Appendix D: Description of REMSAD Atmospheric Mercury Deposition Model and Emission Inventories

This appendix provides a brief description of the U.S. Environmental Protection Agency (USEPA) model, Regional Modeling System for Aerosols and Deposition (REMSAD), for quantifying atmospheric mercury deposition in the United States. Water Board Staff used output from the REMSAD model to characterize atmospheric deposition patterns throughout California. Water Board staff used the REMSAD model to characterize atmospheric deposition in California because it was designed specifically to support TMDL development and implementation and because its simulated spatial distribution of mercury deposition is consistent with observed deposition patterns.

This appendix also provides a summary of the REMSAD emission inventory for 2001, along with the USEPA's National Emissions Inventories for 2002, 2005, and 2008 for comparison, attached as Tables D.1 and D.2. The below description of the REMSAD model refers to these inventories. In addition, they are a critical component of the assessment of emission sources that contribute to mercury impaired reservoirs (see Section 7.4 Atmospheric Deposition in Chapter 7 of this report).

REMSAD Model Objectives and Design

The objective of the USEPA's REMSAD model is to quantify contributions of specific sources and source categories to mercury deposition within each of the lower 48 state and to provide state and local air and water quality agencies with 1) an improved understanding of the sources and mechanisms contributing to mercury deposition; 2) supporting information for development of TMDLs; and 3) assistance in developing implementation plans for TMDLs and related activities designed to help achieve water quality standards. The REMSAD modeling protocols were used to develop the Northeastern States Mercury TMDL approved by USEPA in December 2007 (USEPA 2008a; CDEP et al. 2007). In addition, REMSAD was used for the Devil's Lake TMDL Pilot Project in Wisconsin and to support TMDL development in southern Louisiana and Maryland (USEPA 2008a).

REMSAD is a three-dimensional grid model designed to calculate the concentrations of both inert and chemically reactive pollutants by simulating the physical and chemical processes in the atmosphere that affect pollutant concentrations (USEPA 2008a). The model simulates the transfer of mercury mass between its different oxidation states and its gas and particulate phases, as well as both wet and dry deposition. The REMSAD model uses "tagging", which allows tracking of emissions through space and time. "Tags" can be individual sources, source types, and source regions, both separately and in combination.

The REMSAD modeling domain encompasses the continental United States and portions of Canada and Mexico, with a 12-km horizontal grid resolution over the entire United States

portion of the domain. There are 3,037 model grid cells that intersect California. The annual simulation period is 2001. The baseline point source emissions and meteorological data represent the 2001 time period; estimates of mercury emissions from nonpoint sources (e.g., mobile sources) were not available at the time REMSAD was developed. The meteorological data used in REMSAD were those that were developed by USEPA for use in their evaluation of emissions rules (e.g., the Clean Air Interstate Rule and the Clean Air Mercury Rule) (USEPA 2008a, 2005a and b).

The REMSAD model developers placed emphasis on the preparation and quality assurance of the 2001 mercury emissions inventory (see Table D.1 for a summary of the inventory emissions). The starting point for the mercury inventory preparation was the 2001 emissions data utilized by USEPA in the Clean Air Mercury Rule modeling. Model development included a detailed review and revision of the mercury emissions for each state in order to better represent the 2001 time period. This review was conducted by ICF, USEPA, and state agencies; revisions were incorporated based on information provided by the states (USEPA 2008a).

Tags were assigned to the largest sources in each state as well as a range of source types and potentially important contributors to local and regional mercury deposition in areas with known or suspected mercury water quality programs; not every single source was tagged. In addition, tags were assigned to contributions from global background ("boundary conditions") and re-emissions of previously deposited mercury to mercury deposition. The REMSAD simulations used three alternate specifications of the boundary conditions based on global model simulations. Each of the three global models, the Chemical Transport Model (CTM) (developed and applied by Atmospheric and Environmental Research, Inc.), the Global/Regional Atmospheric Heavy Metals model (GRAHM) (developed and applied by Environment Canada), and the GEOS-Chem model (developed and applied by researchers at Harvard University), utilized the same year 2000 emissions inventory, which includes both natural and anthropogenic sources. Water Board staff used the output from the REMSAD simulation that incorporated the average of the three global models' output for all graphs and calculations in this report.

Peer Review and Comparison to Empirical Results

The REMSAD model was peer reviewed in 1999 (Seigneur et al. 1999) and the modeling in the Devils Lake TMDL Pilot (including the tagging application) was subjected to an external peer review (USEPA 2008a). REMSAD was included in the North American Mercury Model Intercomparison Study for mercury and the performance and response of the model was found to be reasonable (Bullock et al. 2008).

Also, the model developers used a variety of graphical analyses and statistical measures to evaluate REMSAD model performance on a seasonal and annual basis for the most recent model simulation runs. The model developers compared REMSAD wet deposition values to concentration data from 98 Mercury Deposition Network (MDN) monitors and deposition from 53 MDN monitors. The model developers found that the simulated spatial distribution of wet mercury deposition to be consistent with observed wet deposition patterns. The REMSAD simulation results tend to overestimate wet deposition of mercury, as compared to the MDN monitoring data.

The model developers noted that emerging research suggests that the MDN measurement techniques may underestimate wet deposition of mercury by approximately 16%, but they used the MDN data without any adjustment. It was not possible for the model developers to evaluate the simulated dry deposition results because an adequate network of dry deposition monitoring data was not available. However, Bullock and others (2008) noted that simulated dry deposition of mercury was found to vary between REMSAD and two other regional-scale models by nearly a factor of 10 in some locations (especially at high altitudes), and that the REMSAD model simulated much less dry deposition of mercury than the other two models evaluated.

The model results are very similar to empirical (monitoring) wet and dry deposition rates throughout California and eastern Nevada. The similarities are remarkable given: there is a broad range of deposition rates across California; the periods of the precipitation data used in the deposition calculations were different from the wet deposition concentration monitoring periods; and the wet and dry deposition monitoring periods are different from the REMSAD simulation period. Tables D.3 and D.4 provide a qualitative comparison of wet and dry deposition rates observed at different sites in California compared to the REMSAD 2001 deposition values and Figure D.1 shows the study locations.

In addition, the REMSAD results are in closer agreement with observed dry deposition rates than those of a different model used by USEPA for TMDL development for a southern California reservoir. The South Coast Air Quality Management District calculated a dry deposition rate of 11.8 g/km^2/yr (upper bound) for Big Bear Reservoir based on monitoring data for November 2010 through October 11 (SCAQMD 2012; see Table D.4). This rate is very similar to the rate of 8.9 g/km^2/yr estimated by the REMSAD model for 2001. However, the Community Multiscale Air Quality (CMAQ) mercury transport model estimated dry deposition rates of 21 and 40 g/km^2/yr for 2001 and 2002, respectively (Tetra Tech 2008), which are two to four times higher than the rate based on available monitoring data.

Most importantly, the REMSAD model results for California and the United States agree with the Mercury Deposition Network observations that indicate wet deposition rates throughout much of California are low compared to wet deposition rates in the eastern United States.

Changes in California Emissions Data Since 2001

As noted earlier, estimates of mercury emissions from anthropogenic nonpoint sources were not available at the time the REMSAD model was developed. None-the-less, REMSAD model output is expected to characterize deposition patterns across the state adequately for TMDL development.

As summarized in Table D.1, estimates for several nonpoint sources have become available since 2001 and some appear to be substantial. On-road diesel heavy duty vehicles and non-road diesel equipment comprise about 10% to 20% of all anthropogenic emissions, depending on the inventory year. Consequently, the model could under-predict deposition rates in and downwind of areas where there are abundant mobile sources.

However, as Tables D.1 and D.2 illustrate, point source emissions —especially facility emissions— account for much of the anthropogenic emissions in California. Facility emissions

accounted for about 50% of emissions reported in the 2002 and 2005 inventories, and more than 70% in the 2008 inventory.

In addition, Water Board staff reviewed 2008 point and nonpoint source emissions by county to evaluate statewide distributions. Counties with the highest nonpoint source emissions also had the highest point source emissions. As a result, the REMSAD model output is expected to characterize deposition patterns across the state adequately for understanding where California emissions may be substantial contributors to mercury impaired water bodies.

Changes in California Emissions Sources Since 2001

California emissions decreased by more the 50% between 2001 and 2008. Emissions from several California emission sectors decreased, particularly municipal and hazardous waste combustion, fuel combustion associated with energy production and industrial boilers, cement production, and oil and gas production. Some emissions types, such as Portland cement production, vary from year to year as a result of changes in economic demand. Others, such as municipal waste incineration, have had substantial reductions due to implementing emission controls. These decreases, as well as the additional decreases expected from the implementation of recent state and federal regulations, are addressed by the TMDL allocation and implementation strategies described in Chapters 8, 9 and 10 of this report.

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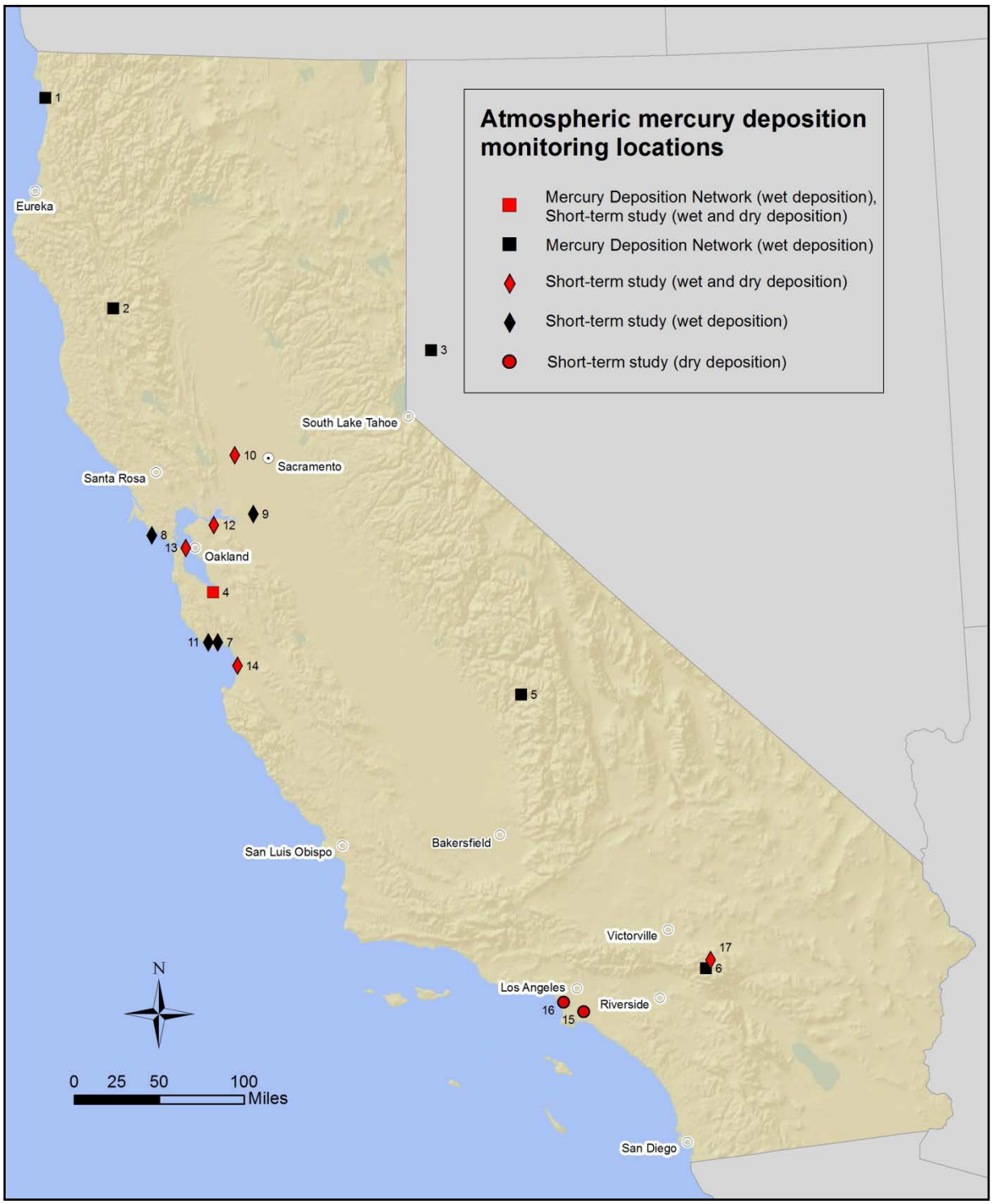

Figure D.1: Atmospheric mercury deposition monitoring sites in California and western Nevada