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COMPUTING ATMOSPHERIC NUTRIENT LOADS TO THE CHESAPEAKE BAY WATERSHED AND TIDAL WATERS[1]

Lewis C. Linker, Robin Dennis, Gary W. Shenk, Richard A. Batiuk, Jeffrey Grimm, and Ping Wang[2]

ABSTRACT: Application of integrated Chesapeake Bay models of the airshed, watershed, and estuary support air and water nitrogen controls in the Chesapeake. The models include an airshed model of the Mid-Atlantic region which tracks the estimated atmospheric deposition loads of nitrogen to the watershed, tidal Bay, and adjacent coastal ocean. The three integrated models allow tracking of the transport and fate of nitrogen air emissions, including deposition in the Chesapeake watershed, the subsequent uptake, transformation, and transport to Bay tidal waters, and their ultimate influence on Chesapeake water quality. This article describes the development of the airshed model, its application to scenarios supporting the Chesapeake Total Maximum Daily Load (TMDL), and key findings from the scenarios. Key findings are that the atmospheric deposition loads are among the largest input loads of nitrogen in the watershed, and that the indirect nitrogen deposition loads to the watershed, which are subsequently delivered to the Bay are larger than the direct loads of atmospheric nitrogen deposition to Chesapeake tidal waters. Atmospheric deposition loads of nitrogen deposited in coastal waters, which are exchanged with the Chesapeake, are also estimated. About half the atmospheric deposition loads of nitrogen originate from outside the Chesapeake watershed. For the first time in a TMDL, the loads of atmospheric nitrogen deposition are an explicit part of the TMDL load reductions.

(KEY TERMS: water policy; simulation; total maximum daily load (TMDL); watershed management; nitrogen; Chesapeake Bay; Community Multiscale Air Quality Model; atmospheric deposition.)

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INTRODUCTION

The U.S. Environmental Protection Agency (USEPA) has established the Chesapeake Bay Total Maximum Daily Load (TMDL), which is a historic and comprehensive watershed restoration plan with rigorous accountability measures to restore clean water in the Chesapeake Bay and in the region's streams and rivers. In the Chesapeake TMDL, reduction in nitrogen and phosphorus nutrient loads are central to restoring water quality, and allocations of these loads to specific Chesapeake watershed jurisdictions and basins were developed. Key measures of Chesapeake

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water quality and restoration are the living resource-based water quality standards of dissolved oxygen, chlorophyll, and water clarity (USEPA, 2003a, b, c, 2004, 2007, 2010a). According to model analyses, the nutrient and sediment allocations, if attained, would reduce pollutant loads sufficiently to achieve all Chesapeake water quality standards. Included in the allocation was a specific allocation of atmospheric nitrogen loads to tidal waters.

Quantification of the deposition loads to the Chesapeake began with assessments of key oxidized (NO_x) and reduced (NH_3) loads in both wet and dry deposition (Fisher *et al.*, 1988; Tyler, 1988; Fisher and Oppenheimer, 1991; Hinga *et al.*, 1991). In the case of the open waters of the tidal Chesapeake and watershed, organic nitrogen deposition also needs to be considered (Knap *et al.*, 1986; Scudlark and Church, 1993; Neff *et al.*, 2002), particularly in the case of linkage to the mass balance watershed model (Shenk and Linker, this issue) and estuary model (Cerco *et al.*, 2010; Cerco and Noel, this issue) in which all estimated nutrient input loads are included. Estimates of coastal ocean loads of nitrogen deposition (Howarth *et al.*, 1995; Howarth, 1998; Paerl *et al.*, 2002; Fennel *et al.*, 2006) are considered in the Chesapeake TMDL as well, and were handled through adjustment of the ocean concentration boundary condition in the estuarine model (USEPA, 2010b).

Building on the nitrogen deposition research and extensive monitoring data in the Chesapeake region are state-of-the-art integrated models of the airshed, watershed, and estuary. The integrated models allow tracking of the transport and fate of nitrogen air emissions, including deposition in the Chesapeake watershed, the subsequent uptake, transformation, and transport to Bay tidal waters, and their ultimate influence on Chesapeake water quality.

The work provides an example of the integration of air and water controls in a large aquatic ecosystem in the United States (U.S.) (Figure 1). For the first time in a TMDL, the loads of atmospheric nitrogen deposition are an explicit allocated load in the Chesapeake TMDL. In determining the allowable loading from air deposition, USEPA separated the nitrogen deposition into two discrete parcels. The first is deposition occurring on the land and nontidal waters which is subsequently transported to the Bay, also called indirect deposition. The second is atmospheric deposition occurring directly onto the Bay's tidal surface waters, also called direct deposition. The nitrogen TMDL allocation given to the USEPA is 7.1 million kg per annum for atmospheric deposition of total nitrogen directly deposited to the tidal waters of the Chesapeake. Reducing current nitrogen deposition loads to achieve the allocation will be accomplished through national programs of NO_x emission reductions.

Atmospheric deposition loads are among the largest input loads of nitrogen in the watershed and indirect nitrogen deposition loads to the watershed, which are subsequently delivered to the Bay, are larger than the direct loads of atmospheric nitrogen deposition to Chesapeake tidal waters. Atmospheric deposition loads of nitrogen deposited in Atlantic coastal waters adjacent to the Bay, which are exchanged with the Chesapeake, are also estimated. About half the atmospheric deposition loads of nitrogen originate from emission sources outside the Chesapeake watershed (USEPA, 2010b).

The 7.1 million kg TMDL allocation of direct nitrogen deposition to the tidal waters is an *explicit allocation* that is quantified and tracked in two-year milestones. On the other hand, the loads of atmospheric deposition to the watershed, i.e., indirect deposition are considered to be a *reference allocation* that is implicitly quantified as one of the loads to the watershed along with point source, manure, fertilizer, and septic system loads. A reference allocation as it is defined here is not a specific legally binding TMDL allocation, but is an agreed to reduction in the watershed's indirect atmospheric deposition loads of nitrogen that allows the states to build their programs of point source and nonpoint source controls upon in order to achieve the nutrient allocation. In effect, it takes some of the TMDL load burden off state point source and nonpoint source programs and places it on programs of national emission reductions in NO_x and the expectations of the future indirect atmospheric deposition reduction brought about by the Clean Air Act. As described by Birch *et al.* (2011), control of atmospheric deposition loads in eutrophic coastal watersheds has the advantage of efficiency in lower total cost when considering all ecosystems and human health because nitrogen reductions occur higher in the nitrogen cascade.

The reference allocation air load is tracked in the integrated Chesapeake Bay Program (CBP) models, specifically by the airshed and watershed models. If there are any reductions in the referenced air allocation brought about by additional national emission reductions, the additional reductions are passed on to the CBP state partners and their watershed point source or nonpoint source controls can be eased by amounts corresponding to the new reference air reductions, using the metric of delivered loads to the Bay.

METHODS

The CBP airshed model is a combination of a regression model of wet deposition (Grimm

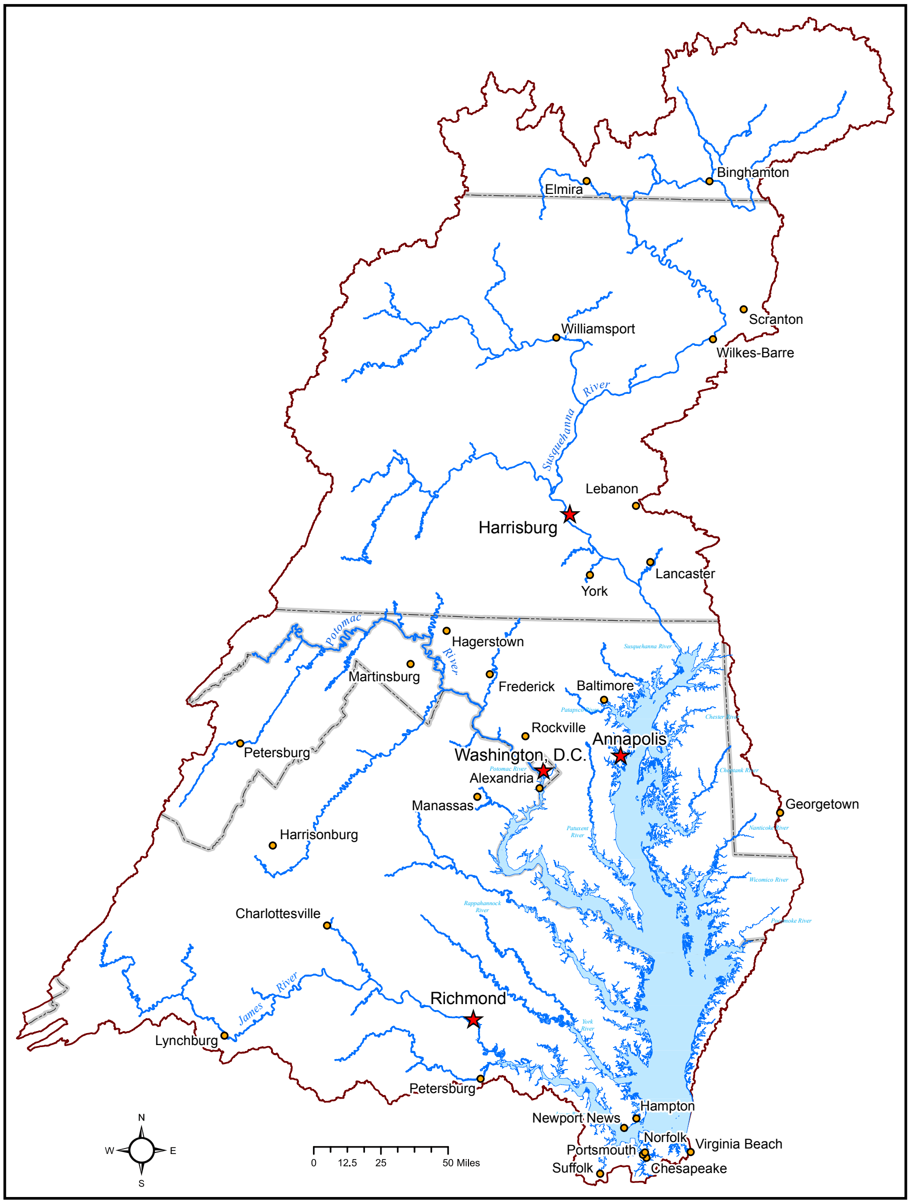

FIGURE 1. The Chesapeake Bay Watershed with the State Boundaries of New York, Pennsylvania, Maryland, West Virginia, Delaware, and Virginia Shown as well as the Major Rivers and Cities in the Watershed.

and Lynch, 2000, 2005) and for estimates of dry deposition, a continental scale Community Multiscale Air Quality (CMAQ) Model application, with North America as the model domain (Dennis *et al.*, 2007; Hameedi *et al.*, 2007). The regression and deterministic airshed models that provide atmospheric deposition input estimates, have gone through a series of refinements, with increasingly sophisticated models of both applied over time (Grimm and Lynch, 2000, 2005; Linker *et al.*, 2000; Lynch and Grimm, 2003). The amount and timing of the wet atmospheric deposition input in the Chesapeake watershed model is hourly because of the need to get high flow and nutrient loads in storms correctly simulated. The dry deposition estimates are monthly constants that are input daily and are based on CMAQ (Dennis *et al.*, 2007; Hameedi *et al.*, 2007). The airshed model tracks the nitrogen deposition load and the progress made toward reducing NO_x nitrogen deposition to reach the 7.1 million kg per annum allocation.

Regression Model of Wetfall Deposition

Wet deposition is simulated using a regression model developed by Grimm and Lynch (2000, 2005; Lynch and Grimm, 2003). The regression model provides hourly wet deposition loads to each land segment based on each land segment's rainfall. The land-segment grid is shown in Figure 2 and described in Shenk and Linker (this issue). The regression model uses single-event precipitation chemistry data from 29 National Atmospheric Deposition Program/National Trends Network (NADP/NTN) monitoring stations and 6 AIRMoN stations (which use the same methods as NADP, but collect single storm wet deposition events) to produce local estimates of wetfall inorganic nitrogen deposition across the entire Chesapeake watershed during the entire simulation period from 1985 to 2005 (Figure 2). The NADP/NTN and AIRMoN are specifically designed to measure ammonium and nitrate wet deposition.

To improve the accuracy of the regression estimates over previous regression analyses (Linker *et al.*, 2000), a number of improvements in the sampling and representation of spatial and temporal patterns of land use activities and intensities and of emission levels were made. Also, detailed meteorological data were assimilated into the regression model to identify contributing emission source areas and to estimate the impact of those contributions on daily deposition rates on a per-event basis.

This version of the regression model included 10 additional NADP/NTN sites in the regression estimates (DE99, MD07, MD08, MD15, MD99, PA47, VA10, VA27, VA98, and VA99) that were placed in operation in and around the Chesapeake Bay watershed since 2001. The sites provided a more complete representation of agricultural influences than the station set used in the earlier analyses.

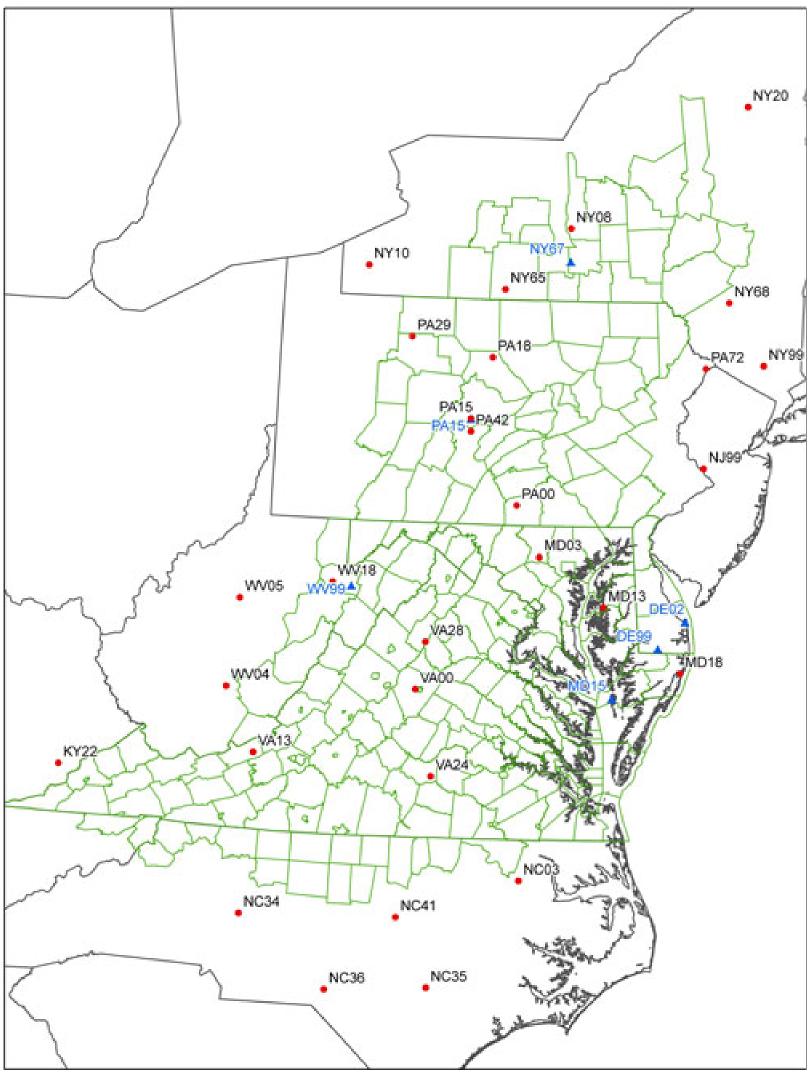

FIGURE 2. Locations of the 29 National Atmospheric Deposition Program/National Trends Network (circle) and 6 AIRMoN (triangle) Precipitation Chemistry Monitoring Sites Used for Development of the Wetfall Regression Model. Figure 2 also shows the land segments of the watershed model, which are the smallest spatial units of atmospheric deposition estimates used in the Chesapeake Total Maximum Daily Load (Shenk and Linker, this issue).

Refinements also involved developing a more accurate and comprehensive representation of the spatial and temporal distribution and intensity of livestock production and other agricultural activities across the Chesapeake watershed model domain. An improved accounting of livestock production activities was achieved by combining county- and watershed unit-specific livestock production statistics with high-resolution (30 m) land use data from the U.S. Geological Survey's National Land Cover Database (NLCD). Estimates of local ammonia emissions from fertilizers and manure applications to croplands were also assimilated into the model using USEPA inventories and high-resolution NLCD to quantify emissions from cropland areas likely to be fertilized, although there are significant uncertainties in the agricultural ammonia emission inventories. Last, localized estimates of NH_3 and NO_x emissions for the Chesapeake watershed model domain and surrounding states were developed by combining facility and county-specific emissions reports from USEPA's National Emissions Inventory database with the NLCD classifications (Grimm and Lynch, 2005).

For each day of precipitation, wetfall atmospheric deposition is estimated by the regression model, which has the general form:

$$\log 10(c) = b_0 + b_1 \log 10(\mathrm{ppt}) + b_{2\mathrm{s}}\ \mathrm{season} + b_3 v_3 + \ldots + b_{\mathrm{n}} v_{\mathrm{n}} + e$$

where c is the daily wetfall ionic concentration (mg/l), b_0 is the intercept, ppt is the daily precipitation volume (inches), b_1 is the coefficient for precipitation term, season is the vector of five binary indicator variables encoding the six bimonthly seasons, b_{2s} is the vector of five coefficients for season terms, and $v_3 \ldots v_n$ are additional predictors selected through stepwise regression of the following four terms:

1. NLCD.
2. Within proximities of 0.8, 1.6, 3.2, 8.0, and 16.1 km of each NADP/NTN site: open water, forested, residential, industrial/transportation, croplands, and vegetated wetlands.
3. Annual emission levels of ammonia and nitrous oxides from USEPA National Emission Trends for:
   - County containing each NADP/NTN monitoring site
   - Four counties nearest to each NADP/NTN monitoring site
4. Twelve-hour back-trajectory exposure of precipitating air-mass to ambient concentrations of transported ammonia and nitrous oxide emissions

$b_3 \ldots b_{\mathrm{n}}$ are coefficients corresponding to $v_3 \ldots v_n$ and e is residual error.

The most significant variables in both models included precipitation volume, the number of days since the last event, seasonality, latitude, and the proportion of land within 8 km covered by forests or devoted to transportation and industry (Grimm and Lynch, 2005). Local and regional ammonia and nitrogen oxides emissions were not as well correlated as land cover. The abilities of those variables to predict wet deposition arise primarily from their relationship with either (1) the spatial and temporal distribution of emissions of ammonium and nitrate precursors from sources within or upwind of the Chesapeake watershed model domain and (2) the chronology and characteristics of precipitation events. Modeled concentrations compared very well with event chemistry data collected at six NADP/AIRMoN sites in the Chesapeake watershed. Wet deposition estimates were also consistent with observed deposition at selected sites.

Volume, duration, and frequency of precipitation events have obvious roles in determining wet deposition rates. However, those parameters alone do not completely describe all the characteristics of a precipitation event. In particular, the intersection of a precipitation event and a volume of air with a particular *history* is estimated by the wet deposition model to have greater relevance to observations at a wet deposition monitor than local and regional emissions in determining wet deposition flux. For this reason, the interactions between storm trajectories and emission sources were incorporated into the model.

Using metrological data from the National Center for Environmental Prediction's North American Regional Reanalysis (NARR), variables were added to daily ammonium and nitrate wet deposition models that predict the rate at which emissions from area and point sources are emitted, dispersed, and transported to specific deposition locations. Surface and upper level vertical and horizontal air movement data from the NARR allowed estimates of the extent to which emissions were transported and mixed into surface and upper level atmospheric layers, and thereby enabled construction of more realistic multilevel air-mass trajectories with which to predict the movement of emissions from multiple source locations to deposition points of interest (Grimm and Lynch, 2000, 2005).

CMAQ Model

The CMAQ Model that was applied in this work was a fully developed, one-atmosphere air simulation model of the North American continent. The CMAQ

Model has more than 1,000 users worldwide and has been applied in many countries (Byun and Schere, 2006; Dennis *et al.*, 2007; Hameedi *et al.*, 2007). Byun and Schere (2006) reviewed the governing equations, computational algorithms, of the CMAQ modeling system, including the simulation approach for wet deposition. A description of the CMAQ dry deposition simulation can be found in Pleim and Ran (2011).

The CMAQ version used in this application was version 4.7.1 using MM5 model output with unidirectional ammonia simulation (Grell *et al.*, 1994). The mesoscale model MM5 is a terrain-following sigma-coordinate model designed to simulate mesoscale atmospheric circulation. CMAQ simulates deposition to the Chesapeake watershed (indirect deposition) and tidal Bay (direct deposition) for every hour of every day for the representative year. To calculate nitrogen deposition budgets CMAQ needs to be a one-atmosphere model incorporating: (1) photochemistry of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) to produce ozone and oxidized nitrogen products, (2) gas- and aqueous-phase oxidation of sulfur dioxide to create sulfuric acid, (3) particle thermodynamics and physics to treat ammonia neutralization of acids that partition the atmospheric species of nitrogen between gases (which rapidly deposit) and particles (which slowly deposit), and (4) cloud, wet scavenging and aqueous chemistry processes for wet deposition.

A variety of input files are needed that contain information pertaining to the modeling domain, which is the entire continental U.S., northern Mexico, and southern Canada. They include hourly emissions estimates and meteorological data in every grid cell as well as a set of pollutant concentrations to initialize the model and to specify concentrations along the modeling domain boundaries. The CMAQ grid cells in this application are generally 36-km grid in size across the U.S., but have a nested finer grid of 12 km covering the Chesapeake airshed and watershed. The initial and boundary concentrations were obtained from output of a global chemistry model, GEOS-Chem (Bey *et al.*, 2001).

The CMAQ Model simulation period is for one year, 2002, characterized as an average deposition year. The 2002 CMAQ simulation year was used to provide the monthly dry deposition estimate for all years of the 1985-2005 simulation period of the Chesapeake Bay watershed and tidal estuary models by adjusting the load for all years by assuming the dry deposition trend to be the same as the linear, long-term wet trend in the separate nitrate and ammonia estimates as described in more detail below. Dry deposition input estimates are derived from the CMAQ Model as monthly average inputs expressed as a daily load to watershed model land segments (USEPA, 1999; Shenk and Linker, this issue).

A 12-km grid was used to better resolve atmospheric deposition loads to the watershed and Bay (Figure 3). The improved spatial resolution of direct deposition loads to tidal waters as well as the deposition loads to the watershed adjacent to tidal waters from metropolitan and mobile sources was an important improvement (STAC, 2007) and allowed better tracking of the deposition fate of these emissions.

Organic Nitrogen Deposition

Organic nitrogen loads are a complex and significant source of nitrogen atmospheric deposition to the Chesapeake (Scudlark *et al.*, 1998; Cape *et al.*, 2011). Estimated loads of atmospheric organic nitrogen are to surface waters of the watershed and Bay only, because it is assumed that all organic nitrogen is derived from aeolian processes, which result in no net change in organic nitrogen on terrestrial surfaces, but do result in a net gain when deposited on water surfaces. Organic nitrogen atmospheric deposition loads are primarily represented as wetfall only, i.e., dissolved organic nitrogen (DON). The magnitude of dry fall organic nitrogen is less well characterized (Neff *et al.*, 2002). Organic nitrogen deposition loads are considered to be uncontrollable loads, which are unaltered by any Chesapeake management practices except in the limited case of peroxyacyl nitrates (PAN, $CH_3COOONO_2$) and an organic nitrate group in the CMAQ simulation involved in products of NO_x photochemistry as discussed below. Even though organic nitrogen loads are uncontrollable they are quantified as input to water surfaces because they contribute to the overall nitrogen load and eutrophication in the Chesapeake.

Wetfall Organic Nitrogen Deposition. Organic nitrogen measurements from Bermuda (Knap *et al.*, 1986) are calculated at about 100 μg/l (as N). Mopper and Zika (1987) reported an average DON concentration from the western Atlantic and Gulf of Mexico of about 100 μg/l (as N). That is consistent with the reported range from the North Sea and northeast Atlantic of 90-120 μg/l (Scudlark and Church, 1993). Scudlark *et al.* (1998) report an annual volume weighted average DON concentration in the Mid-Atlantic coastal areas to be about 130 μg/l (as N). Measurements in that study are consistent with the interannual variation (maximum in spring) reported by Smullen *et al.* (1982). A later study identified methodological problems with some of the previous studies and suggests the wet deposition of organic nitrogen in the Chesapeake watershed would be

FIGURE 3. A Portion of the 12-km Community Multiscale Air Quality Model Grid over the Chesapeake Bay Basin, and Also Showing Watershed Model Segments (Dennis *et al.*, 2007).

closer to 50 μg/l on an annual average basis (Keene *et al.*, 2002). That study also documents the highest concentrations of organic nitrogen in the spring.

The approach CBP has taken is to use 50 μg/l (as N) as representative of an average annual wet deposition concentration to the watershed and tidal waters with the seasonal loading pattern suggested by Smullen *et al.* (1982) and Scudlark *et al.* (1998). That applies an average concentration of 40 μg/l from July to March in rainfall and an average concentration of 80 μg/l from April to June. The load of organic nitrogen would depend on the precipitation, but assuming 100 cm of precipitation, the load would be on the order of 0.45 kg/ha-yr.

Dryfall Organic Nitrogen Deposition. Other than measurements of PAN, there are few measurements of dry deposition of organic N (Neff *et al.*, 2002). The CMAQ simulations used in the Chesapeake TMDL have updated chemical mechanisms that include peroxyacyl nitrates ($CH_3COOONO_2$) and an organic nitrate group (NTR) as products of NO_x photochemistry. The NTR represents several organic nitrates (such as alkyl nitrate) that are produced from ozone photochemistry. Both of these deposition loads are relatively small in magnitude, and both are biologically labile and available. Therefore, the dryfall PAN and NTR are lumped into the oxidized nitrogen atmospheric deposition dryfall inputs (Dennis *et al.*, 2007).

Organic and Inorganic Phosphorus Deposition

All of the atmospherically deposited nutrient loads onto the watershed and tidal Bay were quantified because the output of the Chesapeake airshed model was used as input to the watershed and estuarine models. The watershed and estuarine models tracked all nutrient input loads in an overall mass balance, and the calibration of the watershed and estuarine models was to observed nutrient species in the watershed streams and rivers, and in the tidal Bay, respectively. Organic and inorganic phosphorus deposition loads are considered to be uncontrollable, and are unaltered by any Chesapeake management practices, but because they contribute to the overall Chesapeake phosphorus loads and eutrophication they are quantified as inputs to water surfaces.

Estimated loads of atmospheric organic and inorganic phosphorus are accounted for as an input to surface waters of the watershed and tidal Bay on the assumption that, like organic nitrogen, the load is derived from aeolian processes, which result in no net change in organic nitrogen on terrestrial surfaces, but do result in a net gain when deposited on water surfaces. Following Smullen *et al.* (1982), loads of wetfall deposited organic and inorganic phosphorus are from constant concentrations of 47 and 16 μg/l, respectively, applied to the volume of precipitation at any simulated hour. Seasonally, those loads are treated in the same way as organic nitrogen, assuming that organic phosphorus will follow a pattern similar to organic nitrogen and that an aeolian source of inorganic phosphorus could well increase during the spring due to exposure and tillage of bare soil by agricultural practices. Accordingly, organic and inorganic phosphorus concentrations are set at 74 and 25 μg/l, respectively, from April to June, and at half those concentrations for the other nine months of the year.

Transport of Indirect Atmospheric Deposition Nutrient Loads to the Chesapeake

The Phase 5.3 Chesapeake Bay Watershed Model simulates the fate and transport of indirect watershed atmospheric deposition loads of nitrogen to the tidal Chesapeake (Linker *et al.*, 2000; Shenk and Linker, this issue). Phase 5.3 is an application of the Hydrologic Simulation Program-Fortran (Bicknell *et al.*, 2005). The segmentation scheme divides the Chesapeake Bay watershed into more than 1,000 segments/subbasins, with the average size about 166 km^2. About 280 monitoring stations throughout the Chesapeake Bay watershed were used for calibration of hydrology, whereas approximately 200 monitoring stations were used to calibrate water quality, depending on the constituent being calibrated.

The Bay Watershed Model simulates the 21-year period (1985-2005) on a 1-h time step (USEPA, 2010c). Nutrient input loads from atmospheric deposition are daily. Nutrient input loads from fertilizers and manures are based on an annual mass balance of U.S. Census of Agriculture estimated animal populations and crops, records of fertilizer sales, and other data sources, which are compiled and input to the model as an annual time series of discontinuous inputs based on the estimated timing of agricultural field operations of planting, crop maintenance, and harvest. Best Management Practices (BMPs) are incorporated on an annual time step and nutrient and sediment reduction efficiencies are varied by the size of storms (Shenk and Linker, this issue). Municipal and industrial wastewater treatment and discharging facilities and onsite wastewater treatment systems' nitrogen, phosphorus, and sediment contributions are also included in the Bay Watershed Model (USEPA, 2010d).

Combining the Regression Model of Wetfall Deposition and CMAQ — The Chesapeake Airshed Model

The airshed model is the combination of the regression model of wet deposition and CMAQ estimates of dry deposition. The 21-year time series of daily wet deposition atmospheric deposition loads were developed using the wet deposition regression model of daily inputs. The daily estimated wet deposition loads were input into the Phase 5.3 Chesapeake Bay Watershed Model into aliquots for each hour of precipitation forming a time series which provided the best loading estimates available on the hourly time step of the Phase 5.3 Model. The hourly input loads were particularly useful for simulating storm loads, which is important because high precipitation events are coincident with high nutrient loads. Getting the loading correct at an hourly time step provided the best calibration of nutrient species with observations.

The component of dry deposition in the airshed model was monthly estimates of the 2002 CMAQ average year and was adjusted for all years by assuming the dry deposition trend to be the same as the linear, long-term wet trend in the separate nitrate and ammonium estimates. This approach is based on a comparison of the seasonal trends in air concentrations, which drive dry deposition, in the Clean Air Status and Trends Network (CASTNET) data with the corresponding seasonal trends in wet deposition in NADP/NTN data. The CASTNET is a national air quality monitoring network designed to provide data to assess trends in air quality and atmospheric deposition and provides both wet and dry observations. The data show that the long-term trends are comparable, and that the interannual variability in air concentrations is small, less than 10%, even though the interannual variability in wet deposition can be more than 50%.

Combining the daily time series of wet deposition from the regression model and the monthly time series of dry deposition from CMAQ provided the means to generate atmospheric deposition nutrient loads to the Chesapeake watershed and tidal Bay consistent with the long-term trends, as well as the day to day variation in loads due to wet deposition.

Coastal Ocean Loads of Nitrogen Deposition

The CMAQ Model domain extending out into the Atlantic Ocean provides estimates of atmospheric deposition loads to the coastal ocean at the mouth of the Chesapeake Bay. Coastal ocean nutrient budgets have been made (Howarth *et al.*, 1995; Howarth, 1998; Fennel *et al.*, 2006). Howarth (1998) reported that atmospheric deposition loads are roughly equivalent to watershed loads in the northeast U.S. (Maine to Virginia) and estimated that the watershed inputs of nitrogen to the northeast coastal waters to be 0.27 teragrams. Inputs from direct atmospheric deposition to coastal waters were estimated to be 0.21 teragrams, and inputs from deep ocean upwelling estimated to be 1.54 teragrams, for a total input to the coastal ocean of 2.02 teragrams (Howarth, 1998).

The CMAQ estimates of atmospheric deposition to the coastal ocean region affecting nitrogen loads through the ocean boundary was determined by boundaries that correspond to the proximate region of the coastal ocean exchanging waters with the Chesapeake Bay. The boundary is adjacent to the Atlantic shore, and is inside, or west, of the Gulf Stream. To account for the prevailing north to south current along the coast, the coastal ocean boundary includes more of the coastal waters north of the Chesapeake Bay mouth (USEPA, 2010b).

Atmospheric deposition total nitrogen loads to the coastal ocean are estimated to be about 6.63 kg/ha in the CMAQ 2002 average year. That correlates to 43.8 million kg of total nitrogen deposition to a region of the ocean estimated to exchange waters with the Chesapeake. In the case of the 2020 Maximum Feasible scenario, the nitrogen atmospheric deposition to the same region is estimated to be 29.4 million kg, a reduction of 32%. If that same reduction is extrapolated to the coastal ocean, the direct atmospheric inputs to the coastal ocean would decrease to 0.14 teragram. Assuming the watershed loads discharged to the ocean and the deep upwelling pelagic loads are constant, that would give a combined watershed, direct deposition, and uncontrollable deep upwelling load of 1.95 teragrams, a decrease of 3% relative to the estimated current ocean boundary condition. This approach was used to estimate the relative change in ocean boundary conditions for the six key CMAQ scenarios (USEPA, 2010b).

The ocean loads of nitrogen are unquantified in the Chesapeake TMDL because they are mostly uncontrollable, but they are significant because they contribute to Chesapeake eutrophic conditions and account for about a quarter of the overall total nitrogen loads coming from all sources including the watershed and airshed (Thomann *et al.*, 1994).

RESULTS

Atmospheric loads of nitrogen are from chemical species of oxidized nitrogen, also called NO_Y and includes the mononitrogen oxides NO and NO_2 (NO_x) and compounds resulting from the oxidation of NO_x like nitric acid, and from reduced forms of nitrogen. Reduced atmospheric nitrogen is in the form of ammonia (NH_3) and ammonium (NH_4^+). Oxidized forms of nitrogen deposition originate from conditions of high heat and pressure and are formed from biologically inert diatomic atmospheric nitrogen (N_2). The principal sources of NO_x are industrially sized boilers, such as electric power plants (stationary sources), and the internal combustion engines in cars, trucks, locomotives, airplanes, and the like (mobile sources). Ammonia deposition originates from largely agricultural sources, predominately manures but also volatilization of ammonia from fertilizers. All nitrogen loads from oxidized and reduced nitrogen atmospheric deposition are estimated (using the CMAQ 36-km grid, see below for details) to be about 49% from sources in the watershed states and 51% from sources beyond the watershed (USEPA, 2010b).

Chesapeake Atmospheric Oxidized Inorganic Nitrogen Deposition Trends

Fertilizer and manure loads are estimated from the Agricultural Census at five-year intervals over the 1985-2005 simulation period in (USEPA, 2010e; Shenk and Linker, this issue) and are shown in Figure 4. Over the 1985-2005 simulation, average nitrogen atmospheric deposition levels to the Chesapeake watershed have been declining, particularly for oxidized nitrogen (Figure 5). The atmospheric deposition loads are among the highest sources of nitrogen loads in the watershed but also have the highest rate of reduction from 1985 to the present (Linker *et al.*, 2008).

During the 1985 and 2005 simulation period, wet atmospheric deposition loads of nitrate have tended

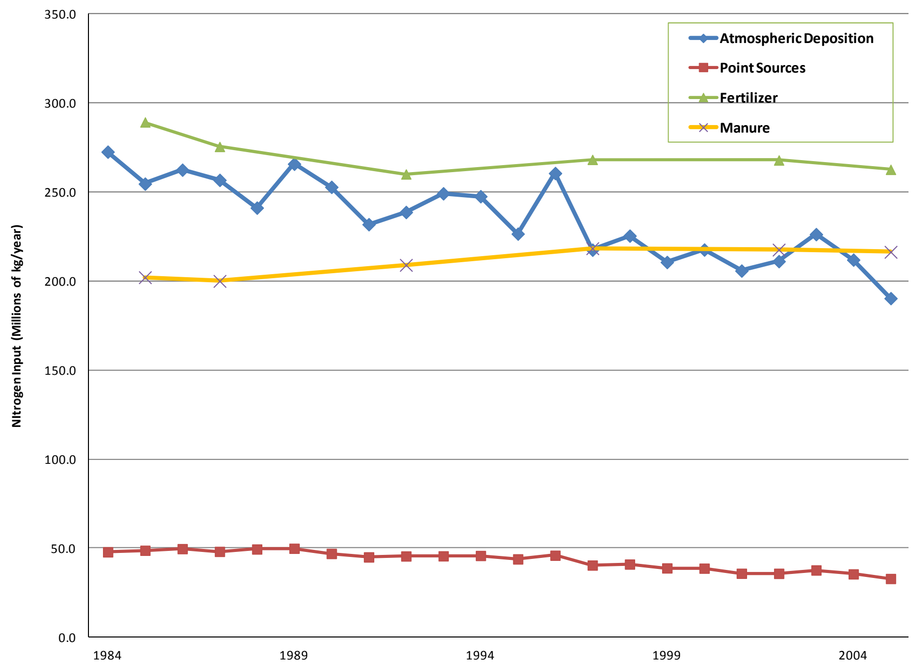

FIGURE 4. Time Series of Estimated Atmospheric, Fertilizer, Manure, and Point Source Total Nitrogen Input Loads to the Chesapeake Bay.

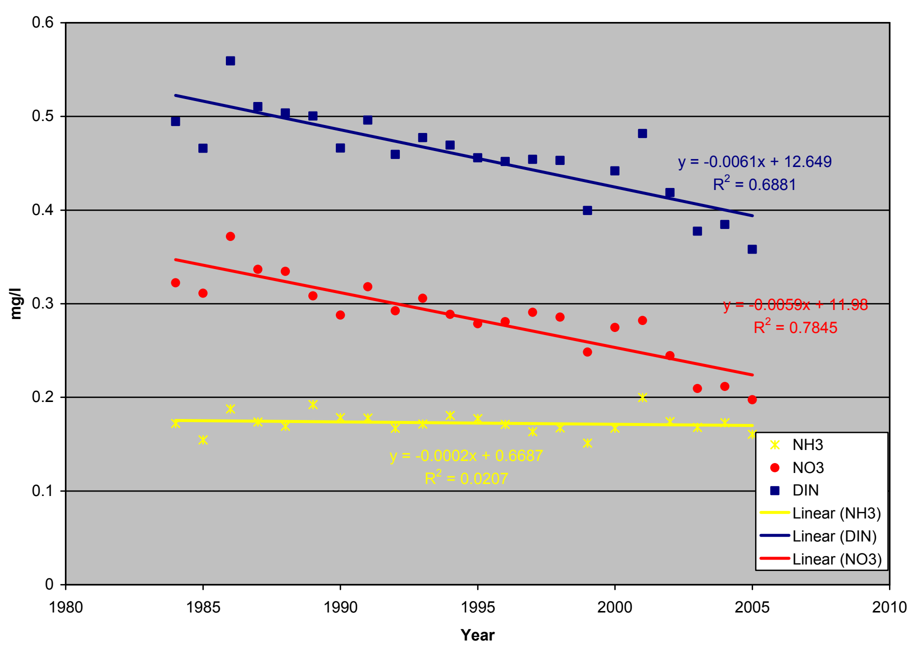

FIGURE 5. Trend of Estimated Average Nitrate and Ammonia Deposition Concentrations to the Chesapeake Watershed. Dissolved inorganic nitrogen (DIN) is the sum of nitrate and ammonia.

to decrease overall in the Chesapeake watershed. Over that 20-year period, wet deposition nitrate loads decreased by about 30% (Figure 5); however, there is considerable variability across the Chesapeake watershed with the greatest reductions occurring in the northern and western portions. The reductions are in part due to the constant downward regulatory pressure on electric generating units (EGUs) over the 1985-2005 period and the high concentration of EGUs upwind of the western and northern portions of the Chesapeake watershed as well as reductions in mobile sources.

In Figure 5, the average annual concentration of wetfall nitrate, ammonia, and dissolved inorganic nitrogen (DIN) is used as an adjustment to smooth out the effects of high and low rainfall years, which introduce substantial short-term variation in deposition load to the watershed primarily because of the volume of precipitation. Use of wetfall nitrate, ammonia, and DIN concentrations, rather than the mass of nitrogen deposition load, provides a reasonable estimate of the overall trend in atmospheric deposition.

Table 1 shows the estimated portion of deposited NO_x loads on the Chesapeake watershed from four sectors including EGUs, mobile sources, industry, and all other sources. Much of the NO_x reduction between 1990 and 2020 is estimated to be due to reductions in EGUs. In addition, both on-road and off-road mobile sources will have ongoing fleet turnover and replacement, which is putting cleaner spark and diesel engines in service, and that is expected to continue beyond 2030. Note that some sources like mobile sources seem to be increasing in percentage relative to other sources like EGUs. Mobile sources and "other" are actually decreasing, and the total estimate deposition load in 2020 is less than that in 1990; however, EGU emission reductions are relatively more than mobile reductions. Total deposited NO_x loads to the Chesapeake watershed are estimated to be 248 million kg in 1990 and 145 million kg in 2020. In 2020, direct deposition to tidal waters is estimated to be 3.5 million kg of NO_x or about 2% of the NO_x deposition load to the watershed.

The Chesapeake airshed is that area where an estimated 75% of the deposition loads to the Chesapeake watershed and Bay originate from. Close to 50% of the NO_x deposition to the Chesapeake watershed and tidal waters is estimated to be from air emission sources located in the six Chesapeake states of Virginia, Maryland, Pennsylvania, West Virginia, Delaware, and New York. The CMAQ Model estimates another 25% of NO_x deposition comes from an area adjacent and mostly west of the Chesapeake watershed which has an area of 1.48 million km^2 (USEPA, 2010c). This area is also called the Chesapeake airshed. The remaining 25% of NO_x deposition is from sources beyond the airshed. The ammonia airshed is similar to the NO_x airshed, but slightly smaller because of shorter range transport of ammonia.

TABLE 1. Estimated Portion of Deposited NO_x Loads on the Chesapeake Watershed from Four Sectors Including Electric Generating Units (EGUs), Mobile Sources, Industry, and All Other Sources in 1990 and 2020. Total annual deposited NO_x loads to the Chesapeake watershed are estimated to be 248 million kg in 1990 and 145 million kg in 2020.

	1990	2020 Preliminary
Power plants (EGUs)	40% (100)	17% (25)
Mobile sources (on-road)	30% (75)	32% (46)
Industry	8% (20)	20% (29)
Other (off-road-construction, residential, and commercial)	21% (53)	31% (45)
Total	(248)	(145)

Note: Units of percent and millions of kilograms in parentheses.

Chesapeake Atmospheric Reduced Inorganic Nitrogen Deposition Trends

Estimated annual average ammonia loads over the entire watershed and tidal Bay have no trend over the 1985-2005 simulation period (Figure 5: NH_3, x symbol). Ammonia deposition is relatively site specific and strongly influenced by local emissions. Local and regional trends in manure, such as, the rise of poultry animal units in the Eastern Shore and Shenandoah and dairy's diminishment in the northern portions of the watershed in the late 1980s, affect regional ammonia deposition in the Chesapeake watershed, leading to increases in ammonia deposition loads on the Eastern Shore and Shenandoah and decreases in the upper portions of the watershed over the 1985-2005 simulation period of the airshed model. The regional trends are not shown due to article size constraints.

CMAQ Scenarios

The CMAQ Model provides estimates of nitrogen deposition resulting from changes in emissions from utility, mobile, and industrial sources due to management actions or growth. For the CMAQ Model, the CMAQ 2002 average year is used, and scenarios include the management actions required by the Clean Air Act in 2010, 2020, and 2030. The future year scenarios reflect emissions reductions from national control programs for both stationary and mobile sources, including the following:

1. Clean Air Interstate Rule
2. Tier-2 Vehicle Rule
3. Non-road Engine Rule
4. Heavy-Duty Diesel Engine Rule
5. Locomotive/Marine Engine Rule

Application of the CMAQ Scenarios to the Watershed and Estuary Models. To develop a watershed and estuary model scenario using one of the CMAQ Model air scenarios described below, a monthly factor is determined from the CMAQ scenario by comparing the CMAQ scenario wet and dry, oxidized, and reduced, atmospheric deposition loads to the CMAQ 2002 Base year. The monthly ratio of the CMAQ scenario to the CMAQ 2002 Base year wet, dry, oxidized, and reduced deposition loads are then used to adjust the base atmospheric deposition loads of wet, dry, oxidized, and reduced deposition in the watershed and estuary models.

CMAQ 1985 and 2002 Scenarios. The CMAQ 1985 and 2002 scenarios represent estimated atmospheric deposition patterns and levels based on estimates of the wet deposition regression model for 1985 and 2002, respectively.

CMAQ 2010 Scenario. The 2010 Scenario represents emission reductions because of regulations implemented through the Clean Air Act authority to meet National Ambient Air Quality Standards (NAAQS) for criteria pollutants in 2010. That includes National/Regional and available State Implementation Plans (SIPs) for NO_x reductions. Other components of the 2010 Scenario include Tier 1 vehicle emission standards reaching high penetration in the vehicle fleet for on-road, light-duty mobile sources along with Tier 2 vehicle emission standards, which were fully phased in by the 2006 model year. For EGUs, the 2010 controls assume that the NO_x Budget Trading Program (including the NO_x SIP call) and the Clean Air Interstate Rule (CAIR) program that regulates the ozone season NO_x are all in place and that the CAIR program is designed for annual NO_x reductions to match the ozone season reductions under the 2010 CAIR first phase conditions (the scenario used the 1997 ozone standard of 80 ppb).

CMAQ 2020 Scenario — The Allocation Air Scenario. The 2020 Scenario was used to establish the estimated nitrogen deposition reductions from air controls in the Chesapeake TMDL. It has all components of the 2010 Scenario and includes the Clean Air Mercury Rule (CAMR), the Best Available Retrofit Technology (BART) used for reducing regional haze, and the off-road diesel and heavy-duty diesel regulations. The 2020 Scenario represents emission reductions due to regulations implemented through the Clean Air Act authority to meet NAAQS for criteria pollutants in 2020 (NAAQS 1997 8-h ozone NAAQS set at 80 ppb). Those include the following:

1. *On-Road Mobile Sources*: For on-road light-duty mobile sources, this includes Tier 2 vehicle emissions standards and the Gasoline Sulfur Program that affects sport utility vehicles, pickups, and vans, which are now subject to same national emission standards as cars.
2. *On-Road Heavy-Duty Diesel Rule — Tier 4*: New emission standards on diesel engines starting with the 2010 model year for NO_x, plus some diesel engine retrofits.
3. *Clean Air Non-Road Diesel Rule*: Off-road diesel engine vehicle rule, commercial marine diesels, and locomotive diesels (phased in by 2014) require controls on new engines.
4. *Off-Road Large Spark Ignition Engine Rules*: Rules that affect recreational vehicles (marine and land based in coordination with earlier NO_x SIP call and Title IV annual NO_x emissions program).
5. *EGUs*: CAIR second phase in place (in coordination with earlier NO_x SIP call); Regional Haze Rule and guidelines for BART for reducing regional haze; CAMR all in place.
6. *Non-EGUs*: Solid Waste Rules (Hospital/Medical Waste Incinerator Regulations).

CMAQ 2020 Maximum Feasible Scenario. The 2020 Maximum Feasible Scenario includes additional aggressive EGU, industry, and mobile source controls with emissions projections that represented incremental improvements and control options that might be available to states to meet a more stringent ozone standard of 70 ppb.

Incremental control measures for five sectors were developed:

1. *EGUs*: Lower ozone season nested emission caps in Ozone Transport Commission (OTC) states; targeting use of maximum controls for coal-fired power plants in or near nonattainment areas.
2. *Non-EGU Point Sources*: Include new supplemental controls, such as low NO_x burners, plus increased control measure efficiencies on planned controls and step up of controls to maximum efficiency measures, e.g., replacing Selective Non-Catalytic Reduction with Selective Catalytic Reduction control technology.
3. *Area (Nonpoint Area) Sources*: Widespread switching to natural gas and low sulfur fuel.
4. *On-Road Mobile Sources*: Increased penetration of diesel retrofits and continuous inspection and maintenance using remote onboard diagnostic systems.
5. *Non-Road Mobile Sources*: Increased penetration of diesel retrofits and engine rebuilds.
6. *Marine Vessels*: Reduced NO_x emissions from marine vessels in coastal shipping lanes.

The 2020 Maximum Feasible Scenario also includes a reduction in ammonia deposition of 15% from estimated ammonia emission programs in the Bay watershed jurisdictions. Estimates of up to about 30% ammonia emission reductions from manures can be achieved through rapid incorporation of manures into soils at the time of application, biofilters on poultry houses, and other management practices (Mark Dubin, 2009, personal communication). From a state and sector analysis of NO_x emissions and deposition, an estimated 50% of emissions from Bay states becomes deposition to the Chesapeake Bay watershed, along with a further 50% of the ammonia deposition load coming from outside the Bay watershed. Assuming that only 50% of the emissions are from watershed sources, a 30% reduction in emissions results in an estimated 15% decrease in wet and dry ammonia deposition for the Maximum Feasible Scenario from ammonia emission control management practices in the Bay watershed jurisdictions.

CMAQ 2030 Scenario. The 2030 scenario is, in some areas, a further decrease in emissions beyond the 2020 Maximum Feasible Scenario due to continuing fleet replacement of heavy diesels, off-road diesels, and mobile sources of all types**.** These emission decreases are offset by continued growth in the Chesapeake Bay region. The emissions projections assume continued stringent controls are in place, such as:

1. *Tier 2 Vehicle Emissions Standards*: Tier 2 fully penetrated in the fleet.
2. *Heavy-Duty Diesel Vehicle Fleet*: Fleet fully replaced with newer heavy-duty vehicles that comply with new standards.
3. *On-Road Mobile Sources*: Increased penetration of diesel retrofits maintained.
4. *Non-Road Mobile Sources*: Capped at 2020 Maximum Feasible Scenario levels.
5. *EGUs and Non-EGUs emissions*: Capped at 2020 Maximum Feasible Scenario levels.
6. *Area Sources*: Emissions capped at 2020 Maximum Feasible Scenario levels, assuming energy efficiency and control efficiencies keep up with growth.
7. *Marine Vessels*: Further reductions in NO_x emissions from marine vessels in coastal shipping lanes.

Scenario Results. In determining the allowable loading from air deposition, the nitrogen deposition was separated into two discrete parcels. The first is deposition occurring on the land and nontidal waters which is subsequently transported to the Bay, also called indirect deposition. The second is atmospheric deposition occurring directly onto the Bay's tidal surface waters, also called direct deposition. Tables 2 and 3 show the indirect delivered nitrogen loads to the Chesapeake Bay from the watershed and the direct delivered loads to the tidal Bay, respectively.

Annual nitrogen loads delivered to the Chesapeake Bay by jurisdiction for key CMAQ scenarios are tabulated in Table 2 in millions of kilograms. The estimated loads delivered to the tidal waters in Table 2 are the simulated airshed model deposition loads of nitrogen which are then input into the Phase 5.3 Watershed Model to track the fate and transport of nitrogen on the land and in the rivers before delivery to the Chesapeake (Linker *et al.*, 2008; Shenk and Linker, this issue). The Phase 5.3 Model simulates plant uptake of nitrogen, denitrification, and other attenuation of the deposition loads on an hourly time step (USEPA, 2010f, g).

To provide a common basis of comparison all the scenarios in Table 2 use the 2002 scenario as a base year in the Phase 5.3 Model. All nonatmospheric nitrogen loads, such as point sources, human and animal populations, and septic system loads in the Phase 5.3 Watershed Model are the same 2002 levels in all these scenarios; only the level of atmospheric deposition changes. The 1985 CMAQ scenario uses the trend of atmospheric deposition described in Figure 5, and the same trend was used for the 2002 atmospheric deposition in the 2002 scenario. The

TABLE 2. Total Nitrogen Delivered to the Bay Under Key Community Multiscale Air Quality (CMAQ) Atmospheric Deposition Scenarios Applied to a 2002 Base Condition of Land Use, Best Management Practices, and Point Source Discharges Showing the Relative Effect of Changing Only Atmospheric Deposition Loads to the Watershed on Total Loads from All Sources.

Basins	CMAQ 1985 Scenario	CMAQ 2002 Scenario	CMAQ 2010 Scenario	CMAQ 2020 Scenario	CMAQ 2020 Maximum Feasible Scenario	CMAQ 2030 Scenario
Susquehanna	72.8	67.2	64.1	62.9	62.4	63.2
West Shore	7.1	6.9	6.8	6.8	6.8	6.8
Potomac	34.9	32.7	31.5	31.0	30.8	31.1
Patuxent	2.2	2.0	2.0	2.0	2.0	2.0
Rappahannock	5.0	4.4	4.5	4.4	4.4	4.4
James	17.2	16.6	16.1	16.0	15.9	15.9
York	4.2	4.0	3.9	3.8	3.8	3.8
East Shore MD-DE	14.3	13.5	13.2	13.2	13.2	13.5
East Shore VA	1.4	1.3	1.3	1.3	1.3	1.3
Total	159.1	148.8	143.6	141.4	140.5	142.0

Note: Units in millions of kilograms per annum as N; Phase 5.2 — August 2009 Watershed Model Version.

TABLE 3. Direct Atmospheric Deposition Loads of Nitrogen (millions of kilograms per annum as nitrogen) to Chesapeake Bay's Tidal Surface Waters for Six Key Scenarios.

Scenario	Wet NO_x Deposition	Dry NO_x Deposition	Wet NH_3 Deposition	Dry NH_3 Deposition	Total Inorganic Nitrogen Deposition	Wet Organic Nitrogen Deposition	Total Nitrogen Deposition	Wet PO_4 Deposition	Wet Organic Phosphorus Deposition	Total Phosphorus Deposition
1985 Scenario	2.98	5.96	1.52	0.89	11.35	0.48	11.83	0.15	0.44	0.59
2002 Scenario	2.18	4.55	1.62	0.96	9.32	0.48	9.79	0.15	0.44	0.59
2010 Scenario	1.48	3.11	1.58	1.25	7.43	0.48	7.90	0.15	0.44	0.59
2020 Scenario	1.16	2.32	1.69	1.47	6.64	0.48	7.11	0.15	0.44	0.59
2020 Maximum Feasible Scenario	1.04	2.03	1.65	1.55	6.27	0.48	6.75	0.15	0.44	0.59
2030 Scenario	1.01	1.95	1.80	1.85	6.60	0.48	7.08	0.15	0.44	0.59

scenarios of 2010, 2020, 2020 Maximum Feasible, and 2030 used estimated atmospheric deposition loads from the CMAQ model. For the estimated Table 2 indirect loads from the watershed, only total nitrogen is tabulated because nitrogen transport and dynamics change the original nitrogen deposition speciation.

The regression and CMAQ models provide estimates of direct atmospheric deposition to the Bay's tidal surface waters. Table 3 lists the estimates of direct atmospheric deposition to the Bay's tidal surfaces for six key scenarios and tabulates the full speciation of nitrogen and phosphorus loads.

The Clean Air Act (CAA) regulations and rules as represented in the 2020 CMAQ Scenario, also called the Allocation Air Scenario, are assumed to be in place in 2020 reducing 11.8 million kg of nitrogen delivered to the Bay as compared to the model estimated 1985 conditions (scenarios to calculate this result are not shown in Table 2). The model estimated reduction in nitrogen directly deposited to Chesapeake tidal waters by the 2020 Allocation Air Scenario is 4.7 million kg compared to the 1985 model estimated conditions.

The largest reduction in total nitrogen deposition is from the 2020 Maximum Feasible Scenario which expands nitrate reductions from the 2020 Scenario as well as applying a 15% reduction in ammonia emissions and deposition. The 15% reduction in emission was assumed to be from improved management of ammonia emissions from manures. The 2030 Scenario has decreased nitrate deposition because of continuing fleet replacement of heavy diesels, off-road diesels, and of mobile sources of all types as compared to the 2020 Scenario. Nevertheless, the emission decreases in the 2030 Scenario are offset by continued growth in the Chesapeake region particularly in the increase in ammonia emissions compared to the 2020 Scenario estimates.

All of the scenarios in Tables 2 and 3, including the 2020 CMAQ Allocation Air, 2020 Maximum Feasible, and 2030 CMAQ scenarios are estimated to achieve widespread, but incomplete achievement of the NAAQS, and the 2020 and 2030 Scenarios have a few monitoring stations estimated to be in nonattainment for the 1997 ozone standard. More tailored SIPs now in development could address the few remaining areas of ozone standard nonachievement.

Chesapeake TMDL Air Allocation

Atmospheric deposition of nitrogen is a major source of nitrogen to the Chesapeake Bay watershed. For that reason, it was necessary to allocate an allowable loading of nitrogen from air deposition in the Chesapeake Bay TMDL. In determining the amount of air controls to be used as a basis for the Bay TMDL air allocation, the projection of the current laws and regulations under the Clean Air Act to 2020 was used and the 2020 Scenario was chosen by the by the CBP as the Allocation Air Scenario.

The 2020 CMAQ Scenario, or the Allocation Air Scenario, was used to set the TMDL air allocation in the tidal Bay. The Allocation Air Scenario was also used to develop the expected future reductions in atmospheric deposition loads to the watershed. Reduced atmospheric deposition loads to the watershed, and subsequently delivered to the Bay, are load reductions tracked in the CBP CMAQ and watershed models. If there are any national, regional, or state reductions beyond the 2020 Allocation Air Scenario, they can be quantified in future versions of the 2020 Allocation Air Scenario so that as these new air deposition reductions are implemented nitrogen reductions in the state Watershed Implementation Plans (WIPs) become more easily attained, and the WIP point or nonpoint source controls can be eased by the same amount that the loads fall below the 2020 Allocation Scenario (as delivered nitrogen loads to the Bay).

Methods have been developed and supported by model simulations to allow exchanges between air controls and more traditional point and nonpoint source watershed management controls of nitrogen which will be described in a subsequent article.

SUMMARY AND CONCLUSIONS

TMDL Allocation of Atmospheric Deposition of Nitrogen to the Watershed

The integrated models allow tracking of the transport and fate of nitrogen air emissions, including deposition in the Chesapeake watershed, the subsequent uptake, transformation, and transport to Bay tidal waters, and their ultimate influence on Chesapeake water quality. Key findings are that the atmospheric deposition loads are among the largest input loads of nitrogen in the watershed and that the indirect nitrogen deposition loads to the watershed, which are subsequently delivered to the Bay, are larger than the direct loads of atmospheric nitrogen deposition to Chesapeake tidal waters. The deposition on the land becomes part of the allocated load to the jurisdictions because the atmospheric deposition on the land becomes mixed with the nitrogen loadings from land-based sources and, therefore, becomes indistinguishable from land-based sources. Furthermore, once the nitrogen is deposited on the land, it would be managed and controlled along with other sources of nitrogen that are present on that parcel of land.

The CAA regulations and rules as represented in the 2020 CMAQ Scenario, also called the Allocation Air Scenario, are assumed to be in place in 2020 reducing 11.8 million kg of annual total nitrogen load delivered to the Bay from the watershed as compared to the model estimated 1985 conditions. This allows the Bay watershed jurisdictions finalize and implement their WIPs to reduce nitrogen loads further with land-based BMPs in order to achieve the allocation loads.

TMDL Allocation of Atmospheric Deposition of Nitrogen to the Tidal Waters

Nitrogen deposition directly to the Bay's tidal surface waters is a direct loading with no land-based management controls and, therefore, is linked directly back to the air emission sources and air emission controls as USEPA's allocation of atmospheric nitrogen deposition.

In determining the amount of emission controls to be used as a basis for the air allocation, USEPA relied on current laws and regulations under the Clean Air Act. These requirements, together with national air modeling analysis, provided the resulting allocated load to air from direct deposition to the tidal waters of the Bay and its tidal tributaries. The model estimated reduction in nitrogen directly deposited to Chesapeake tidal waters by the 2020 Allocation Air Scenario is 4.7 million kg compared to the 1985 model estimated conditions.

The 2010 Chesapeake TMDL included an explicit nitrogen allocation, which was determined to be 7.1 million kg per annum of total nitrogen atmospheric deposition loads direct to Chesapeake Bay and tidal tributary surface waters and was based on the 2020 CMAQ Scenario, also called the Air Allocation Scenario. The loading cap of 7.1 million kg of direct atmospheric deposition to Chesapeake Bay and tidal tributary surface waters will be achieved through the Clean Air Act authority to meet NAAQS for criteria pollutants in 2020. Projected reductions in atmospheric deposition loads to the surrounding watershed over this same period are already accounted for within the individual jurisdiction and major river basin nitrogen load allocations. Any additional nitrogen reductions realized through more stringent air pollution controls at the jurisdiction level, beyond minimum federal requirements to meet air quality standards may be credited to the individual jurisdictions through future revisions to the jurisdictions' WIPs, two-year milestones, and the Bay TMDL tracking and accounting framework.

The atmospheric deposition loads are among the largest input loads of nitrogen in the watershed. The indirect nitrogen deposition loads to the watershed are larger than the direct loads of atmospheric nitrogen deposition to Chesapeake tidal waters by about a factor of 20, however, most of the watershed atmospheric nitrogen deposition loads are attenuated by plant uptake, denitrification, and other loss mechanisms. About half the atmospheric deposition loads of nitrogen originate from outside the Chesapeake watershed. The Maximum Feasible and 2030 scenarios demonstrate that additional reductions in atmospheric deposition of nitrogen are possible, either by controlling manure ammonia emissions, as in the Maximum Feasible Scenario, or in increased fleet penetration of mobile reductions, as in the 2030 Scenario.

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LITERATURE CITED

Bey, I., D.J. Jacob, R.M. Yantosca, J.A. Logan, B.D. Field, A.M. Fiore, Q. Li, H.Y. Liu, L.J. Mickley, and M.J. Schultz, 2001. Global Modeling of Tropospheric Chemistry with Assimilated Meteorology: Model Description and Evaluation. Journal of Geophysical Research 106:23073-23096.

Bicknell, B.R., J.C. Imhoff, J.L. Kittle, Jr., T.H. Jobes, and A.S. Donigian, Jr., 2005. Hydrological Simulation Program—FORTRAN. User's Manual for Release 12.2. U.S. Environmental Protection Agency Ecosystem Research Division/U.S. Geological Survey, Office of Surface Water, Athens, Georgia/Reston, Virginia.

Birch, M.B.L., B.M. Gramig, W.R. Moomaw, O.C. Doering, III, and C.J. Reeling, 2011. Why Metrics Matter: Evaluating Policy Choices for Reactive Nitrogen in the Chesapeake Bay Watershed. Environmental Science and Technology 45(1):168-174.

Byun, D. and K.L. Schere, 2006. Review of the Governing Equation, Computational Algorithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. Applied Mechanics Reviews 59(March):51-77.

Cape, J.N., S.E. Cornwell, T.D. Jickells, and E. Nemitz, 2011. Organic Nitrogen in the Atmosphere – Where Does It Come from? A Review of Sources and Methods. Atmospheric Research 102(2011):30-48.

Cerco, C.F., S.C. Kim, and M.R. Noel, 2010. The 2010 Chesapeake Bay Eutrophication Model. A Report to the US Environmental Protection Agency and to the US Army Corps of Engineer Baltimore District. US Army Engineer Research and Development Center, Vicksburg, Mississippi. http://www.chesapeakebay.net/publications/title/the_2010_chesapeake_bay_eutrophication_model1, *accessed* March 2012.

Cerco, C.F. and M.R. Noel, this issue. Twenty-One-Year Simulation of Chesapeake Bay Water Quality Using the CE-QUAL-ICM Eutrophication Model. Journal of the American Water Resources Association, doi: 10.1111/jawr.12107. EPA-903-R-13-011, CBP/TRS-317-13-10.

Dennis, R.L., R. Haeuber, T. Blett, J. Cosby, C. Driscoll, J. Sickles, and J.M. Johnston, 2007. Sulfur and Nitrogen Deposition on Ecosystems in the United States. Environmental Management December:12-17.

Fennel, K., J. Wilkin, J. Levin, J. Moisan, J. O'Reilly, and D. Haidvogel, 2006. Nitrogen Cycling in the Middle Atlantic Bight: Results from a Three Dimensional Model for the North Atlantic Nitrogen Budget. Global Biogeochemical Cycles 20(GB3007):14, doi: 10.1029/2005GB002456.

Fisher, D., J. Ceraso, T. Mathew, and M. Oppenheimer, 1988. Polluted Coastal Waters: The Role of Acid Rain. Environmental Defense Fund, New York City, New York.

Fisher, D.C. and M. Oppenheimer, 1991. Atmospheric Nitrogen Deposition and the Chesapeake Bay Estuary. Ambio 20:102-108.

Grell, G.A., J. Dudhia, and D.R. Stauffer, 1994. A Description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5). NCAR Tech. Note NCAR/TN-398+STR (Available from the National Center for Atmospheric Research, P. O. Box 3000, Boulder, CO 549 80307). http://www.mmm.ucar.edu/mm5/mm5-home.html, *accessed* January 2013.

Grimm, J.W. and J.A. Lynch, 2000. Enhanced Wet Deposition Estimates for the Chesapeake Bay Watershed Using Modeled Precipitation Inputs. DNR Chesapeake Bay and Tidewater Programs CBWP-MANTA-AD-99-2.

Grimm, J.W. and J.A. Lynch, 2005. Improved Daily Precipitation Nitrate and Ammonium Concentration Models for the Chesapeake Bay Watershed. Environmental Pollution 135(2005):445-455.

Hameedi, J., H. Paerl, M. Kennish, and D. Whitall, 2007. Nitrogen Deposition in U.S. Coastal Bays and Estuaries. Environmental Managment December:19-25.

Hinga, K.R., A.A. Keller, and C.A. Oviatt, 1991. Atmospheric Deposition and Nitrogen Inputs to Coastal Waters. Ambio 20(6):256-260.

Howarth, R.W., 1998. An Assessment of Human Influences on Fluxes of Nitrogen from the Terrestrial Landscape to the Estuaries and Continental Shelves of the North Atlantic Ocean. Nutrient Cycling in Agroecosystems 52:213-223.

Howarth, R.W., G. Billen, D. Swaney, A. Townsend, N. Jaworski, K. Lajtha, J.A. Downing, E.R. Elmgren, N. Caraco, T. Jordan, F. Berendse, J. Freney, V. Kudeyarov, P. Murdoch, H. Zhaoliang, and H. Zhu, 1995. Regional Nitrogen Budgets and Riverine N & P Fluxes for the Drainages to the North Atlantic Ocean: Natural and Human Influences. Biogeochemistry 35(1):75-139.

Keene, W.C., J.A. Montag, J.R. Maben, M. Southwell, J. Leonard, T.M. Church, J.L. Moody, and J.N. Galloway, 2002. Organic Nitrogen in Precipitation over Eastern North America. Atmospheric Environment 36(28):4529-4540.

Knap, A., T. Jickells, A. Pszenny, and J.N. Galloway, 1986. Significance of Atmospheric-Derived Fixed Nitrogen on Productivity of the Sargasso Sea. Nature 320:158-160.

Linker, L.C., G.W. Shenk, R.L. Dennis, and J.S. Sweeney, 2000. Cross-Media Models of the Chesapeake Bay Watershed and Airshed. Water Quality and Ecosystem Modeling 1(1-4):91-122.

Linker, L.C., G.W. Shenk, P. Wang, and R. Batiuk, 2008. Integration of Modeling, Research, and Monitoring in the Chesapeake Bay Program. *In*: Management of Water Quality and Irrigation Techniques, J. Albiac and A. Dinar (Editors). Earthscan, London, United Kingdom, pp. 41-60.

Lynch, J.A. and J.W. Grimm, 2003. Improved Daily Nitrate and Ammonium Concentration Models for the Chesapeake Bay Watershed. U.S. Environmental Protection Agency, Chesapeake Bay Program Office, Annapolis, Maryland.

Mopper, K. and R.G. Zika, 1987. Free Amino Acids in Marine Rains: Evidence for Oxidation and Potential Role in Nitrogen Cycling. Nature 325(15):246-249.

Neff, J.C., E.A. Holland, F.J. Dentener, W.H. Mcdowell, and K.M. Russell, 2002. The Origin, Composition and Rates of Organic Nitrogen Deposition: A Missing Piece of the Nitrogen Cycle? Biogeochemistry 57/58:99-136.

Paerl, H.W., R.L. Dennis, and D.R. Whitall, 2002. Atmospheric Deposition of Nitrogen: Implications for Nutrient Over-Enrichment of Coastal Waters. Estuaries 25(4B):677-693.

Pleim, J. and L. Ran, 2011. Surface Flux Modeling for Air Quality Applications. Atmosphere 2(3):271-302.

Scudlark, J.R. and T.M. Church, 1993. Atmospheric Input of Inorganic Nitrogen to Delaware Bay. Estuaries 16(4):747-754.

Scudlark, J.R., K.M. Russel, J.N. Galloway, T.M. Church, and W.C. Keene, 1998. Organic Nitrogen in Precipitation at the Mid-Atlantic U.S. Coast – Methods Evaluation and Preliminary Measurements. Atmospheric Environment 32(10):1719-1728.

Shenk, G.W. and L.C. Linker, this issue. Development and Application of the 2010 Chesapeake Bay Watershed Total Maximum Daily Load Model. Journal of the American Water Resources Association, doi: 10.1111/jawr.12109. EPA-903-R-13-004, CBP/TRS-310-13-3.

Smullen, J.T., J.L. Taft, and J. Macknis, 1982. Nutrient and Sediment Loads to the Tidal Chesapeake Bay System. *In*: U.S. Environmental Protection Agency Chesapeake Bay Program Technical Studies: A Synthesis, E. Giles Macalaster, D. Allender Barker, M. Kasper (Editors). Chesapeake Bay Program Office, Annapolis, Maryland, pp. 147-259.

STAC (Chesapeake Bay Program Scientific and Technical Advisory Committee), 2007. Workshop on Atmospheric Deposition of Nitrogen: Estimating Local Emission Sources, Near-Field Deposition, and Fate on the Landscape. May 30, 2007. SUNY Binghamton, Binghamton, New York. http://www.chesapeake.org/stac/AtmosphericNDepositionWorkshop.html, *accessed* July 2008.

Thomann, R.V., J.R. Collier, A. Butt, E. Casman, and L.C. Linker, 1994. Response of the Chesapeake Bay Water Quality Model to Loading Scenarios. Chesapeake Bay Program Technology Transfer Report, CBP/TRS 101/94. Annapolis, Maryland.

Tyler, M., 1988. Contribution of Atmospheric Nitrate Deposition to Nitrate Loading in the Chesapeake Bay. Report RP1052, VERSAR Inc. Maryland Department of Natural Resources, Annapolis, Maryland.

USEPA (U.S. Environmental Protection Agency), 1999. Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. EPA/600/R-99/030. http://www.epa.gov/asmdnerl/CMAQ/CMAQscienceDoc.html, *accessed* December 2010.

USEPA (U.S. Environmental Protection Agency), 2003a. Ambient Water Quality Criteria for Dissolved Oxygen, Water Clarity and Chlorophyll *a* for the Chesapeake Bay and Its Tidal Tributaries. EPA 903-R-03-002. U.S. Environmental Protection Agency, Region 3, Chesapeake Bay Program Office, Annapolis, Maryland.

USEPA (U.S. Environmental Protection Agency), 2003b. Biological Evaluation for the Issuance of Ambient Water Quality Criteria for Dissolved Oxygen, Water Clarity and Chlorophyll *a* for the Chesapeake Bay and Its Tidal Tributaries. U.S. Environmental Protection Agency, Region 3, Chesapeake Bay Program Office, Annapolis, Maryland.

USEPA (U.S. Environmental Protection Agency), 2003c. Setting and Allocating the Chesapeake Bay Basin Nutrient and Sediment Loads, the Collaborative Process, Technical Tools and Innovative Approaches. U.S. Environmental Protection Agency, Region 3, Chesapeake Bay Program Office, Annapolis, Maryland.

USEPA (U.S. Environmental Protection Agency), 2004. Technical Support Document for Identification of Chesapeake Bay Designated Uses and Attainability – 2004 Addendum. EPA 903-R-04-006. U.S. Environmental Protection Agency, Region 3, Chesapeake Bay Program Office, Annapolis, Maryland.

USEPA (U.S. Environmental Protection Agency), 2007. Ambient Water Quality Criteria for Dissolved Oxygen, Water Clarity and Chlorophyll *a* for the Chesapeake Bay and Its Tidal Tributaries. 2007 Chlorophyll Criteria Addendum. EPA 903-R-07-005 CBP/TRS 288/07. U.S. Environmental Protection Agency, Region 3, Chesapeake Bay Program Office, Annapolis, Maryland.

USEPA (U.S. Environmental Protection Agency), 2010a. Ambient Water Quality Criteria for Dissolved Oxygen, Water Clarity and Chlorophyll *a* for the Chesapeake Bay and Its Tidal Tributaries: 2010 Technical Support for Criteria Assessment Protocols Addendum. May 2010. EPA 903-R-10-002. CBP/TRS 301-10. U.S. Environmental Protection Agency, Region 3, Chesapeake Bay Program Office, Annapolis, Maryland.

USEPA (U.S. Environmental Protection Agency), 2010b. Chesapeake Bay Total Maximum Daily Load for Nitrogen, Phosphorus and Sediment Appendix L: Setting the Chesapeake Bay Atmospheric Nitrogen Deposition Allocations. U.S. Environmental Protection Agency Chesapeake Bay Program Office, Annapolis, Maryland. http://www.epa.gov/reg3wapd/pdf/pdf_chesbay/FinalBayTMDL/AppendixLAtmosNDepositionAllocations_final.pdf, *accessed* January 2012.

USEPA (U.S. Environmental Protection Agency), 2010c. Chesapeake Bay Phase 5.3 Community Watershed Model: Section 1. EPA 903S10002 — CBP/TRS-303-10. U.S. Environmental Protection Agency, Chesapeake Bay Program Office, Annapolis, Maryland. December 2010. ftp://ftp.chesapeakebay.net/modeling/P5Documentation/SECTION_1.pdf, *accessed* August 2012.

USEPA (U.S. Environmental Protection Agency), 2010d. Chesapeake Bay Phase 5.3 Community Watershed Model: Section 7. EPA 903S10002 — CBP/TRS-303-10. U.S. Environmental Protection Agency, Chesapeake Bay Program Office, Annapolis, Maryland. December 2010. ftp://ftp.chesapeakebay.net/modeling/P5Documentation/SECTION_7.pdf, *accessed* August 2012.

USEPA (U.S. Environmental Protection Agency), 2010e. Estimates of County-Level Nitrogen and Phosphorus Data for Use in Modeling Pollutant Reduction: Documentation for Scenario Builder Version 2.2. CBP/TRS 903R100004 Bin # 304, U.S. Environmental Protection Agency Chesapeake Bay Program Office, Annapolis, Maryland. http://archive.chesapeakebay.net/pubs/sb_documentation_final_v22_9_16_2010.pdf, *accessed* December 2010.

USEPA (U.S. Environmental Protection Agency), 2010f. Chesapeake Bay Phase 5.3 Community Watershed Model: Section 10 Nonpoint Source Nutrient Simulation. EPA 903S10002 — CBP/TRS-303-10. U.S. Environmental Protection Agency, Chesapeake Bay Program Office, Annapolis, Maryland. December 2010. ftp://ftp.chesapeakebay.net/Modeling/P5Documentation/SECTION_7.pdf, *accessed* March 2012.

USEPA (U.S. Environmental Protection Agency), 2010g. Chesapeake Bay Phase 5.3 Community Watershed Model: Section 11 Simulation and Calibration of Riverine Fate and Transport of Nutriets and Sediment. EPA 903S10002 — CBP/TRS-303-10. U.S. Environmental Protection Agency, Chesapeake Bay Program Office, Annapolis, Maryland. December 2010. ftp://ftp.chesapeakebay.net/Modeling/P5Documentation/SECTION_7.pdf, *accessed* March 2012.

USEPA (U.S. Environmental Protection Agency), 2010h. Chesapeake Bay Total Maximum Daily Load for Nitrogen, Phosphorus and Sediment Appendix A. Chesapeake Bay TMDL Contributors. U.S. Environmental Protection Agency Chesapeake Bay Program Office, Annapolis, Maryland. http://www.epa.gov/reg3wapd/pdf/pdf_chesbay/FinalBayTMDL/AppendixAContributors_approved_final.pdf, *accessed* August 2012.