

Nitrogen Emissions, Deposition, and Monitoring in the Western United States

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Nitrogen (N) deposition in the western United States ranges from 1 to 4 kilograms (kg) per hectare (ha) per year over much of the region to as high as 30 to 90 kg per ha per year downwind of major urban and agricultural areas. Primary N emissions sources are transportation, agriculture, and industry. Emissions of N as ammonia are about 50% as great as emissions of N as nitrogen oxides. An unknown amount of N deposition to the West Coast originates from Asia. Nitrogen deposition has increased in the West because of rapid increases in urbanization, population, distance driven, and large concentrated animal feeding operations. Studies of ecological effects suggest that emissions reductions are needed to protect sensitive ecosystem components. Deposition rates are unknown for most areas in the West, although reasonable estimates are available for sites in California, the Colorado Front Range, and central Arizona. National monitoring networks provide long-term wet deposition data and, more recently, estimated dry deposition data at remote sites. However, there is little information for many areas near emissions sources.

Keywords: urbanization, animal feeding operations, hotspots, policy implications, regional haze

Nitrogen (N) emissions and deposition data in the 11 most westerly contiguous states indicate that N deposition has steadily increased over most of this region in recent years (figure 1; Nilles and Conley 2001, Baumgardner et al. 2002), along with a rapid increase in population of 46.8% from 1980 to 2000, compared with 24.2% for the country as a whole (USCB 2001). Continuing rapid population growth in the West highlights the need for greater emissions controls. Population projections from 1995 to 2025 indicate that 10 of the 12 lower 48 states with the highest projected growth rates are western states (Campbell 1996).

Major effects of N deposition have been documented in terrestrial and aquatic ecosystems in the western United States, primarily in response to N enrichment of systems that are naturally N limited (Fenn et al. 1998). These effects include increased greenhouse gas emissions, higher N concentrations in plant tissues, and increased nitrification rates and nitrate (NO_3^-) levels in soil, streams, and lakes (Riggan et al. 1985, Fenn et al. 1996, Williams et al. 1996, Fenn and Poth 1999). Elevated NO_3^- levels in runoff raise concerns about drinking water quality and eutrophication of recipient water bodies (Fenn et al. 1998). Nitrogen enrichment of these ecosystems, a result of chronic N deposition, has caused important

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community changes in vegetation, lichens, mycorrhizae, and phytoplankton, sometimes at relatively low N deposition levels (3 to 8 kilograms [kg] N per hectare [ha] per year; Baron et al. 2000, Fenn et al. 2003a). In urban areas and downwind of urban areas, emissions of nitrogen oxides (NO_x) frequently lead to elevated concentrations of ozone (O_3), a pollutant that may cause severe injury to sensitive plant species (Miller and McBride 1999). Nitrogen emissions also contribute to regional haze and impaired visibility in national parks and other wildlands, a pollution problem that is evident

often at great distances (e.g., hundreds of kilometers [km]) from the emissions source areas; Malm et al. 2000, Tonnesen et al. 2002).

The effects of N deposition in some areas of the western United States are not as well known or studied as in the eastern United States and in Europe (Aber et al. 1989, Bytnerowicz and Fenn 1996, Erisman and de Vries 2000, Driscoll et al. 2001). Emissions data, deposition modeling studies, demographic trends, and the increasingly urbanized character of the West all point to the need for increased monitoring and

studies of ecological effects in the expanding urban zones and adjacent wildlands of the western United States. Estimates of N deposition inputs are generally available for the most intensively studied ecosystems in the West, but N deposition data are minimal or completely lacking for many western sites. Particularly lacking are data on dry deposition inputs, usually the largest component of total atmospheric N deposition in polluted western sites (Bytnerowicz and Fenn 1996), except in areas with frequent fog (Fenn et al. 2000). Deposition rates in the West are likely to be highest downwind of large urban areas, but N deposition rates may also be high in nonurban areas downwind of agricultural sources (Tonnesen et al. 2003). In this article we review current information on N deposition sources and rates in key western ecosystems and on the current state of N deposition monitoring in the West. We also report N deposition rates from a modeling study of aerosol formation and regional haze in the western United States (Tonnesen et al. 2003).

Nitrogen emissions sources and trends

Transportation, agriculture, power plants, and industry are the major sources of N emissions in the West. Transpacific transport of N from Southeast Asia is another source of atmospheric N along the West Coast, although the amounts are uncertain. Nitrogenous emissions from the transportation sector in urban areas are dominated by NO_x , while emissions from fertilized

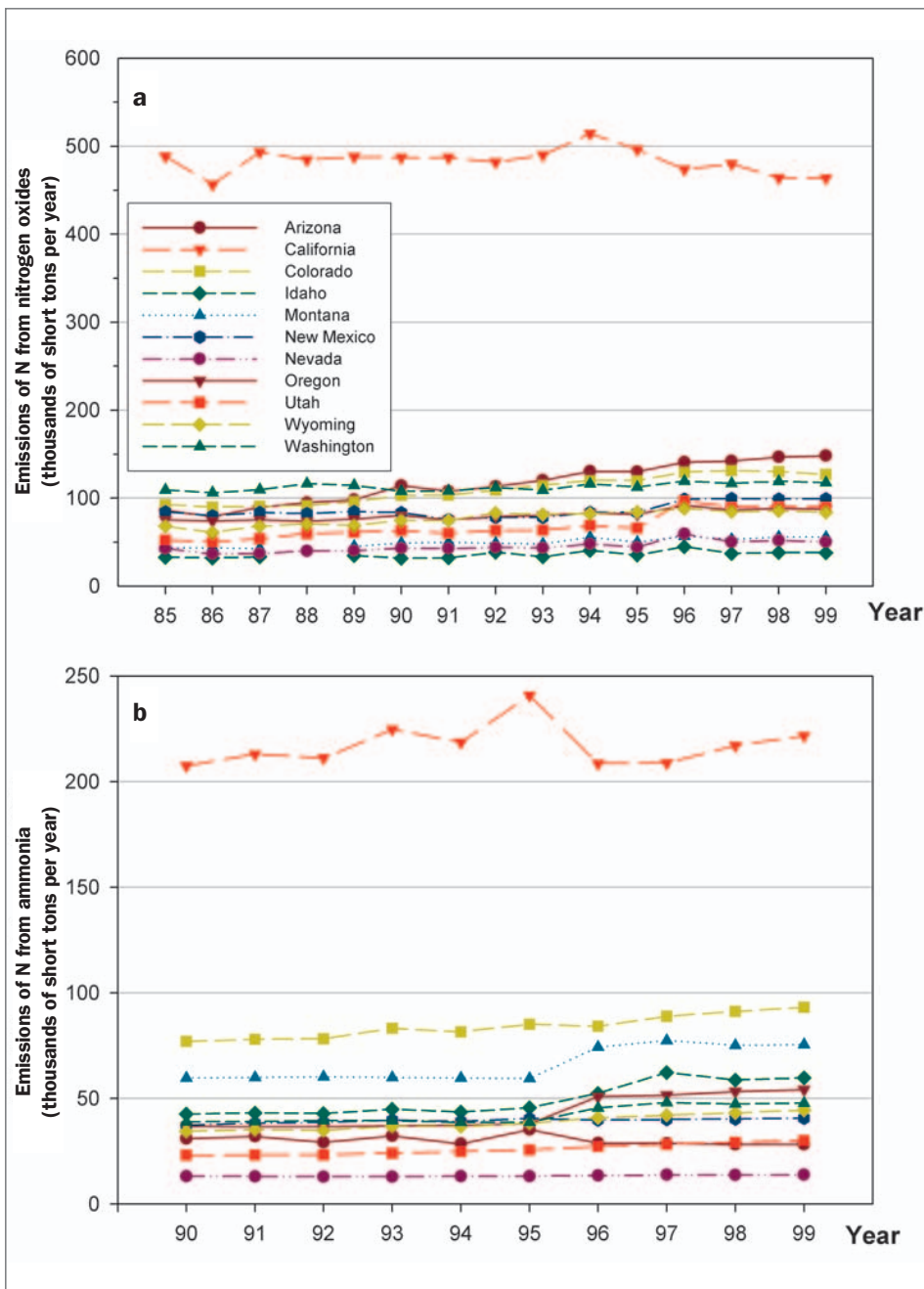


Figure 1. Trends in emissions of nitrogen from nitrogen oxides (a) and from ammonia (b), in thousands of short tons per year (1 short ton = 907.2 kilograms), in 11 western states. Data are from the National Emission Trends database (EPA 2003).

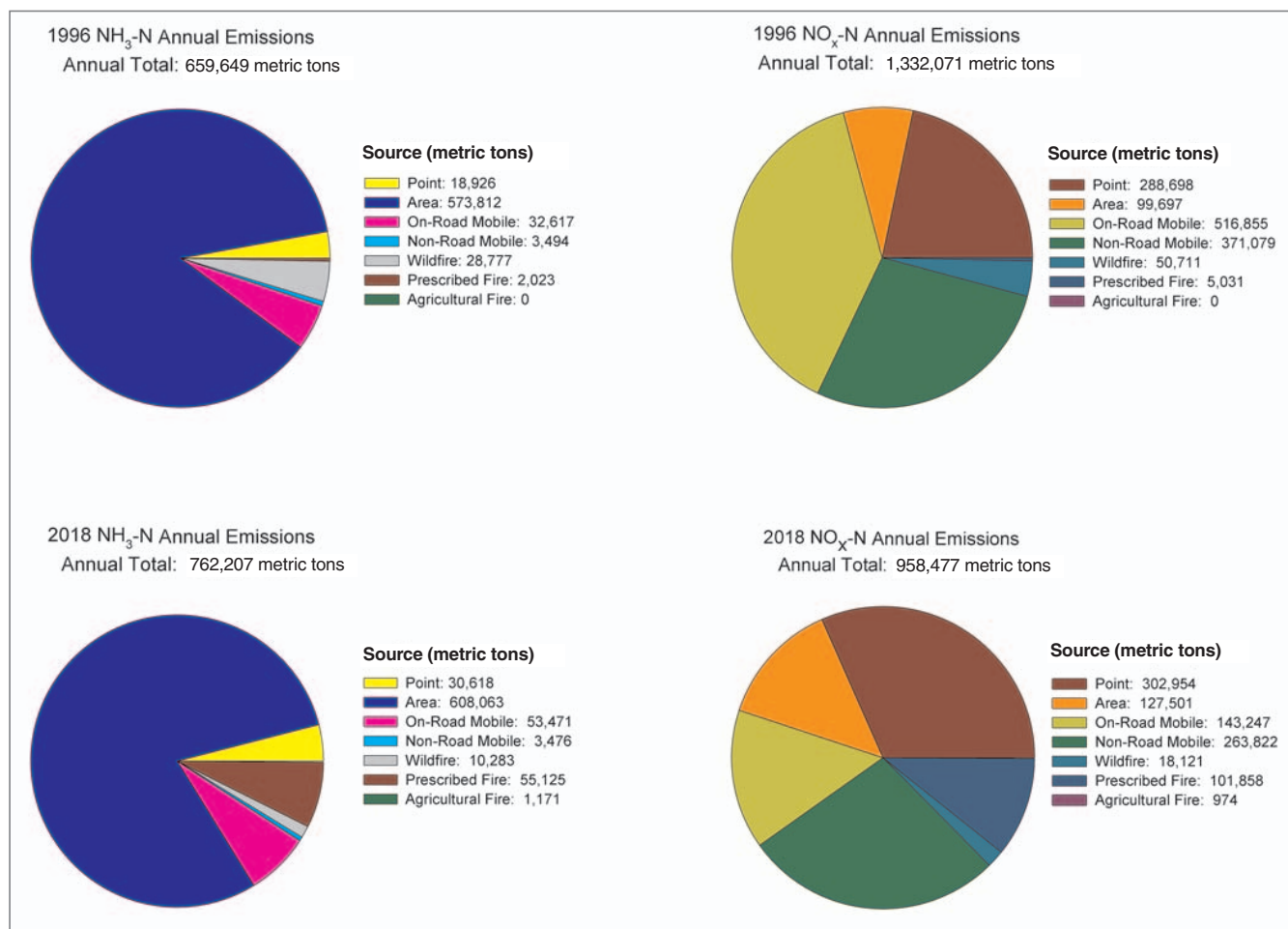


Figure 2. Estimated total emissions of nitrogen, in metric tons, from nitrogen oxides (NO_x-N) and from ammonia (NH₃-N) in 1996 and projected emissions in 2018 in the 11 contiguous western states (Gribovicz 2002). The states include Arizona, California, Colorado, Idaho, Montana, New Mexico, Nevada, Oregon, Utah, Washington, and Wyoming.

crops and concentrated animal feeding operations (CAFOs) are mainly in reduced forms (ammonia [NH₃] and ammonium [NH₄⁺], referred to collectively as NH_x). Figure 2 shows the estimated total emissions of N from NO_x and NH₃ in 1996 and projected emissions for 2018 for the 11 contiguous western states (Gribovicz 2002). Mobile sources, including on-road and off-road sources, make up 67% of the anthropogenic NO_x emissions in these states. Industrial and power plant emissions are also important sources of N pollutants in some areas, accounting for 22% of anthropogenic NO_x. Area sources, primarily crop agriculture and livestock, account for 87% of the estimated 1996 NH₃ inventory. Estimated emissions of NH₃ (659,649 metric tons N from NH₃ per year) were 50% as high as NO_x emissions (1,332,071 tons N from NO_x per year) in the 11 contiguous western states in 1996. However, the accuracy of the NH₃ inventory is highly uncertain, particularly for data on biogenic emissions from soil, which are excluded from the Western Regional Air Partnership (WRAP) inventory (Gribovicz 2002).

Emissions of NO_x in 10 of the 11 contiguous western states increased by 8% to 72% from 1985 to 1999 (an over-

all average increase of 16%; figure 1), based on data from the National Emissions Trends database (EPA 2003). The only western state with a decrease in NO_x emissions was California, where annual NO_x emissions decreased by 5%. However, according to the data published by the state of California (Alexis et al. 2001), NO_x emissions decreased 28% from 1985 to 2000. Emissions of NH₃ increased by an average of 19% in the 11 western states from 1990 to 1999 (figure 1). Precipitation-adjusted NH₄⁺ and NO₃⁻ concentrations in wet deposition increased at 65% and 35%, respectively, of the western National Atmospheric Deposition Program (NADP) sites from 1985 through 1999, while concentrations were generally stable at the remaining sites (Clow et al. 2003). Nilles and Conley (2001) reported significantly increasing trends in volume-adjusted NH₄⁺ and NO₃⁻ concentrations from 1981 to 1998 at western NADP wet deposition monitoring sites, with no decreasing trends. Emissions of NO_x in the western United States are projected to decrease 28% by 2018, while NH₃ emissions are projected to increase by 16% (figure 2; Gribovicz 2002). Projected NO_x reductions are expected to result largely from reduced mobile emissions as stricter

vehicle emissions controls and technical improvements are implemented. However, N deposition is expected to increase in some natural areas as urbanization continues to expand.

Modeling chemical transformations and deposition of nitrogen in the West

Long-term atmospheric modeling efforts to study the formation and transport of regional haze in the western United States were initiated in 2001 by the Western Governors' Association and WRAP (Tonnesen et al. 2002). Because N compounds are key components of the regional haze problem, the haze model simulations also represent the formation and deposition of N species. The atmospheric budget and deposition of N are determined by a complex interaction of multiple processes, including emissions of N species, meteorological dynamics, chemical transformation and partitioning of N species, aerosol dynamics, and rates of wet and dry deposition of aerosol and gas-phase N species.

We report selected results here of a larger study of simulated rates of total (wet and dry) N deposition for an annual modeling study of aerosol formation and regional haze in the western United States (Tonnesen et al. 2003). Model simulations were performed using the Models-3/Community Multiscale Air Quality (CMAQ) model (Buyn and Ching 1999) and data on emissions from the 1996 National Emissions Inventory, with many updates made for the western states (Tonnesen et al. 2002). The CMAQ model is designed to represent both wet and dry deposition of aerosol and gas-phase species. Although the model simulates the rates of deposition for individual N species (at 36 km resolution in these simulations), we present summary results in figure 3 for total NH_x (NH_3 and NH_4^+) and for total NO_y (nitric oxide [NO], nitrogen dioxide [NO_2], nitric acid [HNO_3], and NO_3^-).

Several hotspots for total N deposition stand out in figure 3, particularly adjacent to urban areas in southern and central California, the Pacific Northwest, Salt Lake City, Phoenix, southwestern Utah, northwestern Arizona, and parts of

Colorado. The elevated N deposition in eastern Idaho is consistent with large NH_3 emissions from livestock operations in south-central Idaho (figure 3). There is a high level of uncertainty in the model simulations of N deposition because of uncertainty in model inputs, especially in the emissions inventories that are key to performing budget analyses. The uncertainties are largest for NH_3 , which plays a central role in the conversion of HNO_3 to aerosols. While absolute values for N deposition are highly uncertain, the results indicate likely areas of elevated N deposition in the West. Future modeling studies can provide better estimates of N deposition through the use of better emissions data and a finer model grid resolution.

Monitoring nitrogen deposition

Data on N deposition from three networks, IMPROVE (Interagency Monitoring of Protected Visual Environments), CAST-Net (Clean Air Status and Trends Network), and NADP/NTN (NADP/National Trends Network), rarely indicate the areas most affected by N deposition in the West because (a) dry deposition and fog deposition (including cloudwater), and, in particular, deposition fluxes to canopies, either are not measured or are only partially quantified; (b) monitoring sites are located in remote locations far from the pollution sources; (c) the number of monitoring sites is insufficient; and (d) sites are rarely located in the highest-elevation areas, where ecosystems are likely to be of highest sensitivity. The CASTNet sites are intentionally located away from major sources of sulfur dioxide (SO_2) and NO_x (Baumgardner et al. 2002). This is also true of the IMPROVE sites, because they are intended to address the regional haze problem in federal Class I areas. Measurements of deposition must account for fluxes to vegetation, which function as efficient scavengers of N in areas with high leaf-area index values, particularly in dry forms or in fog. Bulk deposition and wet deposition measurements that do not account for dry deposition or fog or cloudwater deposition to plant canopies can grossly underestimate atmospheric

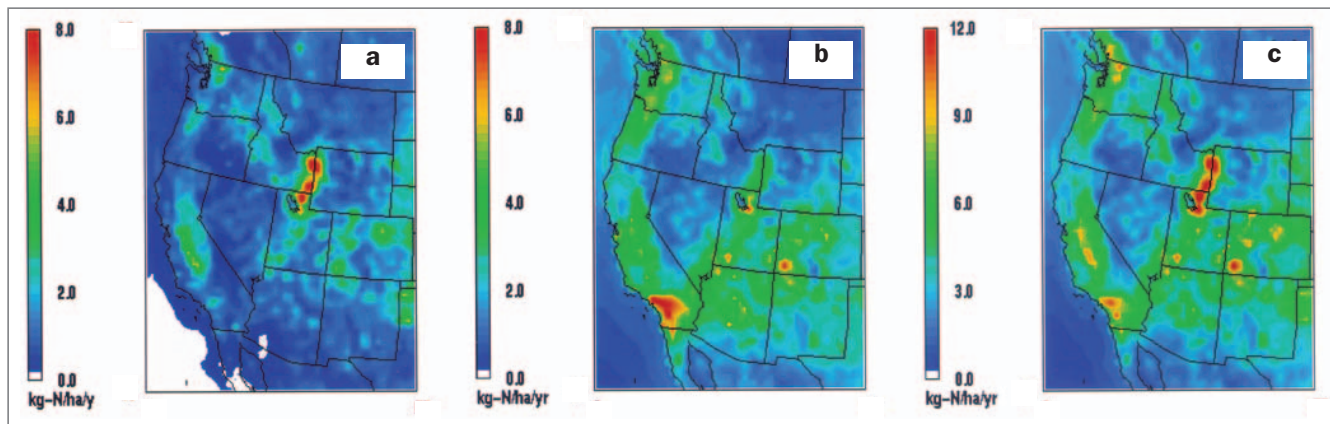


Figure 3. Model-simulated annual nitrogen (N) deposition (kilograms per hectare per year) in the western United States in 1996 for (a) total wet and dry deposition of N from ammonia and ammonium ($\text{NH}_x\text{-N}$), (b) total wet and dry deposition of N from nitric oxide, nitrogen dioxide, nitric acid, and nitrate ($\text{NO}_y\text{-N}$), and (c) total N deposition calculated as the sum of $\text{NH}_x\text{-N}$ and $\text{NO}_y\text{-N}$. Note that the scale for the y-axis in (a) and (b) differs from the scale in (c).

inputs. For example, throughfall fluxes under ponderosa pine (*Pinus ponderosa* Dougl. ex Laws.) canopies at high- and low-deposition sites in southern California were 18 times and 5 times higher, respectively, than bulk deposition in adjacent forest openings (Fenn et al. 2002). Nonetheless, NADP's wet deposition data are frequently cited, because NADP has the most spatially and temporally extensive database on deposition in the United States.

Total N deposition at Tanbark Flat in the Los Angeles air basin in southern California and in surrounding forests is reportedly 20 to 45 kg per ha per year. Wet deposition, at 3 to 5 kg per ha per year, is a very small portion of that total (Riggan et al. 1985, Fenn et al. 2000, 2002). Yet the widely used NADP map figures represent this region as an area with near-background levels of N deposition in precipitation. Furthermore, fog deposition inputs are often similar in magnitude to dry deposition inputs in southern California forests (Fenn et al. 2000) and in parts of the Sierra Nevada (Collett et al. 1989). Fog deposition is not measured in the NADP wet/dry buckets or in dry deposition networks, but is a major N deposition form wherever fog episodes and N emissions sources coincide.

Nitrogen deposition data in rural areas of the West are limited in scope, and where data are available, total deposition estimates are often underestimated and highly uncertain. This is largely because of the technical difficulties and expense associated with measuring the atmospheric concentrations and deposition of the array of dry-deposited N forms. Adding to the challenge of measuring dry deposition of N is the need to quantify N deposition in precipitation, and in some areas N deposition in fog or cloudwater must also be determined. Various approaches have been used to determine annual N deposition and some components of total N deposition. CASTNet is the only monitoring program with a number of sites in the West (currently 22 in the lower western states and 2 in Alaska) that attempts to estimate dry deposition of some of the major N species. However, NO, NO₂, NH₃, and peroxyacetyl nitrate (PAN) concentrations are not measured at the CASTNet sites, and therefore total N deposition is frequently underestimated (Baumgardner et al. 2002).

Various methods have been used to measure dry deposition of N in site-specific studies. The most common of these methods are branch rinsing, throughfall collection, and the inferential method (based on atmospheric concentrations measured with annular-denuder systems or filter packs and calculated using estimated pollutant deposition velocities; Bytnerowicz and Fenn 1996). A limiting factor in the widespread implementation of the inferential method (as used by CASTNet) is the quantity of data needed to calculate dry deposition (e.g., leaf-area index and other plant community characteristics, deposition velocity values for each N species, spatial and temporal distribution of atmospheric concentrations of a number of N compounds, and numerous climatic variables; Baumgardner et al. 2002). Throughfall fluxes generally underestimate total atmospheric N deposition because the canopy retains N (Fenn and Bytnerowicz 1997), but they

are nonetheless a very useful measure of N deposition in solution to the soil or forest floor. The use of passive throughfall collectors, based on retention of ions from throughfall solutions onto ion exchange resin columns (Fenn et al. 2002) or resin bags (Kjønaas 1999, Susfalk and Johnson 2002), is an attractive alternative to more labor-intensive and costly methods, allowing total N deposition to be monitored at a greater number of sites than is practical with conventional throughfall collectors. In subalpine sites in the Sierra Nevada and in the Colorado Front Range, snowpack samples are often analyzed for N to estimate total winter deposition (Clow et al. 2002, Sickman et al. 2002).

Nitrogen deposition in the West

Nitrogen deposition varies widely across the West in terms of total amounts, seasonality, physical forms, and effects. For many western areas, N deposition has not been measured.

The Colorado Front Range. Developments in the Colorado Front Range in recent decades illustrate some of the key factors contributing to increased N deposition in the West: a greater prevalence of large animal husbandry operations and rapid increases in urbanization, population, and distance driven. Nitrogen deposition has increased since the 1980s at high-elevation sites in the Colorado Front Range, and total deposition values currently range from 4 to 8 kg N per ha per year (Baron et al. 2000). The Continental Divide is an orographic barrier that separates the Colorado Rockies into regions of low and higher atmospheric N deposition inputs. Most of Colorado's population lives in the region east of the Continental Divide, which is also an important region for livestock production and agriculture (Baron et al. 2000). The combination of anthropogenic atmospheric N sources, local easterly winds, and orographic precipitation results in elevated N deposition rates to eastern Colorado high-elevation ecosystems (Sievering et al. 1989, Heuer et al. 2000, Williams and Tonnessen 2000). As precipitation increases with elevation, deposition rates increase as well (Williams and Tonnessen 2000). Deposition rates west of the Continental Divide are lower because N sources are limited and because strong westerly winds in the upper atmosphere rarely allow N-enriched air masses from the east to cross the divide (Bossert 1990, Baron and Denning 1993).

California. Wildland ecosystems within the South Coast air basin, which includes portions of four counties in the Los Angeles area, receive the highest N deposition in the country (Fenn et al. 1998, 2002). The most exposed areas are the south-facing slopes of the San Gabriel Mountains and the western and southern edges of the San Bernardino Mountains. Several federal Class I wilderness areas in the South Coast air basin are affected by N deposition. Nitrogen deposition inputs in the low- and midelevation chaparral and mixed conifer forest zones range from 20 to 45 kg per ha per year in the most exposed areas. However, deposition values can be higher than 90 kg per ha per year in years of more extensive

fog exposure, particularly when fog occurs in late summer with unusually high NO_3^- and NH_4^+ concentrations (figure 4; Fenn et al. 2002). Deposition under individual canopies of large trees is even higher, because these stands are relatively open (60% to 70% forest cover; Fenn and Bytnerowicz 1997, Fenn et al. 2000). NO_x emissions decreased by 35% from 1975 to 2000 in the South Coast air basin because of strict regulatory controls (Alexis et al. 2001); this suggests that N deposition may have been higher in past decades.

In the Sierra Nevada and in the San Bernardino Mountains in southern California, NH_x deposition is often equal to or greater than NO_x deposition (figure 4; Bytnerowicz et al. 2002, Fenn et al. 2003b). Throughfall deposition ranges from 10 to 13 kg N per ha per year in chaparral and mixed conifer forest sites in the southwestern Sierra Nevada, and from 1 to 7 kg N per ha per year in central and northern sites along the western side of the Sierra Nevada (figure 4; Fenn et al. 2003b). Total N deposition inputs are expected to be 30% to 40% higher than these throughfall values, because the canopy retains atmospheric N (Fenn et al. 2000). The alpine and subalpine zones of the Sierra Nevada contain large tracts of federal Class I areas that are protected by the Clean Air Act. The higher elevations of the Sierra Nevada of California receive on average about 3 kg per ha per year of total N (dissolved inorganic and organic N) deposition (Sickman et al. 2001). However, this is considered a conservative estimate of total deposition for alpine and subalpine sites in the southern Sierra Nevada. While nearly 90% of precipitation falls as snow during the winter (typically from December to April), 60% of annual N deposition in the alpine and subalpine zones occurs during the months of May through November. This deposition pattern is largely the result of differences in the air quality and storm trajectories between winter and nonwinter periods. Winter storms are derived from relatively clean air masses associated with Pacific frontal systems from the Gulf of Alaska, whereas nonwinter precipitation originates from localized thunderstorms formed from relatively polluted air from the Central Valley of California. In the San Joaquin Valley air basin, which includes the southern two-thirds of the Central Valley, air quality improvements have not kept pace with those in most urban areas of California. There are a number of moderately sized urban areas located along the main axis of the valley, and the area also includes many agricultural emissions sources, making air quality control difficult. Large dairy farm operations, a major source of NH_x , are increasing in the region. According to NADP data, wet deposition of NO_3^- and NH_4^+ in Sequoia National Park, in the more polluted southern Sierra, increased from 1981 to 2001, although the trend was significant only for NH_4^+ (0.037 kg per ha per year; $p = 0.10$). Nitrogen deposition is expected to continue to increase in the Sierra Nevada.

Central Arizona–Phoenix. In the arid Southwest, rainfall is infrequent and seasonal; hence, a substantial proportion of N deposition inputs to terrestrial and aquatic systems occurs as dry deposition. The Phoenix metropolitan area is character-

istic of many rapidly growing cities in the arid and semiarid lands of the southwestern United States, with large tracts of undeveloped desert lands within and downwind of the city. The major source of NO_x is fossil fuel combustion from transportation in the central urban core (Fernando et al. 2001). As part of ongoing research at the Central Arizona–Phoenix (CAP) Long Term Ecological Research site, investigators have employed a combination of measurements and modeling techniques to determine the temporal and spatial characteristics of wet and dry N deposition across the CAP study area. Annual inputs of dry-deposited, oxidized forms of N have been estimated in a detailed modeling effort for the years 1996 and 1998 by means of a diagnostic model. The model results show that NO_x dry deposition has a marked seasonal pattern, peaking over the winter months (October to March) and declining during the summer, tracking the seasonal variation in ambient pollutant concentrations. In the same study, Models-3/CMAQ simulations (Buyn and Ching 1999) gave predicted hourly NO_x and HNO_3 deposition, allowing spatial patterns in NO_x deposition to be evaluated for the modeling domain (10,664 km²), which included the entire Phoenix metropolitan area along with the surrounding desert and agricultural land.

Models-3/CMAQ simulations for a single day episode in July 1996 were used as a basis for estimating dry deposition across the whole study area. Scaling these whole-system predictions to account for seasonal variations, and then adding 3.5 kg N per ha per year for deposited NH_4^+ (as estimated by Baker et al. 2001) and an additional 10% of the subtotal (average 1.2 kg N per ha per year) to account for deposition in particulate form, dry deposition (kg N per ha per year) was estimated to be 13.5 for the urban core, 15.0 for the downwind desert, and 7.5 for the upwind desert (figure 5), with an average for the entire system of approximately 12 kg N per ha per year. Wet deposition to the entire CAP study area was determined from wet bucket samplers at eight sites across the study area. Combining the results from these various techniques gave a total N deposition rate of approximately 13.5 kg N per ha per year, which represents 20% of total N inputs to the study area (Baker et al. 2001). Estimated total N deposition to the most exposed agricultural and desert areas was 28 to 29 kg per ha per year (figure 5), but this occurred over only a small proportion (2%, or about 200 km²) of the modeling domain.

The Pacific Northwest. Excluding urban areas, N deposition in the Pacific Northwest is generally low compared with deposition in other parts of the United States. Annual wet N deposition reported by NADP (8 March 2003; <http://nadp.sws.uiuc.edu>) has been under 1.6 kg per ha each year at 8 of the 10 monitors in operation since the 1980s. Higher annual wet N deposition (up to 3.2 kg per ha) has been detected at only two monitors, Bull Run and Marblemount, east of Portland and Seattle, respectively. However, concentrations of NO_3^- and NH_4^+ at NADP sites in Oregon and Washington have been increasing since monitoring began in 1980.

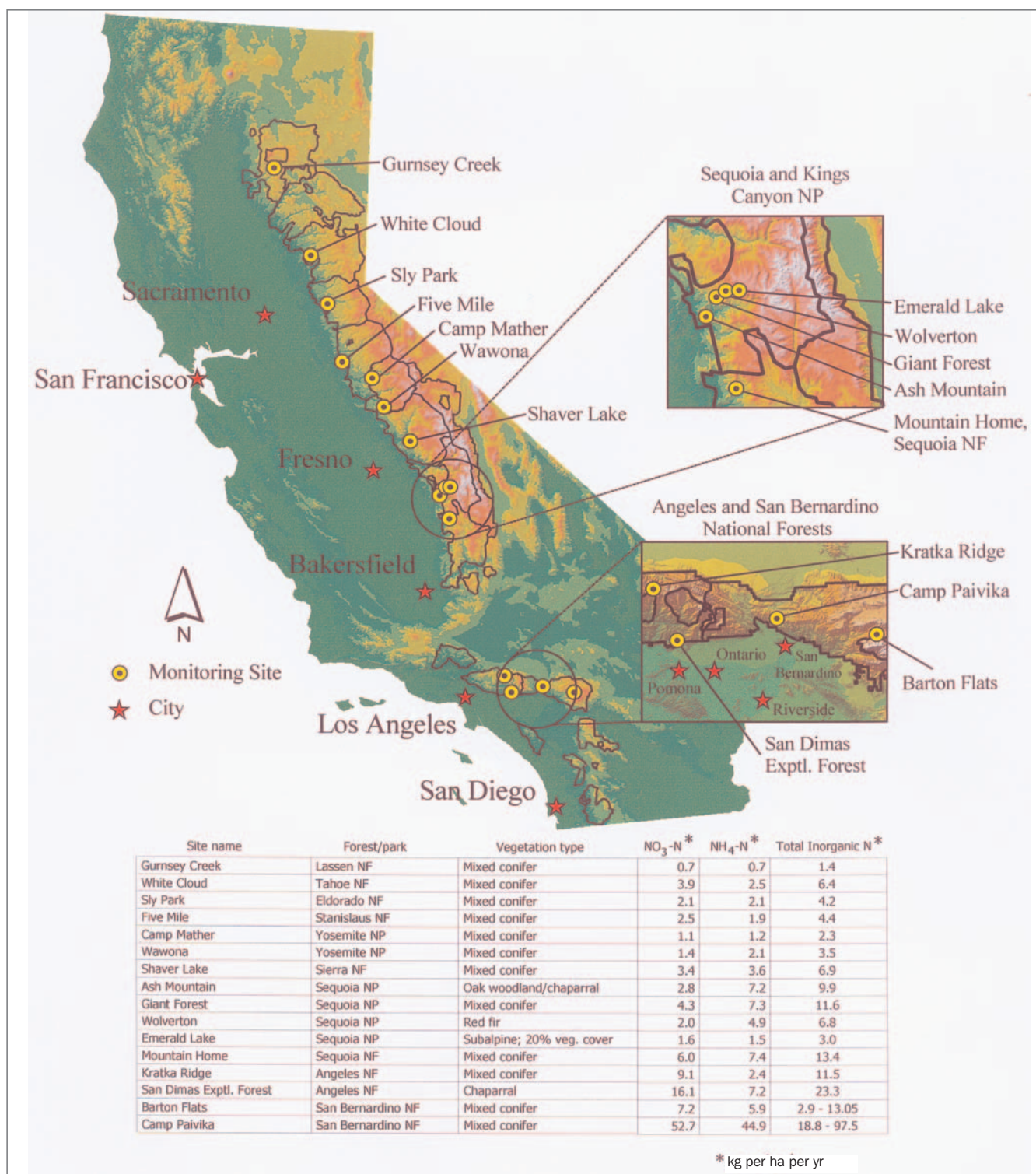


Figure 4. Nitrogen deposition, in kilograms per hectare per year, at sites in the Sierra Nevada and Transverse Ranges of California. Data represent throughfall deposition to mixed conifer forests (1128 to 1920 meters [m] elevation) with a dominant ponderosa pine or Jeffrey pine (*Pinus jeffreyi* Grev. and Balf.) component, except at four locations: (1) Emerald Lake, with deposition to open areas in a subalpine catchment (2800 m elevation); (2) Ash Mountain, an oak woodland (750 m elevation); (3) Wolverton, mainly red fir (*Abies magnifica* A. Murr.), above the mixed conifer zone (2207 m elevation); and (4) San Dimas Experimental Forest, chaparral (800 m elevation). Data are from 2000–2001, except for the lower values at Barton Flats and Camp Paivika, which are from 1996 (Fenn et al. 2000). Abbreviations: N, nitrogen; NF, national forest; NH₄⁺–N, nitrogen from ammonium; NO₃–N, nitrogen from nitrate; NP, national park.

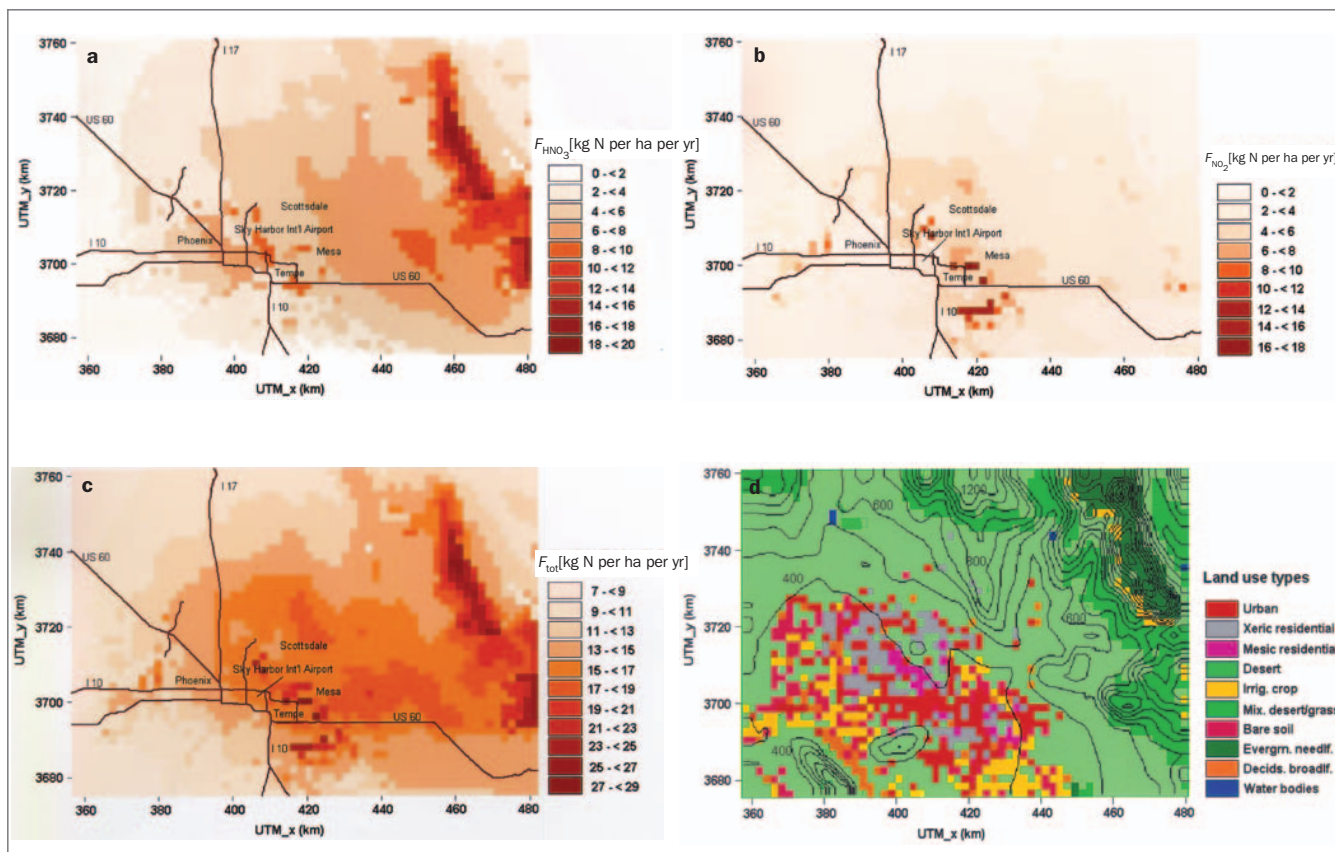


Figure 5. Estimated annual nitrogen (N) deposition (kilograms per hectare per year), as (a) nitric acid (HNO_3), (b) nitrogen dioxide (NO_2), and (c) total N, and (d) land-use patterns in the Central Arizona–Phoenix Long Term Ecological Research study area. Deposition inputs were determined by a combination of measurements and model simulations. Coordinates of the modeling domain are shown in Universal Transverse Mercator (UTM) Zone 12 projection.

Proximity to an urban area is the most important variable predicting an increase in wet deposition of NH_4^+ and NO_3^- .

Transpacific nitrogen transport and deposition to the West Coast of North America

From the mid-1970s to the 1990s, emissions of NO_x and SO_2 from the East Asian region increased at an average rate of approximately 4% per year (Akimoto and Narita 1994), approximately doubling over a period of 20 years. Emissions of N and sulfur from Asia now exceed those from North America, and these emissions can be transported across the Pacific in 6 to 10 days. Peroxyacetyl nitrate, with a lifetime of days to weeks, can be transported across the Pacific on westerly winds (Jaffe et al. 1999), which prevail at mid-latitudes most of the year. This transpacific transport is episodic and can occur either in the atmospheric boundary layer or in the free troposphere (Jaffe et al. 2003). For a number of pollutants, including PAN, higher concentrations were measured on the coast of Washington in 1998, a year with substantially elevated biomass burning in Asia (Jaffe et al. 2001). Although natural sources of fixed N also occur, most N found in the North Pacific atmosphere is likely to be of anthropogenic origin (Prospero and Savoie 1989, Horowitz and Jacob 1999).

Because of the short atmospheric lifetime of NO_x , on the order of 12 to 24 hours, conversion to a longer-lived form is necessary for long-range transport. NO_x is primarily transported as PAN. Once in the free troposphere, PAN can be transported long distances and later moved back into the warmer boundary layer, where it regenerates NO_x and subsequently HNO_3 (Moxim et al. 1996, Kotchenruther et al. 2001a). Given that long-range transport of PAN from Asia to North America is well documented (Jaffe et al. 1999, Kotchenruther et al. 2001b), Asian NO_x emissions almost certainly contribute to the background NO_3^- deposition on the West Coast. The contribution of Asian emissions to the background deposition in the Pacific Northwest is somewhat difficult to ascertain, but is perhaps in the range of 25% to 50%. It is probably not much larger, because global sources of NO_x contribute to a hemispheric accumulation of PAN (Moxim et al. 1996), and PAN decomposition is an important source of NO_3^- in remote regions of the Northern Hemisphere. On the other hand, it is probably not much less, given the quantities of Asian pollutants transported directly to North America. Furthermore, Asian sources contribute approximately 33% of the total carbon monoxide in the background air arriving to the Pacific Northwest in the spring (Jaeglé et al.

2003). Studies are fairly consistent in predicting that Asian NO_x emissions will double again in the next 20 to 30 years (Streets and Waldhoff 2000, Klimont et al. 2001), resulting in an increase in the background NO_3^- deposition in western North America of between 40% and 100% from 1980 to 2020.

Regional haze and visibility impairment

The IMPROVE network was established in response to the Clean Air Act of 1977. The goal of the network is to measure the key aerosol species that are principally responsible for visibility reduction in federal Class I areas. These measurements can be used to establish current conditions, to determine which species contribute to visibility reduction, and to monitor long-term trends toward the national visibility goal of no man-made impairment of protected areas. Since March 1988, the IMPROVE network has sampled fine (diameter < 2.5 micrometers) NO_3^- and other constituents of fine mass in or near select federal Class I areas. We present data for 1997–1999 (figure 6) from 35 sites west of the 100th meridian (9 March 2003; <http://vista.cira.colostate.edu/improve>). These data include as many of the newer sites as possible while excluding the effects of protocol changes from before 1997. Concentrations of atmospheric NO_3^- in the IMPROVE data are probably underestimates of total NO_x concentrations by a factor of 1.5 to 3.0, presumably because HNO_3 and NO_3^- in the coarse particulate fraction are not measured (Ames and Malm 2001).

Visibility impairment caused by regional haze is common throughout the United States. During the 1990s, federal land managers certified visibility impairment in all national parks and some wilderness areas (Watson 2002). As states begin studying implementation strategies to comply with the regional haze regulation (EPA 1999), nitrogenous species in particular will be important for two reasons: First, they contribute considerably to fine mass and visibility impairment in many areas, and second, nitrogenous species are predominantly anthropogenic (Trijonis 1990). The two major chemical species that account for most of the fine particle N are NO_3^- and NH_4^+ . The IMPROVE network directly measures only NO_3^- ; however, a reasonable upper bound for NH_4^+ in the western United States can be inferred from the data by assuming that fine particulate NO_3^- and sulfate are fully neutralized as ammonium nitrate and ammonium sulfate. This technique provides reasonable results at the few sites where NH_4^+ is measured.

Annual average fine NO_3^- concentrations (expressed as NO_3^- ion) in the western IMPROVE sites vary from 45 nanograms (ng) NO_3^- per cubic meter (m^3) at Denali National

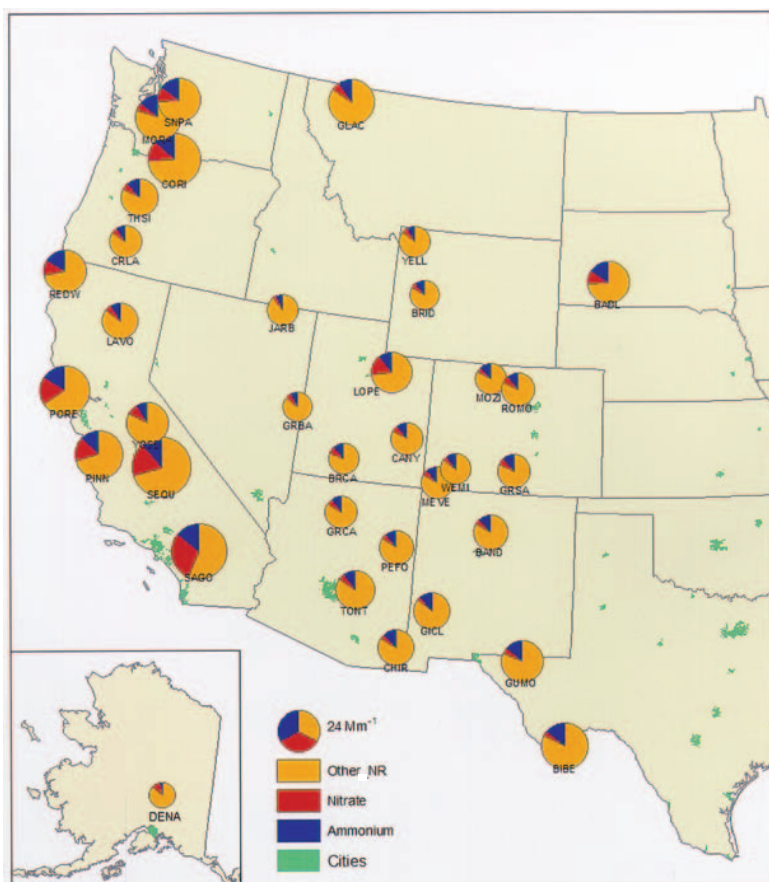


Figure 6. Fraction of reconstructed non-Rayleigh (NR) atmospheric extinction (haze that does not result from atmospheric gases) caused by ammonium, by nitrate, and by other particulate matter. Other particulate matter includes sulfates, organic matter, elemental carbon, fine soil material, and coarse mass. Units for atmospheric extinction are inverse megameters (Mm^{-1}). See Malm and colleagues (2000) for more details of methodology. The area of the circular symbols is proportional to the amount of atmospheric extinction.

Park in Alaska to 2222 ng NO_3^- per m^3 at San Geronio Wilderness in southern California. The highest averages (≥ 400 ng NO_3^- per m^3) are seen at sites that are near large urban areas, such as Point Reyes National Seashore (663 ng NO_3^- per m^3) and Pinnacles National Monument (701 ng NO_3^- per m^3), both near the San Francisco Bay Area. Three sites—Snoqualmie Pass, Washington; Tonto National Monument, Arizona; and Rocky Mountain National Park, Colorado—might be expected to have higher averages because of their proximity to major metropolitan areas, but they do not. Badlands National Park, South Dakota, has anomalously high NO_3^- (326 ng per m^3) for such a remote western site but is characteristic of NO_3^- values for eastern sites. Concentrations from the remaining 24 western continental US sites average about 150 ng NO_3^- per m^3 , an average that could be used as a generic estimate for remote NO_3^- concentrations. By comparison, the 16 sites in the eastern United States have annual

average NO_3^- concentrations ranging from 158 ng NO_3^- per m^3 at Shining Rock Wilderness, North Carolina, to 1148 ng NO_3^- per m^3 at the National Mall in Washington, DC, with an annual mean of about 400 ng NO_3^- per m^3 .

Western annual average estimated NH_4^+ concentrations (expressed as NH_4^+ ion) in fine particulates show considerably less variation than fine NO_3^- , ranging from 139 ng NH_4^+ per m^3 at Denali National Park to 963 ng NH_4^+ per m^3 at San Geronio Wilderness. Proximity to urban areas does not appear to be as important a factor for NH_4^+ as for NO_3^- . The mean value for all 35 western sites is 376 ng NH_4^+ per m^3 , compared with the eastern mean of 1386 ng NH_4^+ per m^3 . Low-elevation sites tend to have higher NO_3^- and NH_4^+ concentrations than higher sites. Nitrogenous species account for as little as 11% of annual average fine mass at Glacier National Park, Montana, and Crater Lake National Park, Oregon, and as much as 35% at San Geronio Wilderness. Patterns for the fraction contributed by nitrogenous species to fine mass are virtually identical to those seen in figure 6 for the contribution of the same nitrogenous species to particulate extinction.

Standard visual range, the distance at which a large dark object is barely visible against the horizon, is inversely proportional to atmospheric extinction. As atmospheric pollutants like ammonium nitrate increase, atmospheric extinction increases and standard visual range decreases (i.e., a vista becomes more hazy). Atmospheric extinction attributed to N ranges from 44% of annual mean particulate extinction at San Geronio Wilderness to 11% of particulate extinction at Jarbidge Wilderness, Nevada (figure 6; see the IMPROVE Web site [<http://vista.cira.colostate.edu/improve/>] for more complete site names and information). Nitrogenous species contribute about 20% of particulate extinction on an annual average basis in the western United States, compared with about 26% in the eastern United States. The lowest individual daily fractions of extinction caused by nitrogenous species are below 1% and are seen at very remote sites like Crater Lake National Park and Denali National Park. The highest individual daily fractions of extinction caused by nitrogenous species are more than 70% of reconstructed extinction and are seen at three California sites: Sequoia National Park, San Geronio Wilderness, and Point Reyes National Seashore.

Current policy and regulatory framework

Clearly, the impacts of N deposition to western ecosystems are important issues. However, issues such as the human health impacts of atmospheric concentrations of O_3 and particulate matter are more traditional policy drivers for NO_x emissions reduction efforts. In the western United States, air quality policy has focused on issues such as regional haze and decreasing visibility in Class I areas, urban air quality in major cities, and O_3 levels. While the ecological consequences of N deposition are important concerns, the air policy community knows less about these consequences and their magnitude in the West than about concerns such as visibility and human health. As a result, ecological issues have not been prominent policy drivers in the West.

Title IV of the Clean Air Act Amendments of 1990 included provisions to decrease NO_x emissions from stationary power generation sources by 2 million tons from levels projected to be reached in 2000. While Title IV was primarily designed to address the ecological effects of acidic deposition in acid-sensitive regions of the United States, the absence of a cap on NO_x emissions made its NO_x provisions less stringent than its provisions for SO_2 . In some measure, this difference was a result of the paucity of knowledge on the contribution of N deposition to acid rain and other ecological and human health effects when Title IV was passed. With the exception of Title IV and, to some extent, the National Ambient Air Quality Standards, many current regulatory efforts to decrease NO_x emissions and N deposition, particularly from stationary sources, focus on the eastern United States and are unlikely to address N deposition levels in the western United States. (For a summary of regulations affecting NO_x emissions from stationary and mobile sources, see EPA 2000, table III-6.)

Still, there are important regulations in place that promise to achieve decreases in NO_x emissions and N deposition in the western United States. While these efforts are aimed at improving air quality, the resulting decline in NO_x emissions will provide corollary ecological benefits. Perhaps the most important of these efforts are the recently adopted Tier II tailpipe emissions standards for cars, sport utility vehicles, and light-duty trucks (Driscoll et al. 2003). In addition to the Tier II standards, the Environmental Protection Agency (EPA) has promulgated similarly tough standards for diesel trucks. In September 2002, the EPA also announced its intention to reduce pollutants from several groups of non-road engines, including large industrial engines, snowmobiles, and all-terrain vehicles. When fully implemented, these non-road vehicle standards will remove more than 1 million tons of hydrocarbons and NO_x each year.

In 1977, Congress set a national visibility goal aimed at "the prevention of any future, and remedying of any existing, impairment of visibility in mandatory Class I Federal areas which impairment results from manmade air pollution" (42 USC 7491 [a][1]). The 1999 regional haze rule (EPA 1999), proposed by the EPA to address visibility issues, may have implications for NO_x emissions and N deposition in the western United States. The rule is designed to improve visibility in 156 Class I areas, including important western sites such as Yellowstone, Yosemite, and Grand Canyon National Parks. States are required to develop state implementation plans to reduce human-generated emissions of SO_2 and NO_x from prescribed fires and from mobile, stationary, and area sources, to reach natural background visibility conditions within 60 years. A major provision directs states to establish emissions limits for older large stationary sources (e.g., power plants, smelters, and refineries) that can be achieved by installing the best available retrofit technology (BART) or alternative measures (e.g., emission trading programs) if such measures promise greater visibility improvements than source-by-source BART controls.

The 1977 amendments to the Clean Air Act also empowered federal land management agencies to help prevent significant air quality deterioration in regions of the country with good air quality. Established by sections 160–169 of the 1977 amendments, the Prevention of Significant Deterioration program specifies that Class I areas should receive the most stringent degree of air quality protection. During the permitting process that precedes construction of major new pollution sources or major modifications of existing sources, federal land managers are directed to consider whether “air quality related values” will be adversely affected by facility construction. If the facility will have adverse impacts on these values in Class I areas, then state or federal action, and possibly both, must be taken to minimize the impacts.

Emissions of NO_x are not the only source of N deposition in the western United States. Other forms of airborne N, such as NH_3 and NH_4^+ , are also of concern. Concentrated animal feeding operations are a large source of these N forms, contributing to atmospheric N deposition through volatilization from waste lagoons and land-based waste application. These operations are regulated at both state and federal levels. Existing regulations focus on controlling effluents from CAFOs directly to surface water and groundwater, though current and proposed CAFO regulations may have some corollary benefits in terms of decreasing atmospheric N sources.

Priority research needs

Improved understanding of western N sources, deposition patterns, and effects will support the development of better policy and management. Efforts to improve our understanding are particularly timely in the context of the current debate over future air quality policy. Over the last several years, various legislative proposals to curb emissions of air pollutants have been introduced in both houses of Congress. These proposals vary in terms of the pollutants addressed, the levels of reductions required, and the timing of these reductions. A common element of the proposals is the imposition of a mandatory cap on NO_x emissions from the power generation sector. However, because cap levels, timing, and other aspects of NO_x emissions reduction vary among proposals, results in the West may differ from one proposal to the next. The information provided through syntheses such as this one are essential in helping policymakers understand and address N-related issues in the West as the policy debate continues. Ongoing research and monitoring efforts will also be crucial in helping evaluate policy changes to determine whether they have achieved environmental goals.

In the western United States, more complete atmospheric monitoring data are needed, including data on N inputs from dry deposition and fog deposition. Deposition data are needed at additional key sites in the West, located within the zone of influence of major emissions source areas. National monitoring networks emphasize remote wilderness and national park areas to the exclusion of other sites closer to major emissions sources. Unless these networks are augmented with additional monitoring sites, data and research on N de-

position and its ecological effects will be skewed away from areas where impacts are likely to be more prevalent and severe (although ecological effects are often observed with relatively low atmospheric deposition levels in remote high-elevation watersheds). In addition, better emissions inventories are needed in many areas. These should include not only major point sources but also mobile sources of all kinds (including non-road sources), combined small area sources, and agricultural emissions from fertilizers and animal husbandry. The NH_3 emissions inventory has the highest level of uncertainty because NH_3 is an unregulated pollutant. Development of emissions factors and emission models for NH_3 is of the highest priority, particularly because NH_3 emissions are projected to increase in the future. The NH_3 inventory affects both the total mass of N and the partitioning of N through the NH_3 – HNO_3 – H_2SO_4 (nitrate–nitric acid–sulfuric acid) equilibrium, which in turn affects the partitioning and lifetime of NO_y . Inventories need to be updated regularly to determine trends and the effectiveness of regulatory controls.

Conclusions

The major N emissions sources in the West are, in order of decreasing importance, transportation, agriculture, and industry. Transpacific N transport also contributes an unknown amount of N deposition along the West Coast. Nitrogen deposition seems to be on the rise in recent years in most areas of the West affected by urban and agricultural emissions, because population and distance driven in these areas have increased. Total NO_x emissions in the western states are projected to decrease slightly by 2018 because of stricter tailpipe emissions standards and the gradual phase-in of newer, cleaner vehicles. By contrast, NH_3 emissions from area sources are projected to increase by 2018. Because of regional variability in activity and in population growth, total N emissions may increase in some areas and decrease in others.

The highest known N deposition levels for wildland areas in the West are at forest and shrubland sites in the Los Angeles air basin (typically 25 to 45 kg per ha per year, although these rates can double in years with high fog deposition) and desert sites in the Phoenix area (29 kg per ha per year), while large areas of the West with sparse populations have low deposition rates. Deposition to the arid ecosystems downwind of Phoenix may exemplify similar situations of high-deposition exposure downwind of other urban regions in the West, but this possibility has not yet been investigated. Current national deposition monitoring networks do not adequately characterize total N deposition levels or identify where the highest deposition rates occur. This is largely because dry deposition and fog deposition to canopies are not accounted for and because monitoring sites are located in sites remote from emissions sources.

Decreasing N emissions will have the triple benefit of decreasing concentrations of tropospheric O_3 (a widespread phytotoxic pollutant), reducing the eutrophying impacts of elevated N deposition in aquatic and terrestrial ecosystems,

and reducing fine particulate pollution and regional haze. This will result in considerable benefits for human and ecosystem health. Legislation that caps and further decreases NO_x emissions from power generation sources will help in achieving these benefits. However, because power generation is not the major source of NO_x emissions in the western United States, controlling emissions from this sector is only one component of an integrated solution. To make significant improvements in air quality and to reduce N deposition inputs and effects on sensitive ecosystems, reductions of NO_x emissions from on-road and non-road mobile emissions sources will be required. Finally, NH_x emissions from agriculture are an important source of N in some areas, although these emissions are not currently regulated.

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