Chapter 16
Importance of a Global Scale Approach to using Regional Models in the Assessment of Source-Receptor Relationships for Mercury

O. Russell Bullock Jr. and Lyatt Jaeglé

Summary Regional atmospheric models simulate their pertinent processes over a limited portion of the global atmosphere. This portion of the atmosphere can be a large fraction, as in the case of continental-scale modeling, or a small fraction, as in the case of urban-scale modeling. Regional modeling of any air pollutant requires that the meteorological and chemical conditions at the boundaries of the model domain be taken into account, especially if the pollutants are long-lived. It was once a common practice for the boundary concentrations of mercury and its reactants to be specified using time-constant values based on limited historical observation. These values were often invariant in the horizontal and vertical dimensions too. This relatively simple procedure for establishing boundary concentrations could be justified based on the previous notion that mercury was a rather inert and long-lasting air pollutant. However, with the subsequent discovery of rapid physical and chemical transformations of atmospheric mercury and significant concentrations of oxidized mercury far removed from known emission sources, the presumption of mercury as an inert substance has generally disappeared. The effect of intercontinental transport is now treated with greater concern in regional atmospheric mercury modeling. Global mercury models are now commonly used to define boundary values for regional mercury modeling. However, the global and regional models must use consistent information for emissions, surface physiology and meteorology to achieve consistent simulation results and associated source-receptor relationships. There is certainly a need for international cooperation on field research and numerical model development to supply the tools needed for confident assessment of source-receptor relationships for mercury on both global and regional scales.

16.1 Introduction

Regional models are typically used for atmospheric modeling when fine spatial detail is desired over a specific area of interest and there is insufficient computing power available to calculate the model over the entire global extent of the atmosphere.

Regional models are also used when there is a lack of necessary information outside of the region of interest. Regardless of motivation, regional models have been used to identify and assess the sources responsible for a variety of atmospheric contaminants including mercury. However, regional models typically require a specification of conditions at the lateral boundaries of their model domains. When these lateral boundary conditions are found to be important to the simulation of conditions within the model domain, a compromise must be made between model fidelity and model practicality. In order to make the simulations more realistic and credible, the model domain must be expanded to the largest extent possible while still allowing the necessary model calculations to be perfomed.

Some examples of regional models that have been used to simulate atmospheric Hg are the Acid Deposition and Oxidant Model (ADOM) (Petersen et al., 2001), the Community Multiscale Air Quality model (CMAQ) (Bullock and Brehme, 2002), the Danish Eulerian Hemispheric Model (DEHM) (Christensen, 1997), the Eulerian Model for Air Pollution (EMAP) (Syrakov, 1995; Ilyin et al., 2002), the Regional Modeling System for Aerosols and Deposition (REMSAD) (ICF, 2005), the Trace Element Analysis Model (TEAM) (Pai et al., 1997) and the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Cohen et al., 2004). However, the HYSPLIT model uses a Lagrangian modeling framework to track pollutant emissions from within its modeling domain and does not treat boundary fluxes of mercury. The following discussion deals only with models using Eulerian frameworks where boundary fluxes are considered in the treatment of global transport.

Mercury (Hg) is known to be a global pollutant due to the long atmospheric lifetime of gaseous elemental Hg (GEM). As a global pollutant, trans-boundary flows of Hg into regional modeling domains are an obvious concern. The mean atmospheric lifetime of GEM is believed to be on the order of months or longer. This long lifetime implies slow deposition where only a small fraction of the GEM transported into a regional model domain is likely to deposit before it is eventually transported out. For this reason, some early modeling assessments of source attribution and trans-national-boundary flux were conducted using rather simple estimates of the boundary concentrations of GEM based on the assumption that they have little effect on the simulated deposition and source-receptor relationships of Hg (US EPA, 1997; Bullock, 2000). However, two additional Hg species are measurable in ambient air, namely reactive gaseous Hg (RGM) and particulate Hg Hg(p), and both are known to deposit from the atmosphere at a significant rate. Boundary values for RGM and Hg(p) concentrations have been a concern since the early days of atmospheric mercury modeling, but they were often assumed to be negligible at regional boundaries far removed from industrial sources. Since then, rapid atmospheric conversion of GEM to oxidized forms was first reported by (Schroeder et al., 1998) and has been reported subsequently at a number of other polar and circumpolar locations. Evidence of significant concentrations of Hg(p) at high altitude has also been reported (Murphy et al., 2006). Because of these recently discovered opportunities for the creation and long-range transport of oxidized forms of Hg, boundary concentrations for all three measurable Hg species have become a greater concern for regional modeling of atmospheric Hg.

To address these concerns, global modeling with relatively coarse spatial resolution has been used as a preliminary step to define lateral boundary conditions for fine-resolution regional Hg modeling (Seigneur et al., 2001; Seigneur et al., 2004; US EPA, 2005). This technique can provide more realistic, time-resolved boundary information to regional models. Of course, global models are subject to the same uncertainties regarding their simulation of emission, transformation and deposition of Hg as are regional models.

Basic scientific uncertainties about the true behavior of atmospheric Hg are discussed in previous chapters of this volume. As discussed below, boundary concentrations derived from global models can differ significantly, as can the responses of the regional model when exposed to these differing boundary conditions.

16.2 Previous Testing and Application

Results from a study of atmospheric Hg models conducted by the Meteorological Synthesizing Centre - East (MSC-E) from 2000 to 2005 provide a good example of how regional modeling can be used to assess source-receptor relationships (Ryaboshapko et al., 2007). Three individual countries roughly equal in size but located in different parts of Europe were selected by MSC-E for source-receptor calculations: the UK, Poland and Italy.

Figure 16.1 (from Ryaboshapko et al., 2007) shows the range of model simulation results for total Hg deposition to these three countries during February 1999, August 1999 and for the entire year of 1999. The regional model simulations

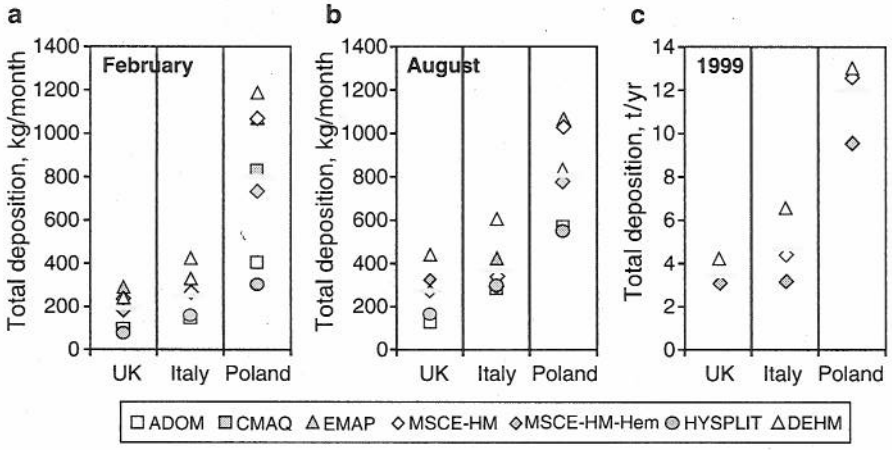

Figure 16.1 Total Hg deposition to the UK, Italy and Poland: (**a**) in February 1999; (**b**) in August 1999; and (**c**) in the whole year 1999. Symbols depict modelling results, light grey dashes show the ensemble average

were also used to calculate four parameters: (NAS) deposition caused by National Anthropogenic emission Sources of a given country; (EAS) deposition caused by all European Anthropogenic emission Sources except the anthropogenic sources of a given country; (GNR) deposition caused by Global anthropogenic sources (excluding European sources), Natural sources and Re-emission; and (ROF) Relative Out-Flow determined as a fraction of national anthropogenic emissions transported outside a given country. Two models (ADOM and CMAQ) stored their output data in such a way that EAS and GNR could not be calculated separately and had to be combined into a single parameter. These source-receptor parameters as calculated for Poland are shown in Figure 16.2 (from Ryaboshapko et al., 2007).

The range of results shown here for Poland is typical of those found for the U.K. and Italy. The differences in the simulated monthly and annual deposition amounts to each of the three countries are certainly significant, as are the differences in source attribution and relative outflow parameters. Because the regional models applied in this first Hg model intercomparison study were free to use different data for meteorology, boundary concentrations, and concentrations of Hg reactants within their individual modeling domains, it was not possible to determine if the simulation differences were primarily due to variations in basic process modeling or if they were simply due to the use of different boundary concentrations or other types of input data.

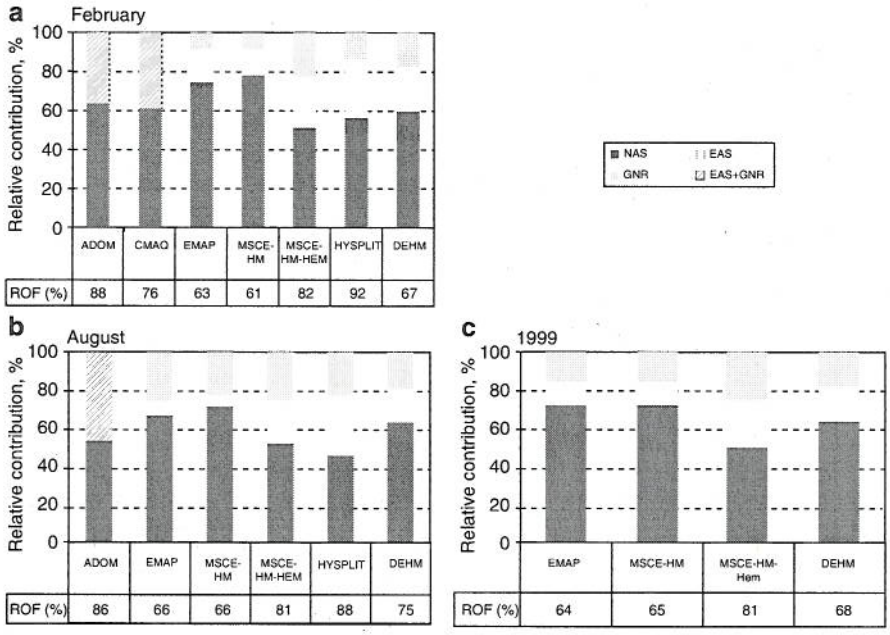

Figure 16.2 Contribution of national anthropogenic (NAS), European anthropogenic (EAS), and global, natural and re-emission sources (GNR) to Hg deposition over Poland: (**a**) in February 1999; (**b**) in August 1999; and (**c**) in the whole year 1999. Relative output flow (ROF) is also presented for each country

16.2.1 *Introduction of Dynamic Global Modeling for Boundary Conditions*

Concurrently with the MSC-E study, researchers at Atmospheric and Environmental Research, Inc., (AER) developed a model nesting approach for Hg using a global chemical transport model (CTM-Hg) and an imbedded regional model covering North America (Seigneur et al., 2001). The global CTM-Hg described in Shia et al. (1999) provided a horizontal resolution of 8° latitude and 10° longitude and a vertical resolution of 7 layers between the Earth's surface and ~12 km altitude and 2 layers between ~12 km and 30 km altitude. The regional model employed was the Trace Element Analysis Model (TEAM) described in Pai et al. (1997). It provided a horizontal resolution of 100 km and a vertical resolution of 6 layers between the Earth's surface and 6 km altitude. Both models were used to simulate the year of 1998. Figure 16.3 (from Seigneur et al., 2001) shows a comparison of annual average total gaseous Hg (TGM) as simulated by the TEAM along with available measurements for 6 U.S. states.

Because TGM includes both Hg^0 and RGM, knowledge about its concentration in air provides only limited guidance for estimating Hg deposition flux. Atmospheric concentrations of RGM are usually a small but highly variable fraction (0.1-10%) of the TGM concentration and RGM is much more readily deposited than Hg^0. Figure 16.4 (from Seigneur et al., 2001) shows a comparison of simulated and observed annual wet deposition fluxes of Hg for 12 U.S. states. The observations were obtained from the Mercury Deposition Network (MDN) described by (Vermette et al., 1995).

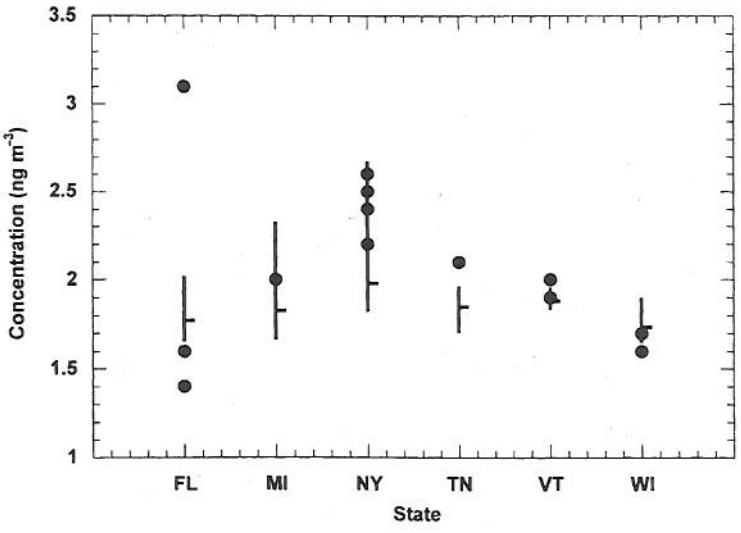

Figure 16.3 Comparison of simulated annual-average gas-phase mercury concentrations ($ng\ m^{-3}$) with measured data for 1998

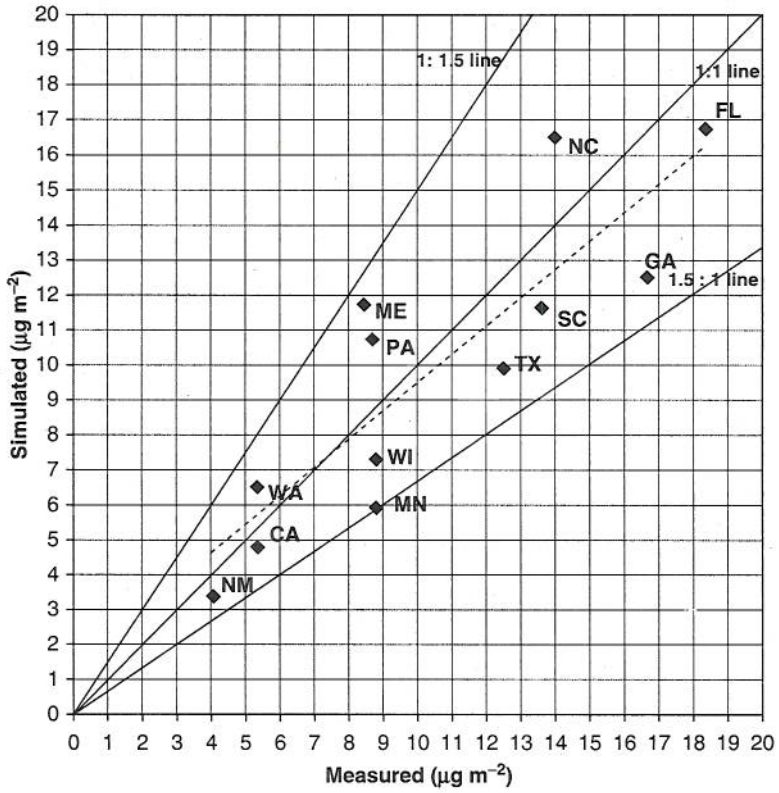

Figure 16.4 Comparison of simulated and measured 1998 wet deposition fluxes ($\mu g\ m^{-2}$) of total mercury over the United States (CA, California; FL, Florida; GA, Georgia; ME, Maine; MN, Minnesota; NC, North Carolina; NM, New Mexico; PA, Pennsylvania; SC, South Carolina; TX, Texas; WA, Washington; WI, Wisconsin)

The observed values shown are the average of all MDN observations for each state where 8 of the 12 states had 2 or more MDN observations. These results certainly demonstrated some modeling skill for Hg wet deposition. However, as discussed in Seigneur et al. (2001), a number of uncertainties affected these modeling results. Some of those uncertainties had to do with global "background" emissions and atmospheric transformations of Hg during intercontinental transport. The lack of information about dry deposition of the Hg species simulated was also identified as a source of uncertainty. Nonetheless, this first integration of regional and global modeling was hailed as an advance towards a better modeling approach for atmospheric Hg. However, the models themselves were recognized to be deficient with regard to scientific process information for Hg and this deficiency remains an issue in atmospheric Hg modeling today.

Another example of using global models to provide boundary data for regional modeling of atmospheric mercury is modeling performed by the U.S. EPA to

investigate proposed reductions in mercury emissions from coal-fired electric generating units (EGUs) within the U.S. For this analysis, the CMAQ model was used to simulated the year 2001 with and without future Hg emission reductions. Input data for initial and boundary concentrations of Hg and other pertinent air pollutants were derived from the global GEOS-Chem model (Selin et al., 2007). This approach has now been adopted by the U.S. EPA and it represents an advance from earlier CMAQ modeling of Hg where time-invariant boundary values were used (Bullock and Brehme, 2002).

To evaluate the accuracy of the CMAQ 2001 simulation, results for annual wet deposition of total Hg were compared to observations from the MDN in much the same way as Seigneur et al. (2001) had done for the TEAM model. However, many more MDN observations were available for the 2001 time frame of the US EPA study. Also, each individual observation was compared to its corresponding model result without any state-by-state averaging. The results of this comparison are shown in Figure 16.5. Discounting one comparison for the MDN station in British Columbia (BC) where the model input data for precipitation were found to be grossly inaccurate, the CMAQ base-case simulation resolved about 60% of the observed variance in annual Hg wet deposition. Figure 16.6 shows the simulated reduction in Hg deposition from all Hg emission controls expected to be in force by 2020. Significant reductions in Hg deposition are confined to a rather small fraction of the continental U.S. This suggests that much of the Hg deposition in the relatively unaffected areas might be attributable to Hg transported into the modeling domain. The following section

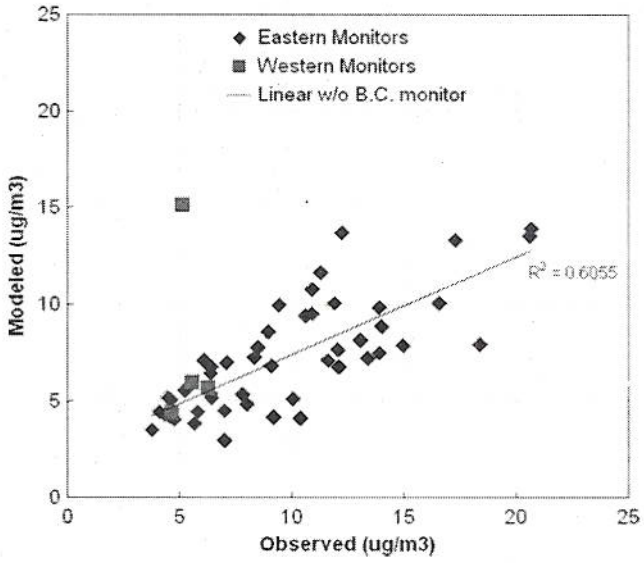

Figure 16.5 Comparison of CMAQ-simulated total Hg wet deposition for 2001 to observations from the Mercury Deposition Network (MDN). The result for the MDN monitor in British Columbia (B.C.) is discounted in the calculation of the least-squares linear fit and the R^2 statistic because of errors in the precipitation input data for that location

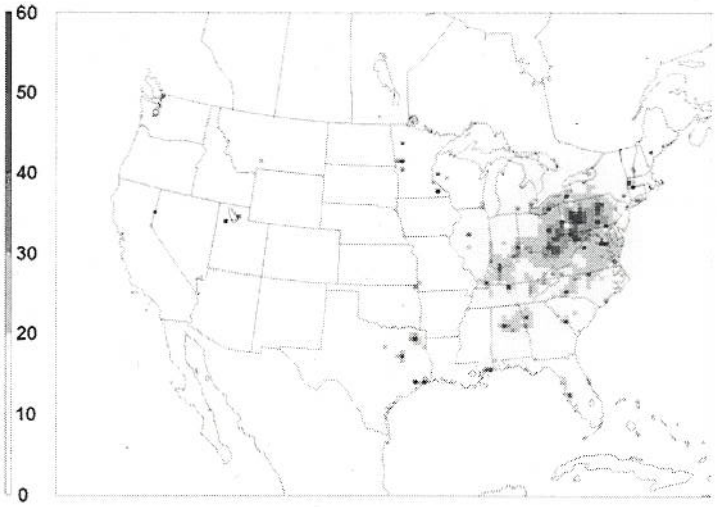

Figure 16.6 CMAQ-simulated results for the percent reduction in mercury deposition by 2020 when proposed regulations are fully implemented

discusses an investigation of the importance of boundary fluxes of Hg to simulations with the CMAQ model and two other regional models.

16.3 Testing Model Sensitivities to Intercontinental Transport

The North American Mercury Model Intercomparison Study (NAMMIS) was organized by the US EPA to apply regional-scale atmospheric Hg models in a tightly constrained testing environment with a modeling domain in North America where standardized measurements of Hg wet deposition are available (Bullock et al., 2008a; 2008b). The intent was to have all regional models in the study use exactly the same input data for initial and boundary conditions, meteorology and emissions, and to have all models applied to exactly the same horizontal modeling domain so that the effects of differing input data could be reduced, thus allowing the effects of differing scientific process treatments to be better understood. The ultimate goal was to determine which scientific process uncertainties are contributing most to observed discrepancies in model simulations of Hg deposition. Three regional atmospheric Hg models were applied: CMAQ, REMSAD, and TEAM. Each was applied to simulate the entire year of 2001 using three different initial condition and boundary condition (IC/BC) data sets, each developed from a different global model. The global models used were the CTM-Hg and the GEOS-Chem models (both described above) and the Global/Regional Atmospheric Heavy Metals (GRAHM) model described in Dastoor and Larocque (2004). All regional modeling results were compared to observed Hg

wet deposition data from the MDN and special event-based monitoring at the Proctor Maple Research Center (PMRC) near Underhill, VT (Keeler et al., 2005).

The global models were not the primary subjects of the study, but they did exhibit significant differences in their simulated air concentrations of Hg^0, RGM and particulate Hg at the lateral boundaries of the regional modeling domain for the NAMMIS. Thus, the three IC/BC data sets provided an opportunity to investigate the effect of uncertainty regarding intercontinental transport of Hg species. Figure 16.7 shows the annual average air concentrations of Hg^0, RGM and particulate Hg across the western boundary of the regional domain as determined from the CTM, GEOS-Chem and GRAHM global simulations. While the patterns of Hg^0 air concentration are somewhat different, it is unlikely that these differences would affect the resulting deposition pattern for total Hg across the regional model domain since Hg^0 is deposited quite slowly under most conditions. However, the differences in RGM and particulate Hg concentration patterns may very well affect the total-Hg deposition pattern and the modeling accuracy as compared to observations. Figure 16.8 shows the resulting R^2 correlation statistic for simulated annual Hg wet deposition compared to observed values for each of the regional model simulations conducted in the NAMMIS. Figure 16.9 shows the mean fractional bias for each simulation while Figure 16.10 shows the mean fractional error. In addition to model-to-model differences in these statistics, these figures also show a considerable sensitivity of all three models to the IC/BC data sets used.

The US EPA has performed additional qualitative analysis of the CMAQ modeling results for Hg wet deposition obtained from the three IC/BC data sets from the NAMMIS (Bullock, 2008). This analysis looked at the concentration patterns of Hg^0, RGM and particulate Hg along all four lateral boundaries for each IC/BC set. It was shown that Hg^0 concentrations at the boundaries had a slight and somewhat negative correlation to the magnitude of the wet deposition flux, both at locations near the boundaries and within the interior of the modeling domain.

This is because the lower Hg^0 concentrations simulated by the global models were associated with higher RGM and particulate Hg concentrations as a product of simulated oxidation of Hg^0 to gaseous and aerosol $Hg^{(II)}$ forms. Indeed, the analysis of CMAQ sensitivity to RGM at the boundaries showed obvious positive correlations with simulated Hg wet deposition, especially near the boundaries. The CMAQ showed less sensitivity to particulate Hg concentrations at the boundaries as compared to RGM, but positive correlations with total-Hg wet deposition were noted.

In general, the NAMMIS and follow-on studies have shown that RGM concentrations specified at the lateral boundaries of regional modeling domains can have a significant effect on the intensity of Hg wet deposition simulated, not only near the boundary but also in interior regions of the domain. Lateral boundary concentrations specified for other mercury species may also impact simulations of wet deposition. It is difficult to draw firm conclusions about the effect of Hg^0 boundary concentrations brought about by simulated or actual intercontinental transport without also considering the effect of boundary fluxes of important oxidants of mercury (e.g., ozone, hydroxyl radical, halogens).

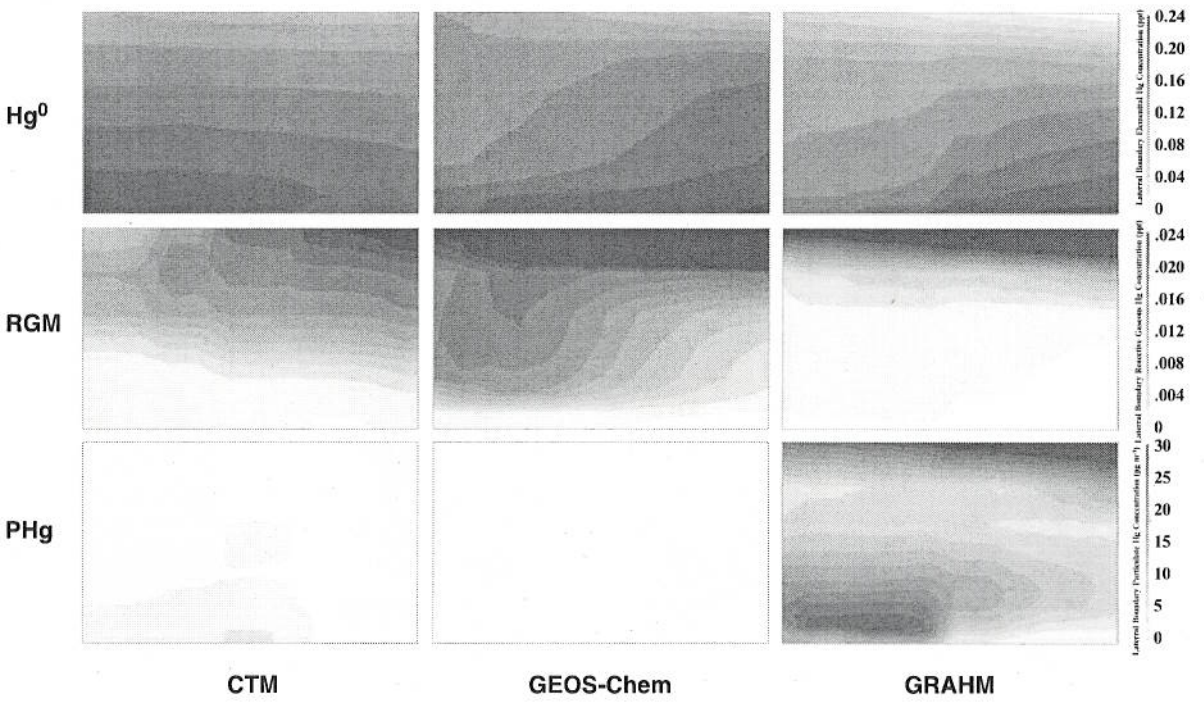

Figure 16.7 Annual average air concentrations of Hg^0, RGM and particulate Hg across the western boundary of the NAMMIS regional domain as determined from the CTM, GEOS-Chem and GRAHM global simulations

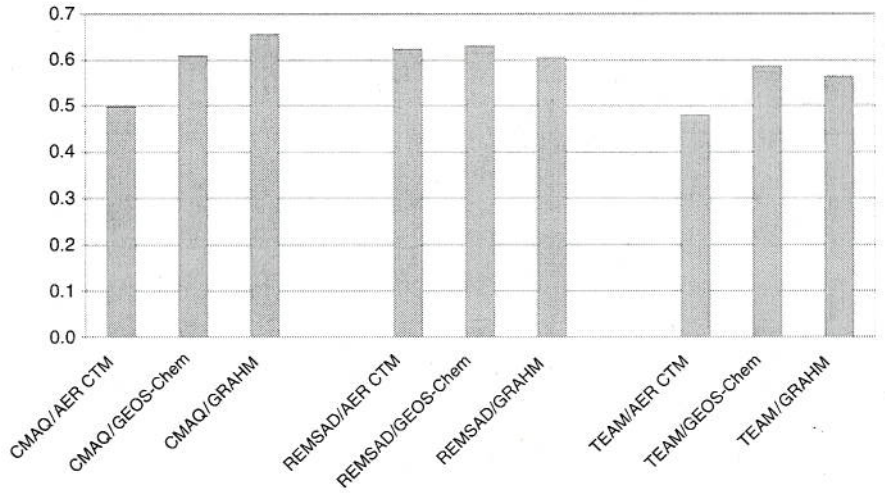

Figure 16.8 R^2 correlation statistics for simulated annual Hg wet deposition compared to observed values for each of the regional model simulations conducted in the North American Mercury Model Intercomparison Study

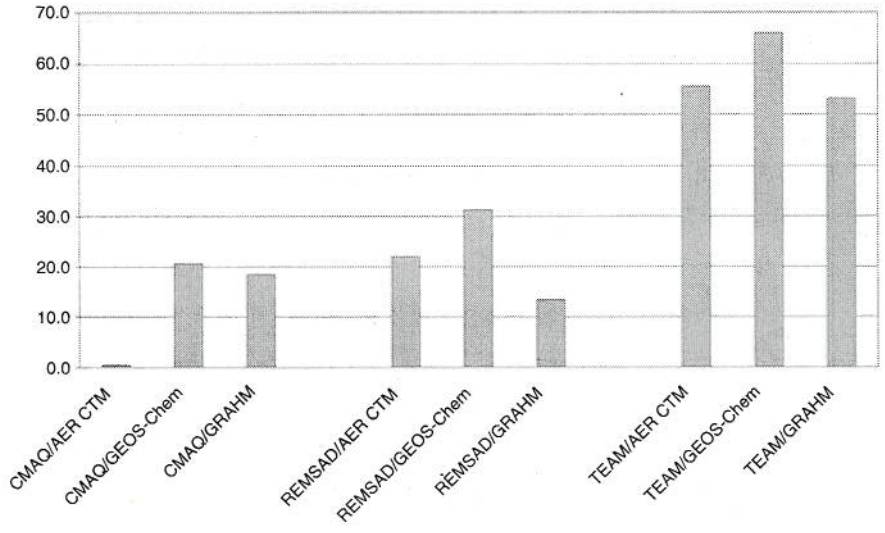

Figure 16.9 Mean fractional bias (percent) for simulated annual Hg wet deposition compared to observed values for each of the regional model simulations conducted in the North American Mercury Model Intercomparison Study

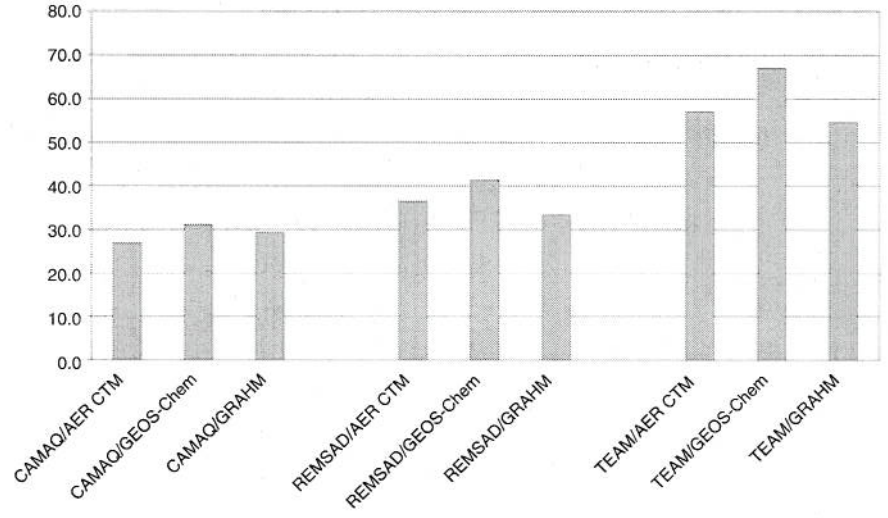

Figure 16.10 Mean fractional error (percent) for simulated annual Hg wet deposition compared to observed values for each of the regional model simulations conducted in the North American Mercury Model Intercomparison Study

16.4 Future Research and Policy Implications

Numerical simulation modeling has played a critical role in the advancement of our scientific understanding of many environmental and ecological issues. As we make new observations and gain knowledge about specific aspects of atmospheric mercury behavior, that knowledge is incorporated into numerical models by adding to or modifying its formulations. These new formulations are tested when model simulations are compared to new independent observations. This iterative process of observation and model development has provided us with the models we have today. Certainly, there remain some serious gaps in our understanding of atmospheric mercury emission, transformation and deposition. Scientific uncertainty affects all atmospheric mercury models, both regional and global. Whether current models are good enough to be used as a basis for environmental protection policy is as much a political question as a scientific one. It is interesting to note that, for the regional models applied in the NAMMIS, the lowest mean fractional error for annual Hg wet deposition was around 30%, which is comparable to the inter-annual variability seen in Mercury Deposition Network observations over the past few years (see http://nadp.sws.uiuc.edu/mdn/maps/). This suggests that the modeling error from the best models currently available introduces a similar level of uncertainty as does the choice of the particular annual period to be simulated.

Gaps in our understanding of chemical and physical processes are discussed in the previous chapter. Here we have shown that regional model simulations of atmospheric mercury are sensitive to the boundary concentrations of Hg specified as

input data. Global models do not require spatial boundary information by nature of their definition. During the last few years, it has become common practice for regional models to be supplied their necessary boundary information based on global model simulations. However, we have shown here that current global models do not agree on the Hg concentrations that the regional models are sensitive to. There is an obvious need for a globally common framework for observation and modeling and a unified assessment of global-scale mercury transport.

In the past 10 years, new and improved information on sources and behaviour of atmospheric mercury has certainly improved our basic scientific understanding. For example, the discovery of gaseous mercury depletion events in circum-polar locations and the subsequent determination that halogens from sea water were chemically oxidizing elemental mercury to forms that rapidly deposited to the local snowpack. High altitude measurements of discrete mercury species have found that elemental mercury air concentrations tend to decrease with height, and that the concentrations of oxidized forms of Hg tend to increase with height, suggesting that significant oxidation of Hg is taking place in the middle and/or upper levels of the troposphere. Laboratory experiments have been used to estimate kinetic rate constants that all models need for their simulations of chemical transformations. Increases in computing speeds and memory capacity have allowed the application of fine-scale modeling over larger spatial and temporal domains. However, we still cannot simulate global scale atmospheric mercury at the fine spatial resolution required for environmental assessment of specific water bodies where Hg contamination is a problem.

One aspect of using global models to provide boundary information for regional models deserves special attention for future development. Global and regional models need to use consistent meteorological data in their respective simulations. For example, all models simulate higher concentrations of RGM in air masses that have not been subject to wet deposition over a long period of time. If a global model simulates no precipitation and high RGM concentrations at a regional model boundary where the regional model simulates some precipitation, the regional model will show excessively large wet deposition fluxes near that boundary. In reality, truly dry air masses do not develop precipitation instantaneously. Instead, cloud water droplets are gradually formed where significant quantities of RGM could be dissolved, chemically reduced to Hg^0 and released back to the air before precipitation and wet deposition could occur. Wind flow patterns also need to be consistent between the global and regional models. The global model might simulate wind flow from an area of concentrated emissions within the regional model domain towards the boundary while the regional model instead simulates wind flow in the opposite direction. This results in the regional model simulating in-flow and deposition of Hg from internal sources. In the opposite case, where the regional model simulates out-flow, the boundary concentrations from the global model are of no consequence at all. So the average effect of inconsistent wind flow between the global and regional models is an artificial recirculation of emissions from within the regional domain. This problem of inconsistent meteorological data can be avoided through the use of nested-grid meteorological modeling where the global model and the

regional model are essentially the same model being applied with the same meteorological information on two different domains with different spatial resolutions within those domains.

Obviously, source-receptor relationships simulated by regional models are influenced by the boundary air concentrations specified for all pertinent pollutants. Regional models may be able to provide useful information about the relative importance of the Hg emission sources within their modeling domains without detailed and accurate boundary flux information. However, the importance of Hg emission sources located outside these domains cannot be assessed with any confidence without accurate boundary concentration data for all Hg species and their reactants. The fact that global models do not agree in their simulated concentrations for pertinent species is certainly cause for concern. We must develop standardized ambient sampling techniques for atmospheric Hg that can be deployed all around the globe and at all levels of the atmosphere, not just at the surface. Then we must actually deploy them with a consistent and continuous coverage of Earth's atmosphere in order to know if any global model is capable of providing realistic boundary data to regional models.

References

Bullock, O.R., 2000. Modeling assessment of transport and deposition patterns of anthropogenic mercury air emissions in the United States and Canada. *Science of the Total Environment* 259, 145–157.

Bullock, O.R., Brehme, K.A., 2002. Atmospheric mercury simulation using the CMAQ model: formulation description and analysis of wet deposition results. *Atmospheric Environment* 36, 2135–2146.

Bullock, O. R., 2008b. The effect of lateral boundary values on atmospheric mercury simulations with the CMAQ model. In: Carlos Borrego and Ana Isabel Miranda, editors. *Air Pollution Modeling and Its Applications XIX.* Springer, Dordrecht, The Netherlands, in press.

Bullock, O.R., Atkinson, D., Braverman, T., Civerolo, K., Dastoor, A., Davignon, D., Ku, J.-Y., Lohman, K., Myers, T., Park, R., Seigneur, C., Selin, N.E., Sistla, G., Vijayaraghavan, K., 2008a. The North American Mercury Model Intercomparison Study (NAMMIS). Part 1: Study description and model-to-model comparisons. *Submitted to Journal of Geophysical Research*

Christensen, J.H., 1997. The Danish Eulerian hemispheric model - A three-dimensional air pollution model used for the Arctic. *Atmospheric Environment* 31, 4169–4191.

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. *Environmental Research* 95, 247–265.

Dastoor, A.P., Larocque, Y., 2004. Global circulation of atmospheric mercury: A modeling study. *Atmospheric Environment* 38, 147–161.

ICF, 2005. User's Guide to the Regional Modeling System for Aerosols and Deposition (REMSAD), Version 8. ICF Consulting/SAI, San Francisco, CA.

Ilyin, I., Ryaboshapko, A., Afinogenova, O., Berg, T., Hjellbrekke, A.-G., Lee, D.S., 2002. Lead, cadmium and mercury transboundary pollution in 2000. EMEP/MSC-E Report 5/2002, Meteorological Synthesizing Centre - East, Moscow, Russia, 131 pp. http://www. msceast. org. html.

Keeler, G. J., Gratz, L., Al-Wali, K., 2005. Influences on the long-term atmospheric mercury wet deposition at Underhill, Vermont. *Ecotoxicology* 14, 71–83.

Murphy, D.M., Hudson, P.K., Thomson, D.S., Sheridan, P.J., Wilson, J.C., 2006. Observations of mercury-containing aerosols. *Environmental Science & Technology* 40, 3163–3167.

Pai, P., Karamchandani, P., Seigneur, C., 1997. Simulation of the regional atmospheric transport and fate of mercury using a comprehensive Eulerian model. *Atmospheric Environment* 31, 2717–2732.

Petersen, G., Bloxam, R., Wong, S., Munthe, J., Kruger, O., Schmolke, S.R., Kumar, A.V., 2001. A comprehensive Eulerian modelling framework for airborne mercury species: model development and applications in Europe. *Atmospheric Environment* 35, 3063–3074.

Ryaboshapko, A., Bullock, O.R., Christensen, J., Cohen, M., Dastoor, A., Ilyin, I., Petersen, G., Syrakov, D., Travnikov, O., Artz, R.S., Davignon, D., Draxler, R.R., Munthe, J., Pacyna, J., 2007. Intercomparison study of atmospheric mercury models: 2. Modelling results vs. long-term observations and comparison of country atmospheric balances. *Science of the Total Environment* 377, 319–333.

Schroeder, W.H., Anlauf, K.G., Barrie, L.A., Lu, J.Y., Steffon, A., Schneeberger, D.R., Berg, T., 1998. Arctic springtime depletion of mercury. *Nature* 394, 331–332.

Seigneur, C., Karamchandani, P., Lohman, K., Vijayaraghavan, K., Shia, R.-L., 2001. Multiscale modeling of the atmospheric fate and transport of mercury. *Journal of Geophysical Research* 106(D21), 27,795-27,809.

Seigneur, C., Vijayaraghavan, K., Lohman, K., Karamchandani, P., Scott, C., 2004. Global source attribution for mercury deposition in the United States, *Environmental Science & Technology* 38, 555–569.

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. *Journal of Geophysical Research* 112, D02308, doi:10.1029/2006JD007450.

Shia, R.L., Seigneur, C., Pai, P., Ko, M., Sze, N.D., 1999. Global simulation of atmospheric mercury concentrations and deposition fluxes. *Journal of Geophysical Research* 104(D19), 23,747-23,760.

Syrakov, Dimiter, Prodanova, Maria and Slavov Kiril (2005)Bulgarian Emergency Response System: Description and Ensemble Performance NATO Science Series Series IV: Earth and Environmental Series Advances in Air Pollution Modeling for Environmental Security Proceedings of the NATO Advanced Research Workshop on Advances in Air Pollution Modeling for Environmental Security Borovetz, Bulgaria 8–12 May 2004-10.1007/1-4020-3351-6_33

US EPA, 1997. Mercury Study Report to Congress. Fate and Transport of Mercury in the Environment, Vol. III. EPA-452/R-97-005, US Environmental Protection Agency, US Government Printing Office, Washington, DC.

US EPA, 2005. Technical Support Document for the Final Clean Air Mercury Rule: Air Quality Modeling, EPA Office of Air Quality Planning and Standards, Research Triangle Park, NC 27711, March 2005. (http://www.epa.gov/ttn/atw/utility/aqm_oar-2002-0056-6130.pdf)

Vermette, S., Lindberg, S., and Bloom, N., 1995. Field tests for a regional Mercury Deposition Network - Sampling design and preliminary test results. *Atmospheric Environment* 29, 1247–1251.