3 DEPOSITION

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3.1 OVERVIEW

To determine the effects of air pollution on ecological systems using the critical load approach, accurate estimates of total nitrogen (N) deposition are essential. Empirical critical loads are set by relating observed ecosystem responses to N deposition (measured, experimentally manipulated, or modeled). Because many manipulation studies are done under natural conditions where the total N deposition is not known, the empirical critical load is often crudely estimated. However, making accurate measurements or models of N deposition is difficult, and such estimates are not available at the fine spatial resolution required for determining empirical critical loads.

The purposes of this chapter are threefold. First, it presents an overview of atmospheric deposition to ecosystems and provides a brief summary of the processes and challenges of measuring and estimating total (wet, dry, cloud, or fog) atmospheric deposition. Second, we present a map (Fig. 3.1) of wet plus dry deposition for the more than 3200 U.S. locations for which we report ecological responses to N deposition in subsequent chapters. Third, we compare two methods of estimating deposition at the national scale (Weathers and Lynch 2008).

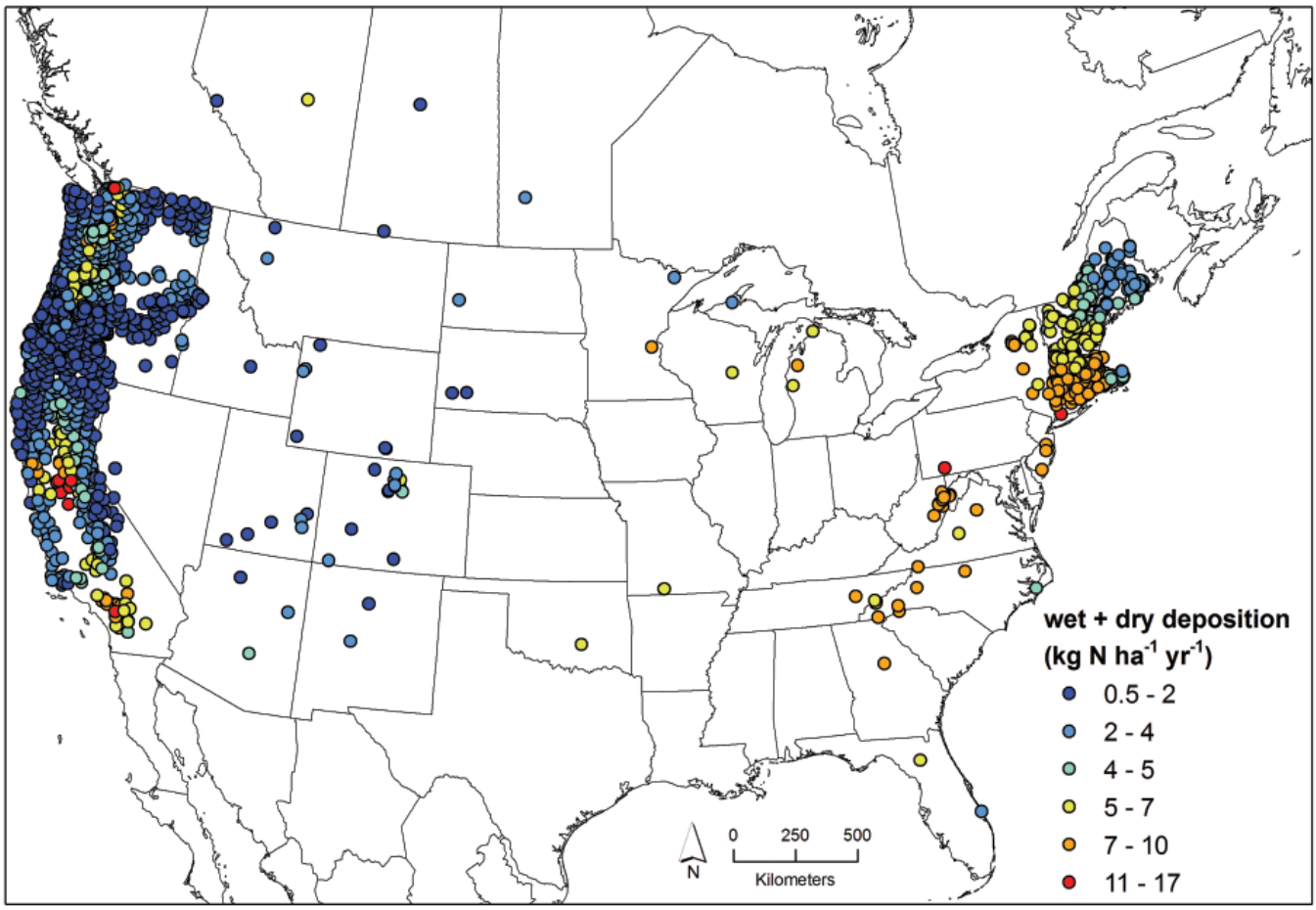

Figure 3.1—Estimates of wet plus dry N deposition (including, as N, wet ammonium (NH_4^+) and nitrate (NO_3^-), and dry nitric acid (HNO_3), ammonia (NH_3), NH_4^+, and NO_3^-) generated by the CMAQ 2001 model for more than 3200 locations across the United States for which we report ecological responses to N deposition in subsequent chapters.

3.2 Background on total N deposition

The primary forms of inorganic N deposition that have been measured routinely are secondary products of nitrogen oxide (NO_x) emissions—nitrate (NO_3^-; particulate and aqueous) and nitric acid vapor (HNO_3) —and ammonium (NH_4^+; particulate and aqueous). The primary anthropogenic source of NO_x is fossil fuel combustion. Transportation accounted for more than 50 percent of the total NO_x emissions in 2002 (Driscoll et al. 2003, Weathers et al. 2007), while the primary sources of ammonia (NH_3) and NH_4^+ are agricultural activities (Driscoll et al. 2003, Weathers et al. 2007). (Chemical formulas and common names for elements and compounds are provided in Appendix 2.)

Nitrogen is deposited from the atmosphere to the Earth's surface in the form of wet (rain and snow), dry (gases and particles), and cloud and/or fog deposition (Fenn et al. 2009; Lovett 1994; Weathers et al. 1988, 2000a, 2000b, 2006). Wet N deposition can be measured directly, but often only some of the components are measured. Dry deposition cannot be measured directly. Moreover, the estimates that exist are either site specific, or are modeled (e.g., dry deposition) assuming flat, homogeneous terrain, e.g. Clean Air Status and Trends (CASTNET) sites (US EPA 2010). For this reason, a combination of monitoring data, deposition models (e.g., the Multi-Layer Model [MLM] for estimating dry deposition), empirical measures of atmospheric deposition (e.g., throughfall), and emissions-based modeling outputs, are often necessary to estimate total N deposition. In the ecoregion chapters that follow, we consider total deposition to include wet (rain and snow) plus dry and, in some cases, cloud/fog.

There are several factors that reduce the accuracy of total N deposition estimates:

- Not all forms (wet, dry, and cloud) of atmospheric deposition are measured or modeled routinely. Thus, estimates that are based on wet deposition only, for example, result in an underestimation of the total N delivered to an ecosystem (Lovett 1994, Weathers et al. 2007). Dry and cloud N deposition can contribute from 50 to 80 percent of total N deposition (Johnson and Lindberg 1993); these forms of deposition are not often estimated.
- Not all of the chemical species of N that are deposited to ecosystems (e.g., NH_3 and organic N) are currently measured and/or modeled, which also leads to an underestimate of N deposition.
- Estimates of dry and cloud deposition depend upon measured air chemistry and modeled deposition. These estimates are therefore likely to be imprecise. For example, the influences of local meteorology, complex terrain (elevation and slope), as well as canopy architecture all affect rates of dry deposition, yet these characteristics of landscapes and canopies are extremely difficult to represent in models, and thus represent serious limitations to accurate estimates of dry N deposition (Lovett 1994; Weathers et al. 1992, 1995, 2000a, 2000b, 2006). Total deposition is therefore often underestimated for high elevation regions and/or for complex terrain (Anderson et al. 1999; Johnson and Lindberg 1993; Weathers et al. 1992, 2000a, 2000b, 2006).
- Estimates of deposition are also highly uncertain for regions with high snowfall, such as the montane west, and regions with a high proportion of dry deposition, such as the arid south and west (see Fenn et al. 2009 for a complete description of issues and potential solutions for these regions).
- Finally, inadequate number and spatial extent of monitoring stations is a crucial issue that adversely impacts deposition estimates across space, particularly in complex terrain. This is a significant problem in the western United States (Fenn et al. 2009), where few monitoring stations exist.

Inorganic species of N most commonly measured or modeled in the field (Fig. 3.2) include: NO_3^-, NH_4^+, and nitric acid (HNO_3). In recent years, dissolved organic nitrogen (DON) has also been shown to be prevalent in precipitation (e.g., Neff et al. 2002) and in clouds

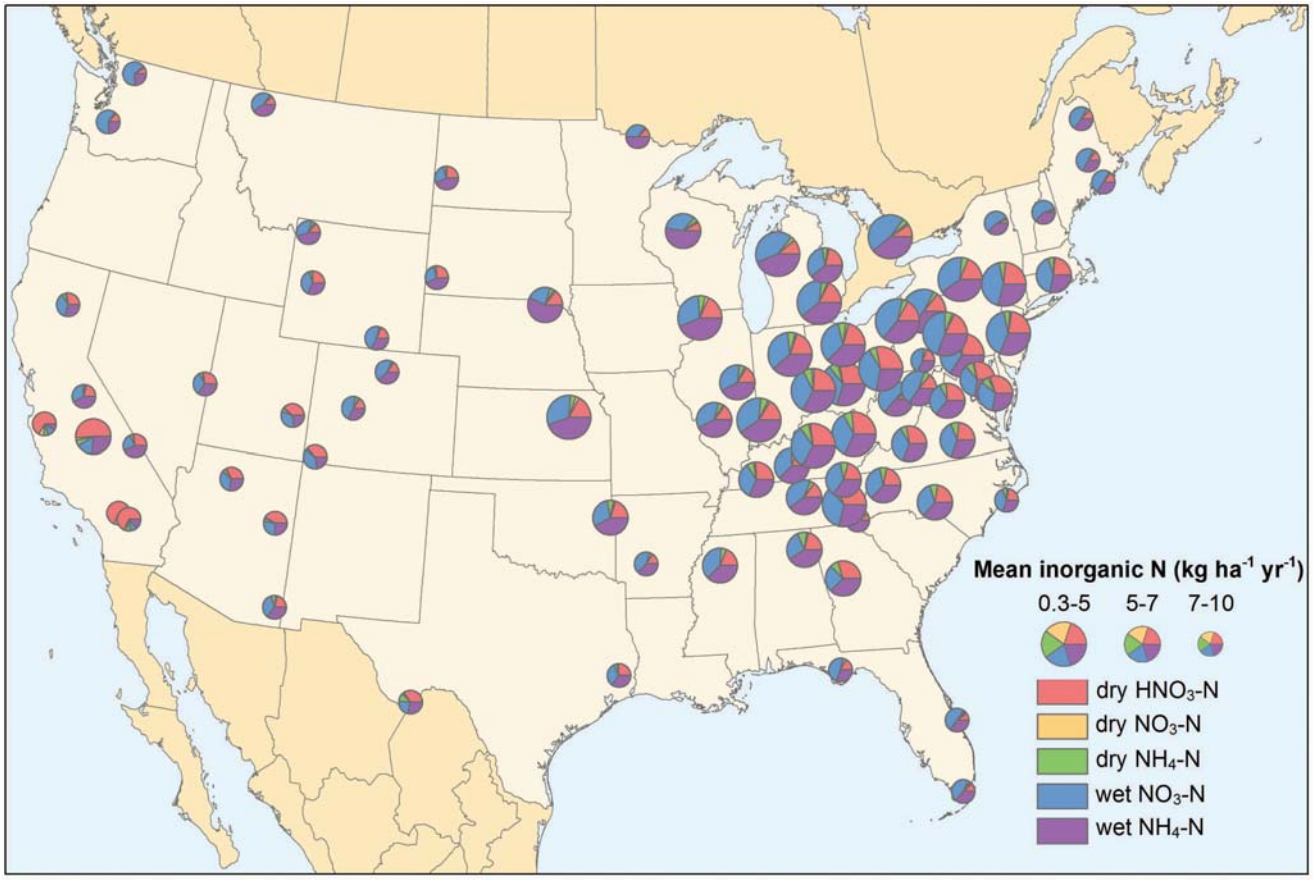

Figure 3.2—Total (wet plus dry, including wet NH_4^+ and NO_3^-, dry HNO_3, particulate NO_3^-, and NH_4^+) mean N deposition for 85 CASTNET site locations for 2004 to 2008. Size of circle indicates relative total deposition. Proportion of wet and dry species for each are shown within circle. Note that HNO_3-N is the largest proportion of dry deposition.

(Weathers et al. 2000a). Other potentially important N species not addressed here, primarily because they are not measured at National Atmospheric Deposition Program (NADP) or CASTNET monitoring stations (see descriptions, Chapter 4), are wet and dry NH_3, NO, NO_2, N_2O_5, peroxyacetal nitrate (PAN) (US EPA 2001). We note that fluxes of these species are estimated by the Community Multiscale Air Quality (CMAQ) model, however (see description, Chapter 4). One of the most important species that is not measured at CASTNET and NADP monitoring stations is NH_3. This form of N is currently increasing in the atmosphere and will be critically important to measure in both the short and long term (Driscoll et al. 2001, 2003).

In sum, estimates of total N deposition are still highly uncertain for several reasons, including the facts that (1) deposition models cannot accommodate complex terrain; (2) important N species are not often measured; (3) snow, dry, fog, or cloud deposition are not often considered, yet represent potentially important vectors for N deposition in some locations; and (4) there is inadequate monitoring (e.g., Driscoll et al. 2001, Lovett 1994, Weathers et al. 2000b, 2006).

3.3 Measurements and Estimates of N Deposition

3.3.1 Wet Inorganic N Deposition

Wet deposition is measured at approximately 250 monitoring stations across the United States as part of the NADP network. Wet deposition is the sum of NO_3^--N and NH_4^+-N delivered via rain and snow. Wet deposition is also estimated at various resolutions (e.g., 36 km x 36 km; 12 km x 12 km) via versions of the CMAQ model (Byun and Ching 1999, Byun and Schere 2006, US EPA 2009; Fig 3.1). The CMAQ model derives estimates of deposition from emissions, atmospheric chemistry, meteorology, and deposition velocities as influenced by land cover (US EPA 2009).

The chemical species we consider here from the CMAQ model are wet NH_4^+ and NO_3^-.

3.3.2 Cloud Deposition Monitoring

There exists only one cloud deposition monitoring network, the Mountain Atmospheric Deposition Program (MADPro), where cloud water samples are collected and measured for chemistry (NO_3^- and NH_4^+). These data are subsequently used to model cloud deposition for a few sites in the eastern United States (Anderson et al. 2006, Baumgartner et al. 2003). Cloud deposition estimates are not routinely extrapolated beyond the mountain tops where they are measured, such as Clingman's Dome, North Carolina, Whitetop Mountain, Virginia, and Whiteface Mountain, New York. Obtaining accurate estimates of cloud/fog deposition across the landscape is a significant problem (Fenn et al. 2009; Johnson and Lindberg 1993; Lovett 1994; Weathers et al. 1988, 2000a, 2000b, 2006). Where it is frequent, cloud/fog deposition can represent up to 80 percent of total N deposition (Anderson et al. 1999, Fenn et al. 2009, Johnson and Lindberg 1993).

3.3.3 Dry Deposition

Dry deposition is estimated at approximately 80 monitoring locations across the United States (US EPA 2010; Fig 3.2). For the ground-based CASTNET and AIRMoN (Atmospheric Integrated Research Monitoring Network) programs, air N chemistry is measured (HNO_3, particulate NO_3^-, and particulate NH_4^+), and dry deposition is modeled based on meteorologic conditions and multiple resistances of the receptor surfaces (CASTNET and AIRMoN; NADP 2009, US EPA 2010). Because protocols differ between the CASTNET and AIRMoN programs and there are fewer than 10 AIRMoN sites, we restrict our analysis to CASTNET data. Researchers have used CASTNET air concentration data to estimate deposition across the landscape (e.g., Fenn et al. 2009, Ollinger et al. 1993, NEG/ECP 2003, Weathers and Lynch 2008). The Environmental Protection Agency's CMAQ model also estimates dry deposition based on emissions, combined with air chemistry, meteorology and land cover-based deposition velocities. The chemical species we consider here from the CMAQ model are NH_4^+-N, NO_3^--N, and HNO_3-N.

3.3.4 Total Deposition

Total (wet plus dry plus cloud) inorganic N deposition has rarely been estimated across the U.S. landscape. In cases in which it has, indices of deposition or estimated air chemistry and meteorologic conditions have been used in combination with statistical and spatial tools (e.g., geographic information systems; NEG/ECP 2003; Weathers et al. 2000b, 2006). Wet plus dry (not cloud) deposition has been estimated across the country via the CMAQ modeling effort (Fig 3.1) as well as through statistical techniques (Ollinger et al. 1993), and for CASTNET locations using CASTNET and NADP data (Fig 3.2). Total flux to the forest floor (including cloud; throughfall) has been measured directly in many locations using bulk precipitation samplers (e.g. Simkin et al. 2004, Fenn et al. 2004). By comparing throughfall to wet-only deposition, estimates of dry (plus cloud, where relevant) deposition can be made (e.g., Ewing et al. 2009, Lovett 1994). However, often it is difficult to interpret these data vis-à-vis atmospheric deposition since canopy processing (uptake, leaching, and transformation) of N is common (Lovett 1994, Weathers et al. 2006).

As noted above, organic N has been identified as an important component of total N deposition for some ecosystems (Neff et al. 2002, Weathers et al. 2000a). However, few data are available to assess its temporal or spatial extent or importance in contributing to total deposition across the United States. Thus, throughout this document, unless otherwise noted, we will refer to total inorganic N deposition (or total N deposition) as the sum of NO_3^--N, NH_4^+-N, and HNO_3-N for dry deposition, deposited via wet and dry deposition (cloud deposition only when noted).

3.3.5 Temporal Trends in Wet plus Dry N Deposition Across the United States

Figure 3.3 shows wet plus dry N concentration and deposition across the United States for the 18-year period from 1990 through 2007, based on data from 34 CASTNET and NADP locations (US EPA 2007). While no trend lines are reported for these data, a 17 percent decrease in total inorganic N deposition over the period was reported. The proportion of wet to dry

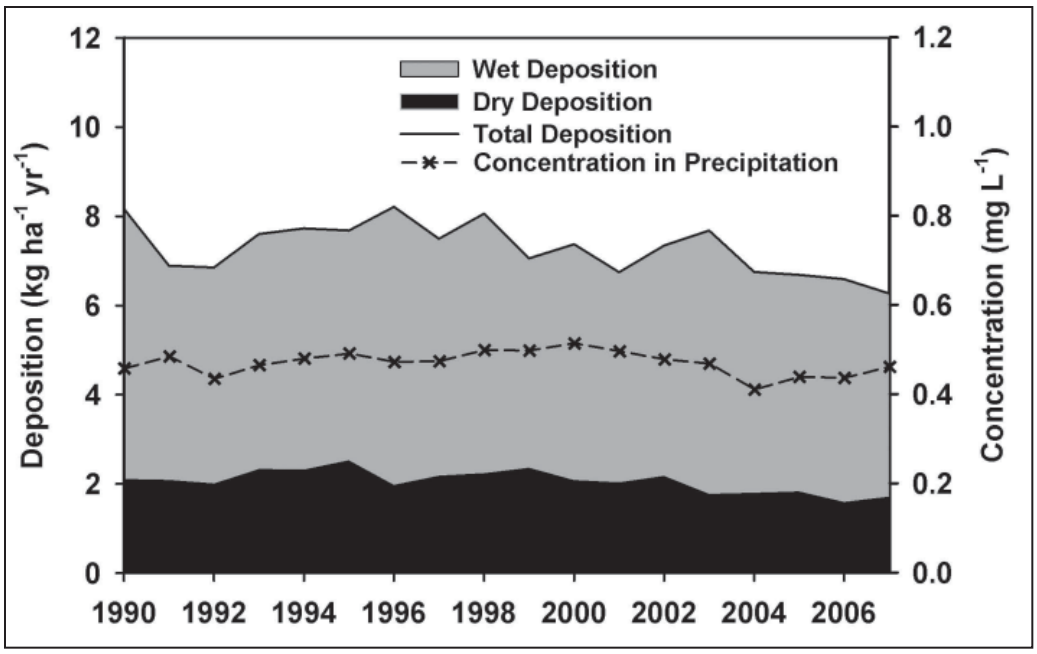

Figure 3.3—Total (solid black line), wet (gray area), dry (black area) deposition (kg N ha^{-1} yr^{-1}) and wet concentration (dotted line in mg L^{-1}) for 34 sites from 1990 to 2007 as estimated and measured by the NADP and CASTNET programs. Figure from US EPA 2007.

deposition also varies commensurate with changes in annual precipitation; when there is more precipitation, wet:dry ratios are greater (US EPA 2007).

3.4 Atmospheric N Deposition Across the Landscape

In this report, we have used the EPA's 2001 CMAQ model v. 4.3 (which use 2001 reported data and is hereafter referred to as CMAQ 2001 model) to provide comparative estimates of wet plus dry deposition to the more than 3200 locations included in this assessment of critical loads. We chose to use the CMAQ 2001 model (using the 36 km x 36 km resolution version) so that we could make comparisons across the country using the same method. For some regions, a 12 km x 12 km resolution version of CMAQ is available, but since it is not available for the whole country, we have not used it here. It is not possible to extrapolate either CASTNET or AIRMoN estimates of dry deposition beyond a 1 km x 1 km region surrounding the stations because the parameters required for the model are site-specific (US EPA 2010). It is also well known that estimates of total deposition that do not sufficiently consider landscape heterogeneity are likely to underestimate total deposition (Driscoll et al. 2001; Fenn and Bytnerowicz 1997; Fenn et al. 1998, 2000, 2009; Holland et al. 2005; Ito et al. 2002; Lovett 1994; Miller et al. 1993; Nanus et al. 2003; Ollinger et al. 1993; Weathers et al. 1992, 1995, 2000a, 2006). These issues notwithstanding, the CMAQ 2001 model represents a current state-of-the-art model to create cross-site deposition comparisons.

3.5 CMAQ Deposition Across Critical Loads Sites/Comparison Between CMAQ and Monitoring Stations

Average annual deposition across the more than 3200 critical load sites varied more than 30-fold, from 0.5 to 17 kg N ha^{-1} yr^{-1} (Fig 3.1). Total N deposition estimates based on the CMAQ 2001 model for CASTNET site locations compared well to those generated using CASTNET dry plus NADP wet data (Fig. 3.4, R^2=0.75). Note that in order to compare these two estimates, total N deposition included wet NO_3^- and NH_4^+ and dry HNO_3 and NH_4^+ as N, only since NH_3 data were not available for CASTNET sites.) However, air concentrations of HNO_3-N, the largest proportion of dry deposition, show a consistent difference (CMAQ < CASTNET), with a positive linear relationship (Fig 3.5; R^2=0.68). This is in contrast to estimates of dry deposition of HNO_3-N, which are also positively correlated (Fig 3.6; R^2=0.45), but with

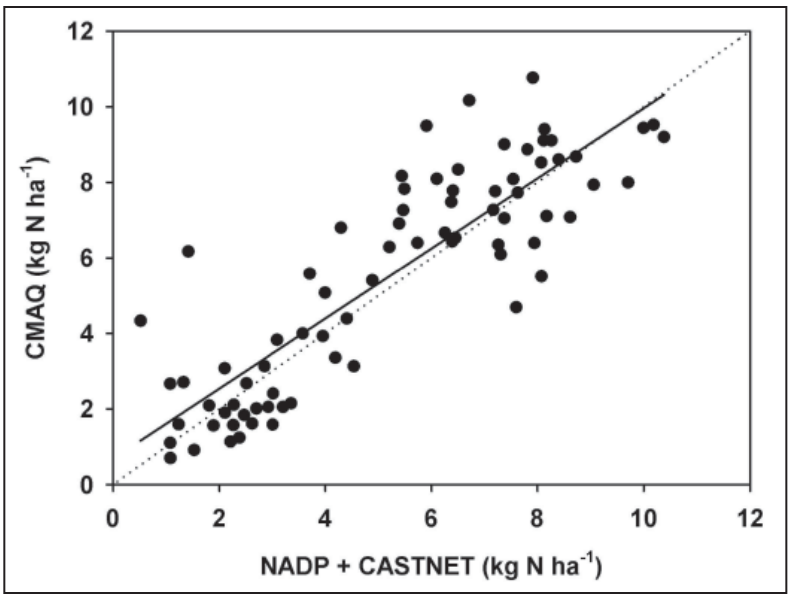

Figure 3.4—Comparison between wet plus dry deposition estimates for 2001 (includes wet NH_4^+ and NO_3^-, dry HNO_3, particulate NO_3^- and NH_4^+) from CASTNET and NADP (NADP site nearest to CASTNET locations) and CMAQ 2001 model for 77 CASTNET site locations. y = 0.9281x + 0.6818, R^2 = 0.75. 1:1 line is shown (from Weathers and Lynch 2008).

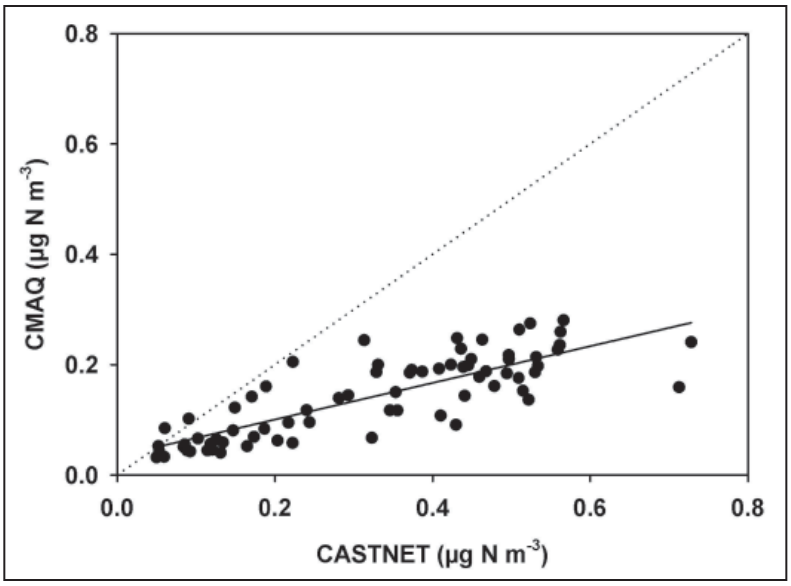

Figure 3.5—Mean air concentration (µg m^{-3}) of HNO_3-N during 2001 measured at 77 CASTNET sites vs. the mean air concentrations generated by the CMAQ 2001 model for those sites during 2001. y = 0.3324x + 0.034, R^2 = 0.6832. The 1:1 line is shown (from Weathers and Lynch 2008).

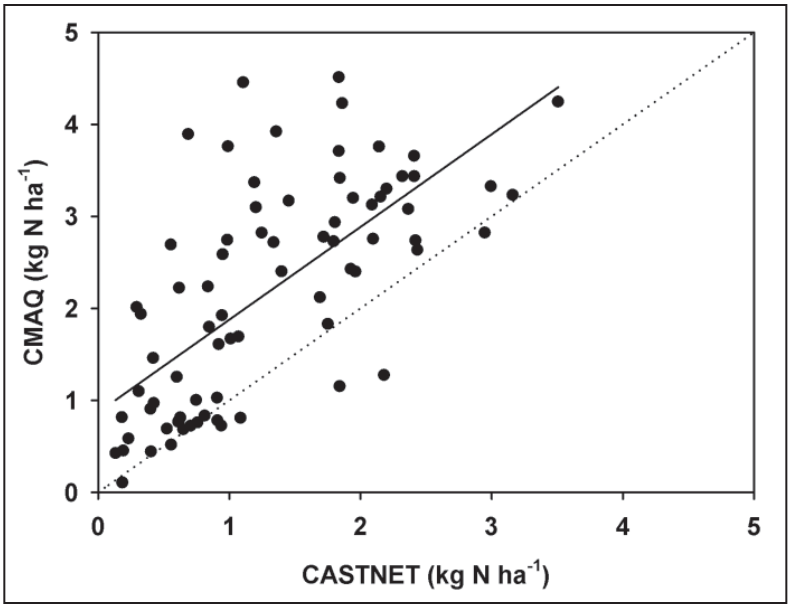

Figure 3.6—N dry deposition (kg N ha^{-1} yr^{-1}) during 2001 modeled at 77 CASTNET sites vs. the N dry deposition generated by the CMAQ 2001 model for those sites during 2001. y = 1.008x + 0.869, R^2 = 0.45. The 1:1 line is shown (from Weathers and Lynch 2008).

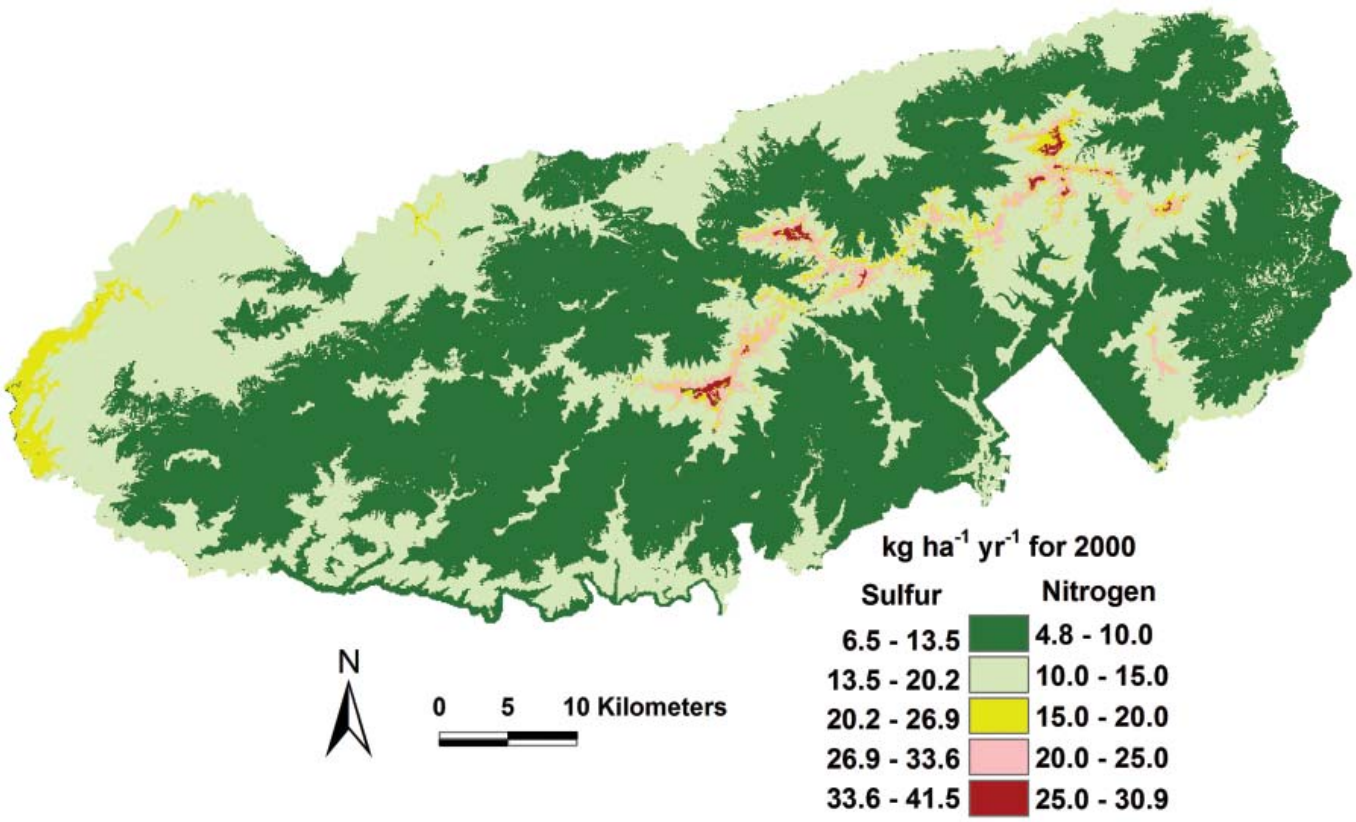

Figure 3.7—Total (wet, dry, and cloud) inorganic N (and S) deposition estimated for Great Smoky Mountain National Park for the year 2000 (from Weathers et al. 2006).

CMAQ estimates greater than CASTNET estimates, suggesting that dry deposition velocities applied to air concentration data for the two different methods vary (these analyses based on Weathers and Lynch 2008).

We note, however, that the CMAQ 2001 model that was available to us at the time of this analysis could not resolve deposition levels below 36 km x 36 km grids compared to deposition that has been measured in the field. For example, Fenn et al. (2000, 2008) have measured 71 kg N ha^{-1} yr^{-1} deposition in the San Bernardino Mountains, an area that, according to the CMAQ 2001 model, would receive only 3 to 5 kg N ha^{-1} yr^{-1}. Similarly, Weathers et al. (2006) have estimated N deposition to be as high as 31 kg N ha^{-1} yr^{-1} at Clingman's Dome, North Carolina (Fig. 3.7), whereas both CMAQ and NADP plus CASTNET deposition for that same location and year would be less than 10 kg N $ha^{-1}yr^{-1}$. In these cases, the CMAQ estimates are several times lower than on-site measures of deposition that include wet plus dry plus cloud deposition.

In summary, the comparison of N deposition estimates from the CMAQ model to CASTNET plus NADP suggest that either method could be used to generate a comparative estimate of wet plus dry deposition to flat, homogenous terrain (Fig. 3.4). The differences between CMAQ and other estimates of N deposition (Fenn et al. 2003, Weathers et al. 2006) are not surprising, given the inadequacy of dry deposition models in handling complex terrain and the lack of cloud deposition estimates (Weathers et al. 2006), as well as the coarse scale (36 km x 36 km) over which the CMAQ model predicts deposition (Weathers and Lynch 2008).

3.6 Future Research Directions and Gaps in Data

Despite decades of research, accurate estimates of total N deposition are lacking. While various methods for estimating deposition (see above) exist, almost all methods estimate only portions of total N inputs to ecosystems. Not all chemical species of N are measured or monitored, nor are all forms of deposition estimated. Priorities for future deposition research include: expanding monitoring station spatial coverage (especially in high elevation and arid sites and in the West); measuring more chemical species of N; generating better estimates of dry (and, where it is an important component of atmospheric deposition, cloud/fog) inputs; creation of models and maps of deposition across heterogeneous landscapes that incorporate or reference existing modeling and/or monitoring data (e.g., Ollinger et al. 1993, Holland et al. 2005, Weathers et al. 2006); expanding the use of passive sampling devices for chemical species such as NH_3 (e.g., Fenn et al. 2009); coupling empirical measurements (such as throughfall) with monitoring data (e.g., Weathers et al. 2006); and comparing estimates of deposition using different methods.

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