K., 2005. Oxidation of Gaseous Elemental Mercury to Gaseous Divalent Mercury during 2003 Polar Sunrise at Ny-Alesund. *Environ. Sci. Technol. 39*, 9156-9165.
- UNEP, Chemicals, 2002. Global Mercury Assessment, 2002. Geneva, Switzerland: Inter-Organization Programme for the Sound Management of Chemicals (IMOC).
- US EPA, October 2003. Technical Support Document for the Clear Skies Act 2003 Air Quality Modeling Analysis (available on-line at: http://www.epa.gov/air/clearskies/air_quality_tech.html).
- US EPA, 2004. Hg Speciation Excel File – correction. US EPA (available on-line at http://www.epa.gov/ttn/atw/utility/hgspec_fuelupdate.xls).
- US EPA, 2005. Emissions inventory and emissions processing for the Clean Air Mercury Rule (CAMR). U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Emissions Monitoring and Analysis Division, Research Triangle Park, NC (available on-line at http://www.epa.gov/ttn/atw/utility/emiss_inv_oar-2002-0056-6129.pdf).

- VanArsdale, A., Weiss, J., Keeler, G., Miller, E., Boulet, G., Brulotte, R., Poissant, L., 2005. Patterns of mercury deposition and concentration in northeastern North America (1996-2002). *Ecotoxicology* 14, 37-52.
- Wesley, M.L., 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. *Atmos. Environ.* 23, 1293-1304.
- Wiener, J.G., Knights, B.C., Sandheinrich, M.B., Jeremiason, J.D., Brigham, M.E., Engstrom, D.R., Woodruff, L.G., Cannon, W.F., Balogh, S.J. 2006. Mercury in Soils, Lakes, and Fish in Voyageurs National Park (Minnesota): Importance of Atmospheric Deposition and Ecosystem Factors. *Environ. Sci. Technol.* 40, 6261-6268.