National Acid Precipitation Assessment Program Report to Congress 2011:

An Integrated Assessment

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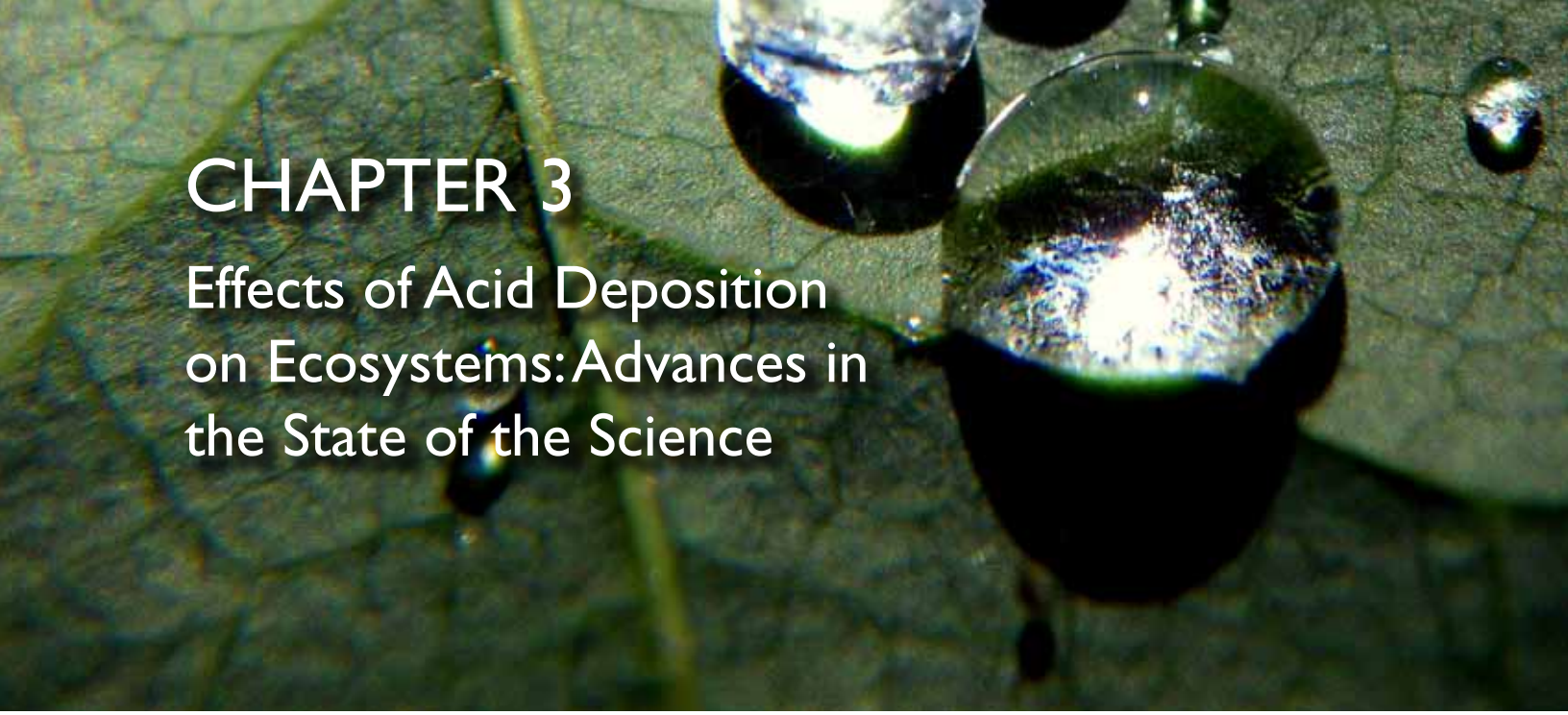

CHAPTER 3

Effects of Acid Deposition on Ecosystems: Advances in the State of the Science

Chapter 2 focused on the environmental results of the ARP, presenting data from national monitoring networks on SO_2 and NO_x emissions, air quality, atmospheric deposition, surface water chemistry, and visibility. This chapter expands on this information by examining the most recent research into how ecosystems respond to acid deposition, especially the processes that control the recovery of ecosystems as acid deposition decreases.

In Chapter 2, two general trends were discussed regarding the current recovery status of affected ecosystems: (1) these ecosystems are trending generally towards recovery, but improvements in ecosystem condition shown by surface water chemistry monitoring data thus far have been less than the improvements in deposition; and (2) ecosystem impacts and trends vary widely by geographic region, but the evidence of improvement is strongest and most evident in the Northeast. These trends are not uniform across the United States, however, and in some regions (e.g., central Appalachian Mountain region), trends in improved water quality are generally not evident.

Despite the strong link in many areas between reduced emissions and reduced acidity of atmospheric deposition, the link is less clear between reduced acidity and recovery of the biological communities that live in aquatic and terrestrial ecosystems that have experienced

State of the Science at a Glance

- Scientists have observed delays in ecosystem recovery in the eastern United States, despite decreases in emissions and deposition over the past 30 years.
- A growing body of literature documents impacts of nitrogen deposition on western ecosystems.
- "Critical loads" are increasingly used by scientists as a tool for quantifying the sulfur and nitrogen deposition levels at which ecosystems are impacted in order to better inform air quality policies.
- Recent literature identifies linkages between a changing climate, an altered carbon cycle, and ecosystem response to acid deposition as an important emerging area of scientific investigation.
- Scientists identify multi-pollutant interactions, including reactions among SO_2, NO_x, ozone, and mercury, as an under-studied area of ecosystem impacts, which if better understood, would likely have implications for future air pollutant policies.

deleterious effects from acid deposition. The recovery of these communities is proceeding at a slower pace than, for example, the improvements in stream and lake ANC would indicate. The goal of this chapter is to synthesize the science in a weight-of-evidence manner to provide policy makers with tangible evidence and likely causative factors regarding ecosystem status and recovery patterns to date. This chapter serves as an update to the 2005 NAPAP RTC (NSTC, 2005), with an emphasis on scientific studies and monitoring since 2003, which was the last year for consideration of research results in the 2005 report. Several issues pertinent to ecosystem response to emission controls and acid deposition are receiving increasing attention in the scientific literature and will be discussed in this chapter, including the (1) observed delay in ecosystem recovery in the eastern United States, even with decreases in emissions and deposition over the past 30 years; (2) emerging ecosystem impacts of nitrogen deposition in the western United States; (3) the application of critical deposition loads as a tool for scientists to better inform air quality policies; (4) the role of changes in climate and the carbon cycle as factors that affect the response of ecosystems to acid deposition; and (5) the interaction of multiple pollutants in ecosystems. Throughout this chapter, the value of long-term environmental monitoring data in informing air quality policy will be highlighted, including the limitations of assessing the current status of some ecosystem indicators for which continuous, long-term data are lacking.

Effects of Atmospheric Deposition on Aquatic and Terrestrial Ecosystems

This section expands on the surface water chemistry section of Chapter 2 and describes the latest studies on the spatial and temporal patterns of recovery in a variety of ecosystems that are sensitive to acid deposition. Factors that control ecosystem sensitivity to deposition are also discussed, with an emphasis on the links between terrestrial and aquatic ecosystems. This section first considers the eastern United States, then the Rocky Mountain region, the far western United States, and finally the coastal areas of the United States. This is a logical division because the effects of atmospheric deposition and issues discussed by scientists and policy makers are somewhat different across these regions and ecosystems.

Ecosystem Recovery in Aquatic and Terrestrial Ecosystems of the Eastern United States

The regions of the United States with an abundance of acid-sensitive waters and ecosystems were well recognized by the end of the 1980s during the decade of intensive research performed under the original NAPAP. These acid-sensitive ecosystems are generally located in upland, mountainous terrain in the eastern United States and are underlain by bedrock that is resistant to weathering, such as granite or quartzite sandstone. The following sections examine trends in surface water chemistry; the impacts of episodic acidification on surface waters; the effects of stored sulfate in the soils of the Southeast; base cation depletion in soils; and how aquatic and terrestrial ecosystems recover from acidifying deposition.

Patterns in Surface Water Chemistry and Soils

Monitoring of lakes and stream water chemistry in the eastern United States has shown the continuation of widespread water quality improvements that are consistent with a trend towards recovery in these aquatic ecosystems (Kahl et al., 2004; Driscoll et al., 2007). Decreasing

Nitrate Concentration Trends

Trends in nitrate concentrations in headwater surface waters are influenced by natural disturbances such as insect defoliation and ice storms, as well as human disturbances such as forest harvesting, and are therefore not as clearly linked to trends in nitrogen deposition as are those of sulfur. Nonetheless, over the long term, decreases in NO_x emissions from the power generation sector are expected to assist in efforts to reduce nitrate concentrations in surface waters.

Aluminum (i.e., Al^{2+} and Al^{3+}) levels in surface water are monitored because acidifying deposition releases aluminum from soils and high concentrations of aluminum are toxic to many aquatic biota.

trends in sulfate concentrations have been the sharpest, generally showing decreases of about 2 to 3 μeq/L/yr since the early 1990s in waters in the Northeast and the Northern Appalachian Plateau. These trends are consistent with the large decreases in SO_2 emissions and sulfur deposition and are paralleled by some improvement in pH, ANC, and aluminum (i.e., Al^{2+} and Al^{3+}) concentrations. However, the improvements in pH, ANC, and aluminum concentrations have generally been much smaller than the decreases in sulfate concentrations. Nitrate concentrations in these surface waters have also shown smaller decreases that have been less persistent and less consistent than those of sulfate concentrations (Kahl et al., 2004; Burns et al., 2005; Driscoll et al., 2007). The most recent trends in surface water chemistry from the EPA LTM monitoring program are reported in Chapter 2 and shown in Table 2-3. These trends are generally consistent with those reported in the studies cited here, indicating sharp decreases in sulfate concentrations, some modest decreases in nitrate concentrations, and increases in ANC in the eastern United States. Streams in the central Appalachian region show the weakest trends in recovery. At least over periods of a decade or more, trends in nitrate concentrations in surface waters are not as clearly linked to trends in nitrogen deposition as trends in sulfate concentrations in surface waters are linked to trends in sulfur deposition (Aber et al., 2002; Burns et al., 2006; Eshleman et al., 2009). Nonetheless, if temporal trends follow observed spatial patterns of nitrogen deposition and surface water nitrate concentrations over the long term, then decreases in these nitrate concentrations are expected to eventually follow decreases in nitrogen deposition (Aber et al., 2003).

Surface water chemistry monitoring programs that document the effects of acid deposition tend to collect samples at regular time intervals, resulting in data that primarily reflect baseflow or non-storm conditions (although there are exceptions). The shortcoming of such a sampling strategy is that surface waters, especially streams and small, shallow lakes, tend to acidify during rainstorms and snowmelt. This phenomenon is termed episodic acidification and has been well documented (Lawrence, 2002; Deviney et al., 2006). Additionally, episodic acidification has been shown to affect surface water biota and to slow ecosystem recovery in waters that otherwise appear to be improving based on data collected at low-flow conditions (Passy et al., 2006; Kowalik et al., 2007). These findings suggest that biological recovery will hinge in part on the acidity of surface waters declining to a level at which minimal episodic acidification occurs because the survival of acid-sensitive aquatic species is dependent on both the magnitude and duration of episodic acidification (Baldigo and Murdoch, 1997). Although low-flow ANC values in the range of 30 to 50 μeq/L have been suggested to provide protection against acidification and biological harm during high-flow periods (Driscoll et al., 2001; Kahl et al., 2004), other studies have shown that episodic acidification occurs in streams with low-flow ANC values as high as 100 to 150 μeq/L (Lawrence, 2002). Some evaluations of ecosystem recovery (e.g., Kahl et al., 2004) provide percent-improvement values for various metrics of stream acidity and the extent of acidic waters in a region. These evaluations are based largely on low-flow conditions or chronic acidification scenarios and do not fully consider episodic acidification. One study in Maryland

The Laurel Prong fork of the Rapidan River in Virginia (photo courtesy of U.S. EPA).

evaluated changes in episodic stream acidification from the early 1990s to the late 1990s and found little change in the magnitude of acidification during storms. However, the study also found a shift towards a diminished role for sulfate in episodic acidification, suggesting a decreasing contribution of acid deposition to this phenomenon in parallel with declining sulfate deposition (Kline et al., 2007). Studies by Rice et al. (2006) and Lawrence et al. (2008) compared the percent of streams or watershed area that is chronically acidified with the percent that is episodically acidified in the Shenandoah National Park, VA, and the western Adirondack Mountains, NY, respectively. In each case, the total percentage of acidified streams or watershed area increased substantially when episodic acidification was considered (see Table 3-1).

One noteworthy exception to the generalizations above about geographic patterns of improvement in surface water chemistry is watersheds in the Southeast (e.g., the central Appalachian region of Virginia) where stream ANC values have generally not improved and sulfate concentrations have shown little change since the early 1990s (Kahl et al., 2004; Webb et al., 2004; U.S. EPA, 2009a; Chapter 2 of this report). The lack of chemical recovery in the streams of the central Appalachian region is believed to be due in part to the old and highly weathered condition of non-glaciated southern soils. These soils contain an abundance of adsorbed sulfate that can be slowly released as sulfuric acid deposition decreases, slowing ecosystem recovery (Webb et al., 2004). In these soils, decades of sulfate release and decreased deposition levels may be necessary to shrink the pool of sulfate in the soils to the point where acidification of streams no longer occurs. Ecosystem recovery of acidified watersheds in the highly weathered soils of the Southeast will likely continue to lag behind that of the glaciated Northeast by several years to several decades.

Recent research has shown that decreases in base cations (e.g., calcium, magnesium, potassium, sodium) in soils may also be slowing the recovery of aquatic and terrestrial ecosystems even though they are receiving reduced loads of acid deposition (Bailey et al., 2005; Sullivan et al., 2006b; Johnson et al., 2008; Warby et al., 2009). The base cations calcium, magnesium, and potassium are important nutrients for the growth and health of forest vegetation and are readily accessed from the pool of cations held on the surfaces of soil particles. These base cations can be displaced from the surfaces of soil particles by hydrogen and aluminum ions and leached into local waters, effectively removing these nutrients from soils and lowering the levels that are available to the plants growing in the

> Ecosystem recovery of acidified watersheds in highly weathered soils of the Southeast will likely continue to lag behind that of the Northeast by several years or, more likely, decades.

Table 3-1. Comparison of Chronically and Episodically Acidified Streams in a Watershed

Location	Percent of Streams or Watershed Area That is Chronically Acidic	Percent of Streams or Watershed Area That is Episodically Acidic	Total Percent of Streams or Watershed Area That is Chronically and Episodically Acidic	Reference
Shenandoah National Park, VA[a]	8.7	22.5	31.2	Rice et al., 2006
Western Adirondack Mountains, NY[b]	35	30	65	Lawrence et al., 2008

[a] Chronic acidification is defined as the percent of watershed area with ANC < 0 μeq/L for at least 168 successive hours at a 4-year recurrence interval, and episodic acidification is defined as the percent of watershed area with ANC < 20 μeq/L for at least 72 successive hours at a 4-year recurrence interval.

[b] Chronic acidification is defined as percent of streams with a base cation surplus (a surrogate for ANC that eliminates the contribution from dissolved organic carbon [DOC]) value < 25 μeq/L during summer, and episodic acidification is defined by the same threshold value during snowmelt conditions.

soils. Acid deposition accelerates this as part of the acidification process, which initially leads to increases in base cation concentrations in surface waters, but eventually acidifies the waters as well. Base cation depletion refers to this accelerated loss of these cations from soils. Depletion of calcium is of particular concern because this cation is in high demand by sugar maple (St. Clair et al., 2008). Although a variety of mechanisms (e.g., maturation of forest vegetation) contribute to base cation depletion, acid deposition and the consequent mobilization of aluminum in soils have been identified as important drivers of depletion (Lawrence et al., 1995). Acid deposition and aluminum mobilization have contributed to dieback and decline of sugar maple (Bailey et al., 2004; Hallet et al., 2006; St. Clair et al., 2008) and to freezing injury in red spruce (DeHayes et al., 1999; Hawley et al., 2006). Recent work has found that declining growth rates can begin in apparently healthy sugar maple trees up to two decades before a strong decline in tree health becomes visually evident (Long et al., 2009). Other studies have found linkages between acid deposition, soil calcium availability, and the reproductive success of bird species (Graveland, 1998; Hames et al., 2002).

The ANC value of a waterbody reflects the sum of the base cation concentrations (calcium, magnesium, potassium, sodium) minus the sum of the acid anion concentrations (primarily sulfate and nitrate). Depletion of base cations from soils over time will provide lower concentrations of these base cations to waters and result in a lower ANC value than would otherwise be predicted based on the acid anion concentrations alone. In this way, losses of exchangeable soil base cations can slow the recovery of aquatic ecosystems (Likens et al., 1996; Lawrence et al., 1999). The scientific consensus is that base cation depletion is slowing the rate of chemical recovery of lakes and streams and, therefore, also the rate of biological recovery in many of the sensitive aquatic ecosystems targeted by Title IV of the CAAA (Driscoll et al., 2001). Re-supply of these base cations generally occurs when chemical breakdown (i.e., weathering) of minerals in bedrock begins to release cations to soils and waters at a rate that is faster than the

Sugar maple showing brilliant fall colors (photo courtesy of Jacquie Tinkler, NPS).

leaching loss caused by acid deposition. Acid deposition must likely decrease below a critical level that will vary among ecosystem locations before large increases in the ANC of acidified waters can occur. The role of base cation depletion is even greater when considering episodic acidification because soil chemistry plays an even greater role in stream chemistry during high-flow conditions than during low-flow conditions (Lawrence, 2002).

Important complications to this simplified conceptualization of the link between acid deposition and base cation depletion arise from the uptake of base cations by vegetation and from other factors. For example, the cation balance is also controlled in part by forest age, health, and root-mining ability, as well as harvesting practices (Blum et al., 2002; Hamburg et al., 2003; Grigal and

Base cation depletion is a high-priority area for future acidification research and is of high relevance to the development of future air quality policies designed to protect ecosystems.

Ohmann, 2005). The exact rate of acid deposition that will allow recovery of base cation–depleted soils is not well known because the rate of mineral weathering is not well quantified in most soils, can be highly variable among locations, and is difficult to measure in the field (Bailey et al., 2003). Current models (such as the Model of Acidification of Groundwater in Catchments [MAGIC]) that are commonly applied to make quantitative predictions about acidification and critical loads in sensitive ecosystems can be, and have been, used to examine the role of base-cation depletion; however, they have not yet been applied widely for this purpose in the United States. Base-cation depletion is a high-priority area for future acidification research and is of great relevance to the development of future air quality policies designed to protect ecosystems.

Recovery of Biota

Aquatic Recovery

Surface water chemistry data commonly have been favored as an ecosystem indicator of acidification because these data are less expensive and more straightforward to obtain than data on aquatic biological populations. In addition, many studies have shown thresholds that link water chemistry values to the survival and reproductive success of organisms, such as brook trout, mayfly, and many others (Schofield, 1976; Baker and Christensen, 1991). Using surface water chemistry data alone to assess ecosystem acidification assumes that when water chemistry degrades below a certain threshold, acid-sensitive organisms will be replaced by acid-tolerant organisms, and that when the water chemistry improves to a chemical threshold capable of successfully supporting more acid-sensitive organisms, then the organisms will soon return to the waterbody. However, this is an oversimplified view of how ecosystems will respond to changes in lake or stream water quality.

Acidification results in changes not only to the constituent or habitat that may be targeted as a threshold indicator but also to a wide variety of other chemical constituents and physical habitats. As a result, a complex set of changes in the ecological community may occur, including a shift in the members of the community from acid-sensitive to acid-tolerant species. Acid-tolerant communities that develop in response to acidification may then resist change as conditions become less acidic, limiting the re-establishment of the acid-sensitive species that were originally impacted by acidification (Frost et al., 2006). Other factors may also slow this process of biological recovery, including physical limitations on dispersal, the proximity of species that may have historically resided in the water prior to acidification (Yan et al., 2003; Langford et al., 2009), and calcium concentrations that tend to be lower relative to ANC during ecosystem recovery than during acidification (Jeziorski et al., 2008; Cairns and Yan, 2009). These factors act to delay biological recovery relative to chemical recovery (Snucins, 2003), and some scientists have suggested that active ecosystem restoration (e.g., reintroduction of previously lost species) may be needed to meet the goal of a "pre-acidification" biological community (Snucins and Gunn, 2003; Binks et al., 2005).

> There have been few studies in the United States on the recovery of aquatic biota over a period of gradually improving acidification status; this is largely because none of the existing U.S. monitoring networks regularly provide such evaluations and because most available data originate from ad-hoc studies.

Recreational fishing in South Rolly Lake in Alaska. (photo courtesy U.S. Fish and Wildlife Service).

When a different pathway of change is followed during recovery than was followed during acidification, this behavior is called "hysteresis." A conceptual representation of hypothetical acidification and recovery patterns in a waterbody is presented in Figure 3-1. Point 1 represents the pre-acidification status of water quality and the biological community. Point 2 shows the status when conditions were most acidic, and Point 3 represents current conditions where hysteretic behavior is exhibited because the rate of recovery does not match the rate of acidification. At this point, the water quality has shown some recovery, but a pre-acidification biological community has not yet been achieved. The two dashed lines represent possible future recovery trajectories for the biological community as future acid deposition levels continue to decrease. These recovery patterns may or may not be linear as shown, and some evidence suggests that "threshold" behavior marked by a sharp change in slope with time may occur in biological communities recovering from acidification (Monteith et al., 2005). One trajectory ends at Point 4, where water quality has fully recovered, but the biological community has only recovered slightly. The other trajectory returns to Point 1, representing a full recovery of water quality and the biological community. Some studies indicate that even with full recovery of water quality (less acidic stream on the right side of the figure), the biological community will likely differ from what existed prior to acidification (Frost et al., 2006; Cairns and Yan, 2009), suggesting that recovery may ultimately be closer to Point 4 than Point 1.

Despite the concerns raised about limitations on recovery from acidification due to competition, dispersal, and other ecological factors that must be considered in addition to chemical thresholds, several studies in Europe and Canada have demonstrated biological recovery in aquatic communities that have become less acidic in response to decreases in acid deposition (Tipping et al., 2002; Findlay, 2003; Halvorsen et al., 2003). Additionally, studies in which lime has been added to waterbodies or soils to hasten recovery from acidification have shown trends towards a pre-acidification aquatic community (Raddum and Fjellheim, 2003), but the success of these efforts has sometimes required active restoration of some species (Driscoll et al., 1996; Clair and Hindar, 2005).

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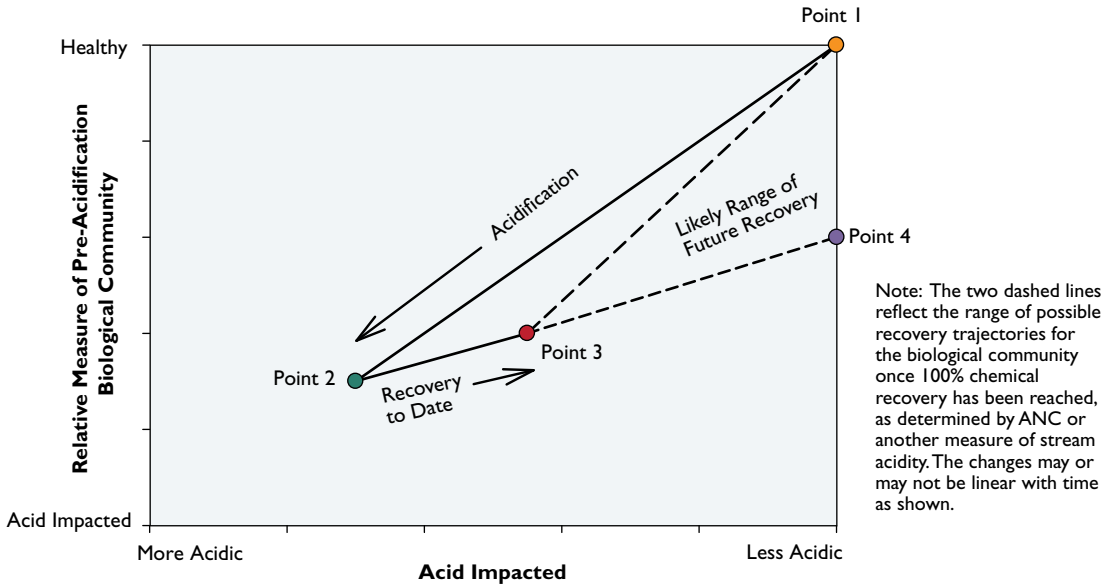


Figure 3-1. Conceptual figure showing hypothetical acidification and recovery patterns in a waterbody that has been acidified by atmospheric deposition (prepared by USGS).

monitoring networks regularly provide such evaluations and because most available data originate from ad-hoc studies. Nonetheless, a few studies have evaluated aquatic communities by sampling waters over a period when acidity was declining, and the general conclusion is that there is little evidence to date of widespread recovery in these communities (Simonin et al., 2005; Burns et al., 2008a; Warren et al., 2008).

Terrestrial Recovery

Few studies have tracked the status and potential recovery of terrestrial ecosystems during the period of decreasing acid deposition since the 1980s. This is due in part to the historical focus on aquatic acidification and a lack of understanding of the terrestrial impacts of acidification at the time that the Clean Air Act was amended in 1990. Since the early 1990s, scientific understanding of terrestrial ecosystem effects of acid deposition has increased greatly, and studies indicate continued degradation of soil base status (calcium and magnesium), as well as continued deleterious effects on sensitive species such as sugar maple and red spruce in the most acid-sensitive regions.

As described previously, several studies have shown the link between acid deposition and losses of base cations from soil. Many of these studies have included data from re-sampled soil locations, generally documenting a lack of improvement or continued degradation of soil base cation status in the Adirondack Mountains and Allegheny Plateau of Pennsylvania (Bailey et al., 2005; Sullivan et al., 2006b; Johnson et al., 2008; Warby et al., 2009). Furthermore, a clear link between the base cation status of soils and the health of sugar maple has been established in recent years (Bailey et al., 2004; Hallet et al., 2006; Long et al., 2009). The role of acidification in red spruce freezing injury was established by the early 1990s (McLaughlin et al., 1993). Recent data have largely confirmed the findings of earlier studies on red spruce (Hawley et al., 2006) and have also found potential effects of acid deposition on other species of forest vegetation (Jenkins et al., 2007; Zaccherio and Finzi, 2007). Currently, no published studies can document an improvement in the status of terrestrial ecosystems

Currently, no published studies can document an improvement in the status of terrestrial ecosystems in the eastern United States relative to acidification effects due to decreased emissions and deposition.

in the eastern United States relative to acidification effects due to decreased emissions and deposition. This conclusion likely results from three factors: (1) few studies have evaluated terrestrial ecosystem health relative to acidification effects over time, (2) soils in the most acid-sensitive regions continue to acidify, and (3) the potential for lags in response in long-lived species such as forest trees.

Current Effects of Atmospheric Deposition on Aquatic and Terrestrial Ecosystems in the Rocky Mountain Region of the United States

As shown in Chapter 2, the Rocky Mountain region of Colorado and adjoining states receives atmospheric deposition levels of sulfur and nitrogen that are generally less than half the deposition levels measured in mountainous parts of the eastern United States. Yet, the Colorado Rockies landscape is generally more sensitive to nutrient over-enrichment from atmospheric nitrogen deposition than landscapes in the eastern United

Central Appalachian basic boulderfield forest, Shenandoah National Park (photo courtesy of Gary P. Fleming, Virginia Department of Conservation and Recreation).

States. This sensitivity is due to the steep slopes, thin or non-existent soil cover, short growing season, and less vegetation per unit landscape area (Clow and Sueker, 2000). Several studies have measured ANC values near or below zero during snowmelt in headwater streams, small headwater lakes, and ephemeral pools, and downward trends in ANC values were evident in some waters through the 1990s (Caine, 1995; Williams and Tonnessen, 2000; Campbell et al., 2004). However, the extent of waters that become episodically acidified in the Colorado Rockies is generally less than in the eastern United States, and no deleterious effects of acidification on fish have been documented.

The character of nitrogen deposition is different in the Colorado Rockies than in the East, with ammonia nitrogen deposition generally exceeding that of nitrate nitrogen.

Atmospheric deposition of nitrogen has been of interest in the Rocky Mountains because both aquatic and terrestrial ecosystems in this region were historically nitrogen limited and have shown species shifts and growth responses at nitrogen deposition levels as low as 1.5 kg/ha/yr (Wolfe et al., 2003; Baron, 2006). Several factors distinguish nitrogen deposition patterns in the Colorado Rockies from those in the eastern United States. First, the character of nitrogen deposition is different in the Colorado Rockies than in the eastern United States. Ammonia nitrogen (i.e., NH_3-N) deposition, which generates greater acidity than does nitrate, generally exceeds that of nitrate nitrogen (i.e., NO_3-N), especially at sites east of the Continental Divide in the Front Range of the Rocky Mountains (Heuer et al., 2000; Burns, 2003; Beem et al., 2010). This pattern is believed to result from the deposition of emissions carried by upslope winds to the Front Range from agricultural sources that are located to the east of the mountains (Baron et al., 2004). As a result, atmospheric nitrogen deposition at many monitoring sites in the Colorado Rockies is not decreasing with decreases in power plant emissions in the western United States. Instead nitrogen loads are still

Loch Vale in Rocky Mountain National Park receives among the highest loads of atmospheric nitrogen deposition in the Front Range of the Colorado Rockies (photo courtesy USGS).

increasing in some western locations. This pattern is especially driven by a tendency towards increasing trends in ammonia deposition (Burns, 2003).

Many effects on terrestrial and aquatic ecosystems from current levels of atmospheric nitrogen deposition have been demonstrated in the Rocky Mountain region. Over the past two to three decades, increases in nitrate concentrations and inorganic nitrogen loads in high-elevation streams and lakes have been documented in a few areas of the Colorado Rockies (Williams and Tonnessen, 2000; Clow et al., 2003). However, throughout a wider range of high-elevation waters in the western United States, trends have been slight enough that other factors such as recent rainfall (Clow et al., 2003) and the melting of glaciers (Baron et al., 2009) can greatly affect nitrate trends. Studies in which historical atmospheric deposition levels were indirectly evaluated through measurements of nitrogen levels in lake sediment cores have concluded that nitrogen deposition over the Colorado Rockies and many western mountain ranges began to increase greatly after the 1950s. This trend is marked by shifts in lake plankton communities from low nutrient species to those indicative of an increased nutrient supply (Wolfe et al., 2003; Saros et al., 2005). The general conclusion across high-elevation waters of

the western United States is that as atmospheric nitrogen deposition increases from low levels of < 2 kg/ha/yr to higher levels, plankton biodiversity tends to decrease, and growth tends to increase as the aquatic-limiting nutrient shifts from nitrogen to phosphorus (Interlandi and Kilham, 2001; Bergstrom and Jansson, 2006). These shifts and changes in plankton communities are assumed to occur at a given location as atmospheric nitrogen deposition increases over time and spatially from regions of low atmospheric nitrogen deposition to regions of high atmospheric nitrogen deposition.

The changes described above for aquatic ecosystems are also paralleled by observed changes in terrestrial plant communities, especially those in high-elevation alpine climatic zones of the Colorado Rockies. Increases in atmospheric nitrogen deposition tend to increase growth and decrease species diversity in alpine plant communities, with sedge and grass communities generally favored over shrubs (Bowman et al., 1995; Seastedt and Vaccaro, 2001). Changes in vegetation nitrogen content of individual species can occur at atmospheric nitrogen deposition levels as low as 4 kg/ha/yr, and whole community changes were observed at values of 10 kg/ha/yr, as determined by nitrogen addition experiments (Bowman et al., 2006).

Together, the results of studies highlight the high sensitivity of aquatic and terrestrial ecosystems in the Rocky Mountain region to atmospheric nitrogen deposition. Deleterious effects are generally observed at deposition levels that are significantly less than those commonly measured in high-elevation ecosystems of the eastern United States. These results are pertinent to the management of ecosystems in this region because of the large number of National Parks (e.g., Rocky Mountain, Grand Teton, Yellowstone) that are managed as Class 1 Wilderness areas under Clean Air Act provisions to prevent significant deterioration of ecosystems (Porter and Johnson, 2007). Thus, a more active Federal role in land management and ecosystem protection from air pollutant deposition may be necessary in the large tracts of Class 1 wilderness land that are present in the west.

Much of the Grand Teton National Park is managed as a Class 1 wilderness area (photo courtesy of NPS).

Current Effects of Atmospheric Nitrogen Deposition on Terrestrial and Aquatic Ecosystems in the Far Western United States

In many areas of the far western United States (lands to the west of the Rocky Mountains, especially the West Coast states), nitrogen loads to terrestrial systems have historically been low compared with loads to terrestrial systems in the eastern United States. The native plant communities (e.g., coastal sage scrub, desert scrub, annual grasslands) are adapted to thrive in low-nitrogen environments. When nitrogen loads to these habitats increase, the native plant and soil communities have difficulty competing with invasive species that are better adapted to more nitrogen-rich environments. In California, nitrogen deposition levels as low as 5–8 kg/ha/yr have major effects on epiphytic lichens (i.e., lichens that grow on trees and shrubs) and contribute to changes in native plant communities by enhancing invasion by exotic annual grasses (Fenn et al., 2003a; Weiss, 2006). In coastal sage scrub, desert scrub, and annual grasslands in California, nitrogen deposition favors non-native grass invasions, which can lead to altered plant communities. In southern California chaparral (Egerton-Warburton et al., 2001) and coastal sage scrub ecosystems (Egerton-Warburton and Allen, 2000), the diversity of the mycorrhizal fungi communities that grow in association with roots also is negatively impacted by nitrogen deposition. Shifts in these plant communities are of additional concern because the invasive vegetation that results

from increased nitrogen loads also may increase the risk of wildfires due to increased fuel availability. In the desert scrub ecosystems of Joshua Tree National Park, invasive grasses can provide sufficient fuel load to carry fire, particularly in a wet year. This increased fuel load puts the Park's native vegetation at risk, including its namesake, the Joshua tree. Fire occurrence in the desert is a rare event in the absence of the invasive grasses (Allen et al., 2009).

Increased nitrogen deposition also can impact forests in the far western United States. Nitrogen deposition effects, in combination with the physiological effects of ozone, decrease the amount of roots and increase susceptibility of ponderosa pine to bark beetle attack and mortality (Jones et al., 2004). Forest sustainability is thus threatened, and fire risk is increased (Grulke et al., 2009). Studies have been conducted to assess various methods of reducing nitrogen saturation symptoms. Using fire as a management technique to reduce excess nitrogen was found to be ineffective in chaparral ecosystems, presumably because of the limited capacity of fire to reduce the large nitrogen stores in the mineral soil of these ecosystems (Meixner et al., 2006). Similarly, in forests affected by excess nitrogen, a combination of periodic nitrogen release by fire and significant reductions in nitrogen deposition are needed to mitigate the effects of nitrogen accumulated from long-term nitrogen deposition (Gimeno et al., 2009).

Nitrogen deposition effects on the lichen community are pronounced and widespread in the forests, oak woodlands, and chaparral of California, as evidenced by major shifts from dominance of lichen species that thrive in low-acid, low-nitrogen environments to communities dominated by lichen species that thrive in acidic, high-nitrogen environments (Fenn et al., 2008; Jovan, 2008; Jovan and McCune, 2005). Similar effects of nitrogen in lichen communities are well documented in forests near urban and agricultural emissions source regions in Washington and Oregon and in the Columbia River Gorge corridor (Fenn et al., 2007; Geiser and Neitlich, 2007).

In the streams and lakes of the far western United States, increased nitrogen deposition can alter lake and stream chemistry. At moderate nitrogen

Desert scrub ecosystem in Joshua Tree National Park may be impacted by nitrogen deposition (photo courtesy of USGS).

deposition levels of 10–14 kg/ha/yr for chaparral catchments and of 17 kg/ha/yr for forested catchments in the southwestern Sierra Nevada and in the Transverse Ranges near Los Angeles, high concentrations of nitrate are found in stream water. In chaparral, forested, and high-elevation wilderness catchments in the Los Angeles air basin, up to 40% of the nitrate exported in stream water was directly deposited from the atmosphere and transported through the system without biological assimilation (Michalski et al., 2004), clearly indicating that the source of this excess nitrogen was air pollutant deposition in these watersheds.

High-elevation watersheds in the Sierra Nevada appear to export nitrate during early snowmelt, even with relatively low atmospheric nitrogen deposition similar to the high tendency to export nitrate discussed above for the Colorado Rockies. Although the role of nitrogen deposition on nitrate leaching in the high-elevation Sierra Nevada is not entirely clear, nitrogen deposition does not appear to have as severe an impact on lake and stream water chemistry, particularly in Sierra Nevada catchments, where soils tend to be thicker than those of the Colorado Front Range. Differing responses between the two regions may be due to higher

nitrogen deposition in the Colorado Rockies or to environmental differences. For example, the greater extent of soil freezing in the Colorado Rockies may reduce the nitrogen retention capacity of these watersheds (Sickman et al., 2002). However, watersheds in the Sierra Nevada are expected to be sensitive to increases in nitrogen deposition in the seasonal snowpack. This expectation is based on the large pulses of nitrate that are flushed to surface waters during snowmelt-driven spring runoff (Sickman et al., 2003); a short growing season; the limited soil and vegetation cover; and the limited biological nitrogen demand during the early spring runoff period (Fenn et al., 2003b).

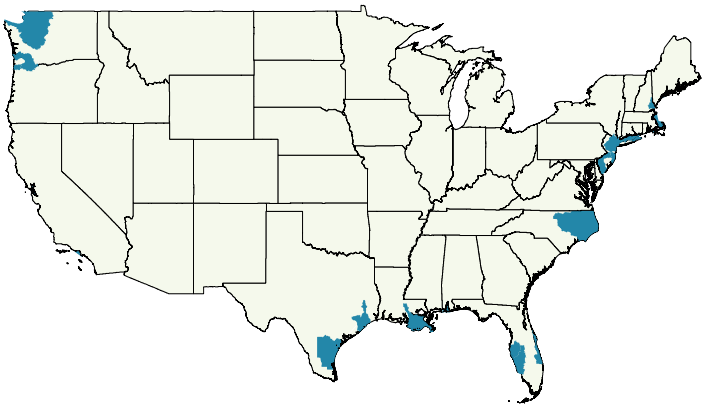


Figure 3-2. Map of major estuaries in the United States (prepared by U.S. EPA).

Effects of Atmospheric Nitrogen Deposition on Coastal Estuaries

Eutrophication and associated hypoxia is widespread in estuaries of the United States (Figure 3-2),including Long Island Sound, the Chesapeake Bay, and the Gulf of Mexico near the Mississippi River Delta (Diaz, 2001; Kemp et al., 2005). Nitrogen and phosphorus are the nutrients that commonly limit algal growth in coastal ecosystems. The role of nitrogen in estuarine eutrophication and hypoxia is often attributed to a combination of the delivery of excess nitrogen by rivers and the direct atmospheric deposition of nitrogen onto the water surface. These same mechanisms also deliver nitrogen to the open ocean, and atmospheric nitrogen deposition has been shown to affect the growth of marine plankton (Krishnamurthy et al., 2010). The potential sources of nitrogen to estuaries include runoff from fertilizer and animal waste applied to agricultural landscapes, human waste in urban/suburban landscapes, and atmospheric nitrogen deposition. The relative role of atmospheric nitrogen deposition to total nitrogen loads to coastal estuaries differs widely, from about 15% to 25% in the Mississippi Delta region of the Gulf of Mexico, to 20% to 50% in Chesapeake Bay and Tampa Bay, and as much as 60% to 80% in some coastal river estuaries of northern New England (McIsaac et al., 2001; Boyer et al., 2002; Poor et al., 2006; Whitall et al., 2007; Alexander et al., 2008; Howarth, 2008). The relative contributions of nitrate and ammonia as sources of atmospheric nitrogen to estuaries also vary widely across coastal regions of the United States. Nitrate is generally the dominant form of nitrogen in atmospheric deposition across the United States; however, ammonia can be the dominant atmospheric species of nitrogen in parts of estuarine watersheds, particularly those with abundant agricultural land (Lawrence et al., 2000; Whitall et al., 2003). The most recently available data indicate that the majority of U.S. estuaries show signs of eutrophication (Bricker et al., 2008), and the number of impacted estuaries in the United States and globally has increased greatly since the 1960s (Diaz and Rosenberg, 2008). Although most U.S. coastal waters have not shown changes in nutrient loads or trophic status since the 1990s (Alexander and Smith, 2006), about 20% to 25% of these coastal waters show improved trophic status in recent decades (Alexander and Smith, 2006; Bricker et al., 2008), most notably Tampa Bay. The role of decreased loads of atmospheric nitrogen deposition to observed decreases in estuarine eutrophication is likely only minor, but is not well quantified (Greening and Janicki, 2006).

The contribution of headwater streams to the total nitrogen loads in rivers can be substantial. For example, small streams account for more than two-thirds of the water volume and approximately two-thirds of the nitrogen delivered to large rivers in the Northeast (Alexander et al., 2007). Despite a higher removal rate (denitrification) of nitrate from headwater reaches than from large river channels during transport (Alexander et al., 2008), the colder temperatures at the higher elevations of the headwaters act to slow this rate of loss (Schaefer and Alber, 2007). Therefore, a substantial

amount of the nitrogen deposited on upland forested areas from the atmosphere may later be exported to larger rivers and estuaries. These studies emphasize the connection of upland ecosystems to coastal ecosystems and indicate that the effects of atmospheric nitrogen deposition extend beyond acidification in sensitive upland landscapes.

The issue of ocean acidification has emerged in recent years, with the observation that rising levels of carbon dioxide (CO_2) in the atmosphere are causing decreases in ocean pH and alkalinity, with numerous implications for the health of marine ecosystems (Caldeira and Wickett, 2003). Some recent studies have explored the role of atmospheric nitrogen and sulfur deposition on ocean acidification and concluded that acid deposition contributed no more than a few percent to ocean acidification trends in recent years (Bates and Peters, 2007). However, the contribution of acid deposition to marine pH declines is likely greater in coastal regions closer to continents (Doney et al., 2007). Further scrutiny of the role of acid deposition on the pH and alkalinity of coastal ecosystems may be needed in the future given the serious consequences of ocean acidification that are currently forecast.

Eutrophication and Hypoxia

Eutrophic waters are those with an overabundance of nutrients, such as nitrogen or phosphorus. As nutrient levels in the water increase, the growth of algae and plankton increases. When these organisms die, they are decomposed by bacteria that use the dissolved oxygen in the water. "Dead zones" characterized by hypoxia (i.e., low levels of dissolved oxygen in the water) or anoxia (i.e., no dissolved oxygen in the water) may result, harming or killing fish, crabs, shrimp, and other organisms. Benthic (bottom-dwelling) organisms such as oysters that are unable to move to more oxygenated waters are particularly susceptible.

Emerging Issues at the Interface of Science and Policy in Managing the Effects of Acid Deposition on Ecosystems

Critical Loads

Critical loads, which were introduced in Chapter 2, can be viewed as providing a synthesis of scientific knowledge about thresholds of harm to or alteration of ecosystems that can inform the development, implementation, and assessment of air quality policy and the management of sensitive ecosystems. Critical loads have been widely used for large-scale applications and policy purposes in Europe and Canada since the 1980s (Jeffries and Ouimet, 2005; Spranger et al., 2008), but historically have not been widely used for these purposes in the United States (Porter and Johnson, 2007). For example, critical load maps for soils and waters are produced by several European countries that are parties to the *Convention on Long-Range Transboundary Air Pollution*. Although a signatory to this Convention, the United States did not commit to the provisions regarding critical loads contained in the Convention (see recent discussion in Spranger et al., 2008). Academic critical load studies in the United States also date to the 1980s (Henriksen and Brakke, 1988), and many studies continue to be published (Baron, 2006; Fenn et al., 2008; Sullivan et al., 2008); however, most past work has been of limited geographic scope. Recently published studies provide examples of larger-scale assessments of critical loads in the United States, including nationwide critical load maps for forest soils (McNulty et al., 2007) and a review of published studies throughout the United States for empirical critical loads of nitrogen (Pardo et al., 2011).

Critical loads can be calculated for any species of concern or for a sensitive biological community within an aquatic or terrestrial ecosystem if a threshold of atmospheric deposition can be identified below which negative biological effects on the species or alteration of the community are not expected to occur. Because various species in an ecosystem may have differing sensitivities to air pollutant loads, a large number of species-specific or community-specific critical loads are likely to exist for a given location (Sullivan et al., 2008).

Critical loads are often linked to thresholds at which the organism experiences a certain biological effect, which may range from stunted growth to reduced reproductive levels to increased mortality. Critical loads are usually calculated in reference to a chemical quantity believed to provide an index of acidification or nutrient over-enrichment. Potential indices include ANC levels or nitrate concentrations in a waterbody, or the base cation saturation level of a soil. Multiple critical loads and ecological effects end points provide policy makers or land managers with multiple options to determine the level of protection that is warranted based on the species or communities of greatest concern at a given location. This is part of the process of establishing a target load, which may differ from the strictly science-based critical load. The target load reflects inclusion of economic, social, cultural, or other considerations to establish the protection level of the ecosystem in question, as well as the load that is deemed "achievable" by managers or regulators (Porter et al., 2005; Porter and Johnson, 2007).

In addition to a variety of critical loads applicable for each ecosystem based on the species and biological effects being considered, there are different types of critical loads and different methods for establishing critical loads. Critical loads can be determined by an empirical approach based on field- or laboratory-based studies that define a chemical threshold of sensitivity for a species or community. A recent example is the establishment of a critical load of 1.5 kg/ha/yr of atmospheric nitrogen deposition to protect against eutrophication (nutrient over-enrichment) in lakes in the Rocky Mountain National Park. This load was established based on evidence from lake sediment cores that showed a shift in phytoplankton species as nitrogen deposition values rose above 1.5 kg/ha/yr in the 1960s (Porter and Johnson, 2007). Critical loads may also be determined by a mathematical expression, such as the Steady-State Mass Balance model for soils. This model compares the concentrations of neutralizing base cations (e.g., calcium, magnesium) to those of potentially toxic acidifying cations (e.g., aluminum) in soils (Sverdrup and deVries, 1994). Both the empirical and mathematical expression approaches develop steady-state critical loads based on an assessment of current levels of atmospheric deposition relative to some threshold of harm or change. Critical loads can also be calculated using a dynamic approach that involves more sophisticated modeling of time-varying patterns of emissions, deposition, soil chemistry, and lake or stream chemistry. This approach often uses some of the acidification models (e.g., MAGIC or photosynthetic evapotranspiration-biogeochemical [PnET-BGC]) described in the text box and can provide information on the time expected to reach a threshold based on a given emissions policy as it is implemented over time (Sullivan et al., 2008). The dynamic critical load approach is especially valuable for evaluating the success of air quality policies and programs as they are implemented over multiple years.

Despite the lack of a long history of applying critical loads to inform air pollutant policies in the United States, interest in the use of critical loads in the United States has increased in recent years with the advent of a Critical Load Science Committee within NADP (NADP, 2009), several recent workshops and meetings on this topic, and several publications exploring greater use of critical loads as a policy-relevant environmental assessment tool (Porter et al., 2005; Fisher et al., 2007; Burns et al., 2008b; Glavich and Geiser, 2008). Recently published studies have generally found that for many regions well known for their sensitivity to either acidification or excess nitrogen deposition (e.g., the Adirondack Mountains of New York, the central Appalachian region of Virginia, the Front Range of Colorado), the current levels of atmospheric deposition generally exceed the critical loads for many species of concern (Baron, 2006; McNulty et al., 2007; Sullivan et al., 2008). Case studies exploring the application of the critical load approach for the Adirondack Mountains and the central Appalachian region are also included in Chapter 2 and Chapter 4 of this report. Finally, consideration of two key factors is warranted when using scientific information on critical loads to inform public policy implementation: (1) critical loads can have high uncertainty that stems from model assumptions and data, as well as uncertainty in the threshold itself (Skeffington, 2006; Li and McNulty, 2007),

and (2) other variables such as forest harvesting and climate change will affect critical loads in addition to atmospheric deposition (Watmough et al., 2003). When critical load calculations are being used to inform air pollutant policies, scientific uncertainty must be properly considered, along with the role of other mitigating variables.

Atmospheric Deposition and Ecosystem Services

In addition to the health benefits of the ARP discussed in Chapter 1 of this report, benefits to ecosystem services also might be expected, and these benefits can be evaluated by considering the varied material and intangible goods and services by which ecosystems support human well-being (PCAST, 2011). Ecosystem services can be defined as the sum total of benefits that humans receive from the resources and processes of natural ecosystems (Corvalan et al., 2005). These benefits range from extractive services or provisioning (e.g., wood products, minerals, water) to regulating services (e.g., climate regulation, nutrient cycling) to cultural services (e.g., outdoor recreation). The ability of ecosystems to provide these services over the long term is impacted by disturbances (Beier et al., 2008), including atmospheric deposition. The response of ecosystems to disturbances, such as acid deposition, depends on the resiliency of the ecosystem in question. Over the past two decades, researchers have begun to establish the value of these various ecosystem services (Costanza et al., 1997) and to develop approaches for valuating both the direct use-related benefits, as well as the non-use benefits of ecosystems; most studies indicate these non-use benefits exceed those of the more traditional market-based benefits derived from resource extraction (Walsh et al., 1990).

Models Used to Investigate the Ecosystem Effects of Acid Deposition

Models play a pivotal role in understanding the ecosystem effects of acid deposition, as well as helping to inform air quality policy. Below are the principal models that have been used to study ecosystem effects.

Initial Models—These models were simple and used an analysis of surface water chemistry based on charge balance or titration by strong acids (Henriksen, 1979).

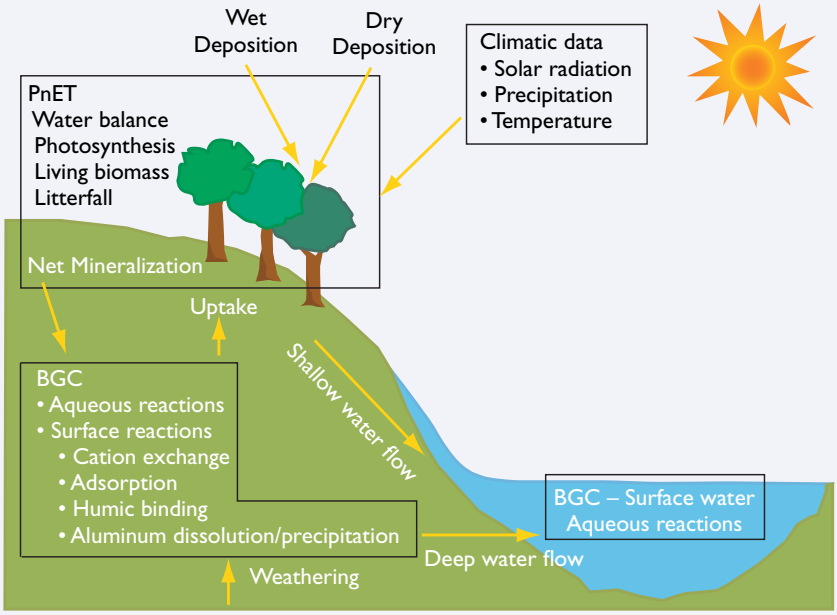
Dynamic Models—Later, more formal computer models were developed that simulate biogeochemical processes and hydrology occurring in catchments (see the figure on the following page). The dynamic models listed all share an ability to make predictions about future concentrations and loads of chemical constituents (e.g., pH, ANC) in lakes and streams given the chemistry of atmospheric deposition.

- Integrated Lake-Watershed Acidification Study (ILWAS) model (Gherini et al., 1985)
- Birkenes model (Christophersen et al., 1982)
- Model of Acidification of Groundwater in Catchments (MAGIC) (Cosby et al., 1985c)
- Photosynthetic evapotranspiration-biogeochemical (PnET-BGC) (Gbondo-Tugbawa et al., 2001).

All of the dynamic models can be combined with atmospheric models, such as the Community Multiscale Air Quality model (CMAQ), that estimate atmospheric deposition to landscapes from present emission levels or future emission scenarios (Schwede et al., 2009).

(continued)

Models Used to Investigate the Ecosystem Effects of Acid Deposition (continued)



Conceptual diagram of the biogeochemical processes and chemical stores of the PnET-BGC model (figure courtesy of Charles Driscoll, Syracuse University).

Other dynamic models, such as the following, have been applied more commonly in ecosystems of the western United States:

- DayCent/PHREEQC (DayCent-Chem) (Hartman et al., 2007)
- Alpine Hydrochemical Model (AHM) (Meixner et al., 2004).

Steady State Models—These are another class of models that focus only on a steady-state approximation of either water or soil chemistry (Henriksen and Posch, 2001). Steady-state models, such as those listed below, are computationally simpler and demand less input data to run:

- Steady-State Water Chemistry (SSWC)
- Simple Mass Balance Equation (SMBE).

Several recent surface water chemistry modeling studies have explored likely future ecosystem responses to existing air pollutant policies (e.g., Title IV), along with some future emissions scenarios (Chen and Driscoll, 2004; Sullivan et al., 2004; Chen and Driscoll, 2005a; Chen and Driscoll, 2005b; Lawler et al., 2005; Sullivan et al., 2008). These modeling studies not only generally confirm the success of Title IV at improving the acid-base status of surface waters, but also highlight regional differences and point out that additional emission reductions will be needed to realize the goal of full ecosystem recovery to conditions resembling pre-acidification conditions. Chapter 4 of this report includes a modeling analysis in which MAGIC was used to assess aquatic ecosystem recovery in several regions of the eastern United States as a result of implementing current policies and programs in comparison to results achieved as a result of implementing three alternative future emission-reduction scenarios.

A recent review discusses and summarizes studies on estimating the monetary value of the effects of sulfur and nitrogen emissions and deposition on ecosystems (Krupnik and Siikimaki, 2008). The studies cited can be divided into those that estimate the monetary value of terrestrial, aquatic, and transitional (mainly wetland) ecosystems. Several studies have generally estimated the value of adverse ecosystem effects, but many fewer studies have attempted to relate specific adverse effects that can be directly attributed to sulfur and nitrogen deposition. Some studies have estimated the value of benefits from minimizing or eliminating ecosystem disturbances such as insect defoliation of forests (Kramer et al., 2003) and estuarine eutrophication (Morgan and Owens, 2001), in which acid deposition is clearly one contributing factor among many. However, the monetary role of acid deposition cannot be estimated in most of these studies.

The greatest challenge in developing specific data on the economic benefits of changes in acid deposition due to emission reductions lies with the availability of comprehensive scientific evidence that defines the extent and magnitude of the adverse effects that can be directly attributed to acid deposition from among multiple ecosystem stressors (Krupnik and Siikimaki, 2008). In one notable study, Banzhaf et al. (2006) estimated ecological benefits of a policy that would substantially decrease adverse effects of acid deposition in the Adirondack Park of New York. These benefits are in the range of about $300 million to $1.1 billion annually for the Adirondack Park only and do not consider ecological benefits in other regions of the United States from these deposition reductions. As described in Evans et al. (2008), researchers are applying economic methods to show the total value people place on ecological improvements to important resources, such as the Adirondack Park, expected from further reductions in acid deposition. However, as discussed in Chapter 1 of this report, no studies have comprehensively evaluated the monetary benefits to ecosystem services of the ARP across all ecosystem types that would be sufficient to make comparisons to overall health benefits and costs of the ARP. Significant future analytical work and basic ecological and economic research is needed to build a sufficient base of knowledge and data to support an adequate assessment of ecological benefits.

Forest harvesting is an example of an ecosystem service that provides monetary benefit to land owners (photo courtesy of Douglas Burns, USGS).

What are the Implications of Ongoing and Future Changes in Climate and the Carbon Cycle for the Effects of Acid Deposition on Ecosystems?

The Earth has warmed by an average of 0.74° C between 1906 and 2005, and there is a high likelihood that recent warming over the past several decades has been driven in large part by increases in atmospheric concentrations of CO_2 and other greenhouse gases (IPCC, 2007). Under a range of likely future scenarios of greenhouse gas emissions and a range of climate model predictions, the Earth is likely to warm by another 1.4°–5.8° C during the twenty-first century (IPCC, 2007).

Ongoing and future predicted changes in the climate and the carbon cycle have numerous implications for the response of ecosystems to atmospheric deposition of sulfur and nitrogen. In addition to warming air temperatures, a wide array of other environmental variables are predicted to change with the climate, including water availability and the rates of many nutrient cycling processes. These additional environmental changes and their effects are likely to vary by region (Boisvenue and Running, 2006). In short, ongoing and future predicted climate change is likely to greatly affect

the interactions of sulfur and nitrogen deposition with ecosystems in a complex manner that does not easily lend itself to simple summaries and has high uncertainty (McLaughlin and Percy, 1999). For example, although the effects of an average increase in air temperature are likely to induce widespread changes in ecosystems, an increase in the magnitude and frequency of a wide variety of climatic events (e.g., floods, droughts, fires) also is expected. Some have suggested that these events are likely to affect ecosystems to a greater extent than long-term average changes in temperature (Dale et al., 2001; Jentsch et al., 2007). Despite these complexities, a brief assessment of likely interactions between ecosystems, acid deposition, and climate change is warranted in this current assessment given the potential magnitude and multi-faceted impacts of the projected changes. This section focuses on ecosystem effects and does not discuss the interactions between climate change and greenhouse gases or how these interactions may affect the transport, atmospheric interactions, and atmospheric deposition of sulfur and nitrogen. These atmospheric changes have been discussed elsewhere (Tagaris et al., 2008) and are beyond the scope of this assessment.

> Ongoing and future predicted climate change is likely to greatly affect the interactions of sulfur and nitrogen deposition with ecosystems in a complex manner that does not easily lend itself to simple summaries and has high uncertainty.

Implications of Climatically Driven Changes in the Species Composition of Ecosystems

Perhaps the greatest impact of climate change is likely to be wholesale changes predicted for the biological communities found in many ecosystems sensitive to acid deposition effects, particularly those at high elevations (Ibanez et al., 2007). For example, a migration northward and to higher elevations is predicted for many tree species under most current climate change forecasts as they are replaced by species better adapted to a warmer climate (McKenney et al., 2007; Lenoir et al., 2008). Evidence indicates that migration of lower elevation forests to higher elevations is already occurring in Vermont, suggesting this shift is ongoing in mountainous areas that have warmed in recent decades (Beckage et al., 2008). Similar cold-to-warm water species shifts are also predicted for aquatic ecosystems (Mohseni et al., 2003; Durance and Ormerod, 2007). The rate at which these ecosystem changes will occur is not well known and is likely to reflect interactions with other contemporaneous changes, such as those in water availability, light, insect defoliation, land use, and air pollutant deposition. Overall, the evidence suggests that many species may reach an out-of-equilibrium state with respect to a future warmer climate (Higgins and Harte, 2006; Mohan et al., 2009), and this state may result in increased sensitivity of many terrestrial and aquatic ecosystems to the effects of other disturbances, such as sulfur and nitrogen deposition. A European study (Alcamo et al., 2002) concluded that climate change is likely to have little effect on the sensitivity of forested ecosystems to air pollutant deposition. However, the weight of evidence from other studies indicates that, despite some uncertainty in the effects of climate change on ecosystems (Aber et al., 2001), ongoing and likely future climate change will provide an added stressor to many ecosystems in the United States. Furthermore, several studies indicate that the combination of atmospheric deposition of sulfur and nitrogen with high ozone concentrations will act as additional ecosystem stressors in many regions, such as the eastern United States (McLaughlin and Percy, 1999; Mohan et al., 2009). The increases in extreme events, such as droughts and fires, which are predicted with future climate change, will likely act as additional stressors to many of these ecosystems, with greater effects expected in the western United States (Dale et al., 2001; Knapp et al., 2008).

Climate Warming Will Affect Many Temperature-Sensitive Biogeochemical Processes in Ecosystems

Many biogeochemical processes affect the cycling of atmospherically deposited sulfur and nitrogen in ecosystems and play a pivotal role in the

acidification of ecosystems, including mineral weathering, nitrification, and sulfur and nitrogen mineralization. The rates of nearly all of these processes are likely to be affected by changes in climate. For example, the rates of mineral weathering and nitrification increase with increasing temperature if all other factors are held constant (White et al., 1999; Dalias et al., 2002). Studies have shown widely varying biogeochemical responses to past and predicted climate warming in the United States, and these varying responses reflect interactions with precipitation, snow and ice, and seasonal variations. For example, Campbell et al. (2009) predict higher nitrate concentrations in soil water and streams at Hubbard Brook, NH, due to warming-induced increases in nitrogen mineralization and nitrification, yet they also predict decreased rates of mineral weathering due to decreases in soil moisture. At the Rocky Mountain National Park in the western United States, stream nitrate concentrations have been increasing in recent years due to the melting of glaciers and snow fields by increased warming in summer through fall and the leaching of nitrate that occurs when the underlying soils are exposed to microbial processes (Baron et al., 2009). These results highlight the sensitivity of long-term stream water quality to changes in snowfall, snowmelt, and the mass of glaciers in high-elevation watersheds of the western United States. Data from the European Alps show a recent trend towards increasing solute concentrations in high-elevation lakes, consistent with accelerated mineral weathering rates driven by warmer air temperatures and less-extensive snow cover that lasts for a shorter duration (Mosello et al., 2002; Rogora et al., 2003). Increased mineral weathering rates would likely result in a greater ability of soils to neutralize acid deposition and might accelerate recovery in sensitive ecosystems when accompanied by decreasing atmospheric deposition rates of sulfur and nitrogen. However, a recent study in the Colorado Rockies showed that regions underlain by sulfide minerals such as pyrite may experience greater acidification with warmer air temperatures due to increased mineral weathering rates and the subsequent formation of sulfuric acid (Mast et al., 2010).

Climate change studies in Loch Vale have documented increasing air temperatures, earlier snowmelt, and earlier runoff. These changes may have important impacts on water availability, water quality, and ecosystem function (photo courtesy of USGS).

For a range of likely twenty-first century climate change scenarios in Europe, Posch (2002) concluded that ecosystems will generally become less sensitive to acid deposition, primarily due to increasing rates of mineral weathering in soils driven by warmer temperatures. The amount of land in Europe subjected to sulfur and nitrogen deposition loads in excess of the critical load for acidification is likely to decrease with climate change, except in areas predicted to become more arid. Decreases in the exceedance of critical loads for nitrogen-driven nutrient over-enrichment are also predicted with future climate change in Europe (Posch, 2002), but these decreases were less than those modeled for decreases in critical load exceedances for acidification. In contrast, existing studies in the United States predict higher nitrate concentrations (which should lead to greater critical load exceedances for nutrient over-enrichment) with future climate change (Campbell et al., 2009; Wu and Driscoll, 2010). These opposing forecasts for expected future nitrate chemistry in surface waters indicate the current high uncertainty about the role of climate change and point to a need for additional research and improved models to explore this important issue (Spranger et al., 2008).

Implications of Changes in Carbon Dioxide Concentrations in the Atmosphere

One of the fundamental factors that affects the rates of photosynthesis and terrestrial ecosystem growth is the availability of CO_2. Global atmospheric CO_2 concentrations have increased by nearly 40% since the mid-eighteenth century and are expected to double during the twenty-first century (IPCC, 2007). The ecosystem "CO_2 fertilization effect" has been much discussed in the literature (Tans et al., 1990; Nowak et al., 2004). Indeed, measurements and satellite observations of forested ecosystems, as well as CO_2 manipulation experiments, have generally reported increasing net ecosystem growth (primarily in forests) as CO_2 concentrations have increased in temperate regions of the world, although there have been fewer monitoring-based studies in the United States than in Europe (Norby et al., 2005; Boisvenue and Running, 2006). The ecological responses have not been consistent, however, in all studies and across all regions. A wide variety of factors (e.g., disturbance history, water availability, solar radiation), in addition to experimentally altered CO_2 concentrations, can contribute to these diverse patterns in net terrestrial ecosystem growth trends (Nemani et al., 2003; Boisvenue and Running, 2006; Potter et al., 2006).

Research at the Forest-Atmosphere Carbon Transfer and Storage (FACTS-I) facility in the Duke Forest has shown that nitrogen deposition can affect the uptake of atmospheric carbon dioxide as forests grow (photo courtesy of U.S. Department of Energy's Brookhaven National Laboratory).

There are several challenges in trying to quantify the role of CO_2 in forest growth. Many factors generally co-vary across sites, and experiments necessary to understand the interacting effects of these multiple factors have not been undertaken (Norby and Luo, 2004; Rustad, 2008). Also, the nearly impossible task of designing experiments that adequately consider variations in the numerous factors that influence ecosystem growth has been noted (Aber et al., 2001; Boisvenue and Running, 2006). When variations in atmospheric sulfur and, especially, nitrogen deposition are added to this mix of factors, the difficulty in attributing cause and effect to observed changes in ecosystems increases further (Bytnerowicz et al., 2007), and some argue, can only be interpreted through the use of models (Aber et al., 2001). Nonetheless, results from a CO_2 enrichment experiment that simulated likely twenty-first century concentrations showed increased soil acidification and increased rates of mineral weathering, suggesting that important interactions with terrestrial ecosystem effects of atmospheric deposition may occur in the future (Andrews and Schlesinger, 2001).

Changes in the Water Cycle are Key to Understanding Ecosystem Effects

Under various likely twenty-first century climate-change scenarios, most climate models predict an overall increase in global precipitation of about 1–3% per degree of temperature increase (Held and Soden, 2006). Recent observations and satellite measurements generally support an even greater increase in precipitation of about 5–10% per degree of warming (Wentz et al., 2007; Zhang et al., 2007). Such a change is expected to increase the wet deposition of sulfur and nitrogen in the eastern United States if all other factors remain the same (Civerolo et al., 2008). However, the prediction of climate "wetting" is less certain than the prediction of climate warming, and changes in precipitation patterns are predicted to vary widely over the United States, with forecasts for a wetter climate in the eastern United States and for a drier climate in large parts of the western United States (Milly et al., 2005). These drier climate patterns are already evident in the western United States, which is exhibiting smaller winter snowpacks and earlier snowmelt in mountainous areas (Mote et al., 2005).

Earlier snowmelt has also been noted in upland watersheds in the Northeast (Burns et al., 2007). Model predictions with likely future climate scenarios indicate continued diminishing snowfall, less snowpack, and less of a role for snowmelt in the hydrologic cycle of temperate regions of the Northern Hemisphere (IPCC, 2007).

These patterns may affect the cycling of sulfur and nitrogen pollutants through ecosystems and watersheds by impacting episodic acidification, soil freezing, and the flows and temperatures in surface waters. When snow melts, surface-water chemistry is often at its most acidic and has its highest nitrate concentrations; therefore, a diminished snowmelt may lessen the episodic acidification of aquatic ecosystems (Moore et al., 1997).

However, these predicted trends of diminishing snowmelt are uncertain, and an increase in mid-winter rain has been noted in the eastern United States (Hodgkins et al., 2003). This increase in winter rain will also likely trigger strong episodic acidification, even when the rain falls on a diminished snowpack. Additionally, increases in large rainfall events have been observed and are predicted to increase with climate change in the twenty-first century (Murdoch et al., 2000). Increased episodic acidification from these large rainfall events may offset the predicted decreasing trend in snowmelt. In addition, diminished snow cover may change soil freezing patterns in winter, which are dependent on the magnitude of warming. Frequent freeze-thaw behavior in soils can increase nitrate leaching and raise nitrate concentrations in surface waters (Fitzhugh et al., 2003).

Finally, earlier and diminished snowmelt may result in lower stream flows and warmer water temperatures in the later spring and summer. Warmer temperatures can result in increased growth rates. Increased aquatic growth can lower nitrate concentrations through biological uptake (Sommaruga-Wogarth et al., 1997); however, increased growth also can have negative impacts (e.g., hypoxia; see the section *Effects of Atmospheric Nitrogen Deposition on Coastal Estuaries* in this chapter for further discussion). Warmer temperatures and lower flows in streams may also have deleterious effects on cold-water fish species (Mohseni et al., 2003).

A nearly universal prediction from models of future climate is that droughts will increase in severity and duration, even in areas such as the eastern United States where overall increases in precipitation are predicted (IPCC, 2007). This forecast has strong implications for the impacts of acidification on aquatic ecosystems. When soils are dry, oxidized nitrogen and sulfur species accumulate and are released and flushed into local waterbodies upon re-wetting. Several studies have shown increased concentrations of nitrate and sulfate and decreased ANC values in surface waters when wet conditions return following drought periods. These impacts are especially apparent in wetland-influenced watersheds (Tipping et al., 2003; Watmough et al., 2004; Schiff et al., 2005).

This pattern of nitrogen and sulfur storage and release has further implications for the episodic acidification of waterbodies. Soils affected by acid deposition often contain levels of stored nitrogen and sulfur that are equivalent to decades of atmospheric deposition. Therefore, the aquatic ecosystem effects of severe, post-drought episodic acidification in lakes and streams and other surface waters may persist for decades after deposition levels have declined below critical loads (Tipping et al., 2003; Eimers et al., 2007).

Rapid shifts from periods of dry to wet conditions are also expected to increase in the future. Increases in acidification driven by such climatic shifts have been shown to directly affect aquatic biological communities such as diatom algae (Faulkenham et al., 2003) and to alter physico-chemical variables that affect aquatic life, such as the penetration of ultraviolet radiation (Yan et al., 1996).

Nitrogen Availability has Important Implications for the Response of Ecosystems to Climate Change

One of the most uncertain and important ecosystem-related issues regarding the interaction of the atmospheric deposition of pollutants with changes in climate and CO_2 levels concerns the role of nitrogen availability and its impact on growth and carbon sequestration. Growth in the majority of U.S. and global ecosystems is limited by nitrogen availability, suggesting that atmospheric nitrogen

deposition may be acting to increase growth and carbon sequestration in many ecosystems across the United States. However, in terrestrial ecosystems, excess atmospheric deposition of nitrogen also contributes to ecosystem acidification, calcium depletion in soils, loss of diversity, and excess runoff of nitrate, which can, in turn, lead to over-enrichment in freshwater and estuarine ecosystems. Therefore, when considering atmospheric deposition of nitrogen, the potential "benefit" of increased growth and carbon sequestration must be weighed against the potential "costs" of resulting nutrient over-enrichment, decreased ecosystem services (e.g., biodiversity), and the promotion of invasive species (Fenn et al., 2003b). These issues are brought into sharp focus by studies that seek to determine the role of nitrogen in the present and future growth of terrestrial ecosystems (particularly forests) and the sequestration of carbon as climate and atmospheric CO_2 concentrations continue to change.

One general conclusion is that net ecosystem growth in temperate and boreal forests has increased coincident with climate change in the United States, across Europe, and in parts of Asia over the past few decades (Goodale et al., 2002; Nemani et al., 2003; Boisvenue and Running, 2006; Magnani et al., 2007). However, many local and regional exceptions to this generalization can be found (Korner, 2003). Another general conclusion is that atmospheric nitrogen deposition acts to increase net ecosystem growth and carbon sequestration in mid-latitude forests (Reich et al., 2006; Magnani et al., 2007), although the magnitude of this nitrogen-driven carbon sink is likely much less than originally estimated by some studies (Sutton et al., 2008). Recent estimates of this sink are approximately 3% to 21% of annual CO_2 emitted globally from fossil fuel combustion (Churkina et al., 2009; Thomas et al., 2009; Zaehle et al., 2010). The interactions of carbon and nitrogen are critical in controlling the magnitude of the terrestrial sequestration of a large amount of the anthropogenic (i.e., man-made) CO_2 that would otherwise reside in the atmosphere and further alter the global climate. As CO_2 in the atmosphere increases, terrestrial carbon sinks (especially forests) have been shown to increase; however, a point may be reached when nitrogen and other nutrients become limiting (Johnson, 2006; van Groenigen et al., 2006). The role of atmospheric nitrogen deposition in the global carbon cycle highlights the complex linkages among biogeochemical cycles and the important link between air pollutant deposition and global climate change.

Dissolved Organic Carbon—Acid Deposition Interactions: A Case Study of Climate Change

Widespread increases in the concentrations and loads of dissolved organic carbon (DOC) in surface waters have been reported in the United States and Europe (Driscoll et al., 2003; Evans et al., 2006; Monteith et al., 2007). DOC is formed as organic matter decomposes and dissolves in water. DOC levels in surface waters are important because of the demonstrated link between atmospheric nitrogen and sulfur deposition and DOC loads in these waters (Clark et al., 2010). DOC plays a pivotal role in human health concerns related to water supply and distribution through the formation of disinfection by-products (Escobar et al., 2001; Chow et al., 2003). Additionally, DOC plays an important role in a wide array of aquatic ecosystem effects and interactions, including light penetration, water temperature, thermal stratification, plankton growth, pH and acidification, and the transport of trace metals (Snucins and Gunn, 2000).

A variety of causes have been offered to explain why DOC concentrations are changing in remote fresh waterbodies that are not greatly influenced by human land use. These causes include the following:

- Decreasing atmospheric sulfur deposition (Evans et al., 2006; Monteith et al., 2007);
- Climate warming (Worall and Burt, 2007; Clair et al., 2008);
- Changes in precipitation amount (Hudson et al., 2003; Worrall et al., 2003);
- Changes in incident solar radiation (Hudson et al., 2003);
- Decreases in sea salt deposition (Monteith et al., 2007); and
- Chronic inputs of atmospheric nitrogen deposition (Findlay, 2005).

CASTNET monitoring site in Rocky Mountain National Park, Larimer County, CO (photo courtesy of U.S. EPA).

Reduced levels of sulfur deposition appear to be playing an important role in increased DOC concentrations in many surface waters in North America (Monteith et al., 2007), and the strength of this association is likely to be greatest where atmospheric deposition loads are greatest (Clark et al., 2010). Decreases in atmospheric sulfur deposition over the past 10–20 years have resulted in increasing pH and decreasing ionic strength in many waters, both of which should increase the solubility of organic matter in soils and sediment and, therefore, the DOC levels in surface waters. These observed increases in DOC concentrations may simply reflect ecosystem recovery and a return to the "natural" levels of DOC that were present before widespread acid deposition acidified aquatic ecosystems. Also, some observed DOC increases that appear to be driven by drought occurrence may actually result from the drought conditions favoring oxidation of previously reduced sulfur compounds that often originate from atmospheric deposition (Clark et al., 2006).

Climate variation also probably partly explains increasing surface-water DOC trends in some regions (Hudson et al., 2003; Worrall et al., 2003). Fundamentally, warmer soil and sediment temperatures should increase the decomposition rate of organic matter and DOC levels in waters. However, variations in moisture, nutrient availability, the availability of dissolved oxygen, and other variables would be expected to mask a simple DOC to temperature relationship (Giardina and Ryan, 2000). For example, the Arctic regions of North America have experienced some of the greatest warming trends on Earth. Climate warming is expected to significantly increase DOC concentrations and fluxes in Arctic surface waters (Clair and Ehrmann, 1998; Frey and Smith, 2005). However, data from the Yukon basin supports decreased DOC export, which may result from the conversion of soil carbon to CO_2 in the active soil permafrost layer (Striegl et al., 2005). Also, research conducted in Canadian lakes has highlighted the important role of other climate-related factors (e.g., drought-wetting cycles, variations in solar radiation) on surface water DOC concentrations (Dillon and Molot, 1997; Schindler, 1998; Hudson et al., 2003).

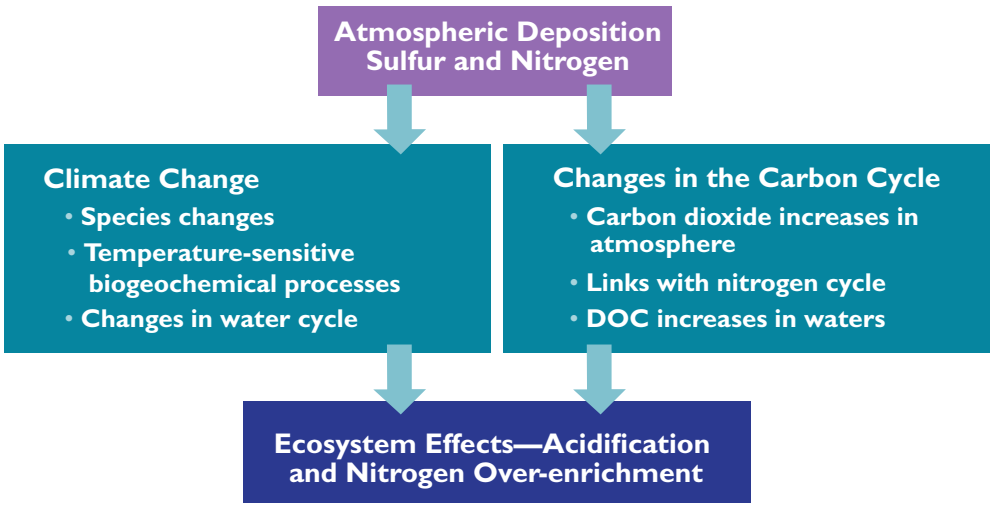
Regardless of the relative roles of climate change and changes in atmospheric deposition of sulfur and nitrogen in driving these recent patterns of increased DOC concentrations and loads in surface waters, this issue highlights the interplay of acid deposition and climate change. Increases in DOC concentrations generally result in decreases in ANC; therefore, these increased DOC concentrations are likely slowing the rate of recovery of aquatic ecosystems, as inferred from measurements of ANC (Monteith et al., 2007). Because of the widespread importance of DOC for aquatic ecosystems, future monitoring and modeling efforts should continue to scrutinize and consider the interplay of climate and atmospheric deposition. Historically, the recognition and study of DOC trends was not much discussed or considered until data from long-term monitoring networks, such as the LTM and TIME programs in the United States and other programs across the world, showed the magnitude and widespread geographic nature of these trends.

Terrestrial ecosystems store large amounts of carbon and the potential for these ecosystems to sequester or release additional carbon will likely contribute to future climate change. Research indicates that atmospheric nitrogen deposition affects the ability of terrestrial ecosystems to sequester atmospheric carbon dioxide.

Summary of Climate Change—Atmospheric Deposition Linkages

The results summarized here provide a strong scientific basis for joint consideration of climate change and air quality policies affecting atmospheric nitrogen and sulfur deposition. Topics considered under the umbrella of global climate change include past, ongoing, and future expected changes in a wide range of factors that affect ecosystems, such as warmer air temperatures, water availability, and the frequency of large magnitude events (e.g., droughts, floods, fires). The combination of patterns in the atmospheric deposition of sulfur and nitrogen with these global climate change factors has implications for the growth of ecosystems, expected shifts of species, and the acid/base chemistry of surface waters. Untangling the interactions among multiple biogeochemical cycles and the resulting ecosystem responses to changes in these cycles is complex and uncertain. For example, global climate change generally provides an additional set of interacting stressors in ecosystems that are negatively impacted by acid deposition (see Figure 3-3). However, climate change also may increase rates of chemical weathering in some locations and, therefore, ecosystems may be able to sustain greater amounts of sulfur or nitrogen deposition before experiencing negative effects. Conversely, atmospheric nitrogen deposition may currently be limiting global warming through enhanced ecosystem carbon sequestration in U.S. regions that are only lightly to moderately impacted by air pollutant deposition.

Currently, the degree of uncertainty regarding many of the predicted effects of global climate change on ecosystems is high and often not well quantified. Environmental monitoring programs are helpful in understanding climate-change effects on ecosystems, but often are not adequate in geographic scope or in the number of parameters measured to fully understand these effects. For example, the leading programs that measure atmospheric deposition (NADP and CASTNET) and surface-water chemistry (LTM and TIME) generally do not measure many climatically relevant variables, such as snow-water equivalents and air and water temperature. Additionally, several of the papers reviewed here highlighted the critical need for more multi-factorial and interdisciplinary experiments to test the effects of varying air/soil/water temperature, CO_2 concentrations, and



Note: Although the rate of atmospheric deposition of sulfur and nitrogen are the principal drivers of the ecosystem effects discussed in this chapter, ongoing and future changes in climate and the carbon cycle are expected to interact with these ecosystem effects in ways that are currently difficult to predict. Ongoing research efforts are helping to unravel the role of climate change on ecosystems so that more accurate model forecasts can better constrain the role of climate and carbon.

Figure 3-3. Climate change, acid deposition, and other stressors (prepared by USGS).

pollutant deposition levels on ecosystems. There is also a need for consideration of climate-change effects in acidification and nitrogen effects models, such as MAGIC and PnET-BGC, as they are used to provide predictions of the expected impacts of future air pollutant deposition scenarios. Work of this type is ongoing, but not yet widely available in the published literature. Evaluations of the ability of these models to incorporate changing climatic effects would also be helpful in providing feedback to discussions about future policies. Finally, past and future disturbances that are not necessarily climate-change driven (e.g., land disturbance, land use changes, invasive species, fire) can have ecosystem effects that are as great or greater than those derived from climate change and are also worthy of consideration in future scenario modeling.

Multi-Pollutant Interactions in Ecosystems

Throughout this chapter, the interacting effects of nitrogen and sulfur deposition have been discussed in reference to the acidification and nutrient over-enrichment of ecosystems. In the previous section, the interacting effects of acid deposition and changes in the carbon cycle and climate change were discussed. There are also important interactions between acid deposition and the ecosystem effects of ozone and mercury, and these interactions will be discussed in this section. In recent years, interest has been increasing in development of air quality policies that target multiple pollutants (NRC, 2004; Brook et al., 2009; NARSTO, 2011). This section focuses on the ecosystem aspects of two pollutants (i.e., ozone and mercury) for which clear links have been demonstrated.

Ozone Interactions with Atmospheric Sulfur and Nitrogen Deposition

Ozone is taken up by plants and, in high concentrations, can directly damage plant cell membranes, reduce the rate of photosynthesis and growth, and cause species shifts in forests (Karnosky et al., 2007). In short, ozone is yet another pollutant stressor that can interact with sulfur and nitrogen deposition and changes in climate and CO_2 concentrations to affect ecosystem health (McLaughlin and Percy, 1999). Ozone concentrations and trends are highly variable across the globe and the United States, but leveling or slight decreases in concentrations in North America have been noted in recent years (Vingarzan, 2004; Oltmans et al., 2006). Trends in ozone concentrations are affected by trends in (1) the sources of ozone, such as fossil fuel use, (2) the principle atmospheric precursors of ozone (i.e., NO_x, volatile organic compounds, and carbon monoxide), and (3) climatic factors, particularly summer stagnation episodes that favor the highest concentrations. Results from models that simulate likely twenty-first century climate, combined with chemical transport model outputs, indicate a likelihood of increased future ozone concentrations in the United States (Jacob and Winner, 2009). Additionally, tropospheric ozone (ozone in the lowest portion of the Earth's atmosphere) provides the third-strongest warming influence of all greenhouse gases (Mickley et al., 1999), and recent work suggests that ozone also contributes indirectly to warming by slowing growth, and subsequently CO_2 uptake, in terrestrial ecosystems (Sitch et al., 2007).

Few studies have simultaneously considered the ecosystem effects of ozone combined with sulfur and nitrogen deposition. The added complication of ongoing changes in the climate and the carbon cycle provide additional challenges for understanding patterns and predicting likely ecosystem impacts of ozone, sulfur, and nitrogen loading. The inability of current models to adequately consider the simultaneous effects of these pollutants has been noted (Aber et al., 2001; Dewar et al., 2009). Despite the lack of adequate models and sufficient experiments to understand multi-factorial variations of these pollutants, evidence supports a joint, interacting role of ozone and nitrogen deposition in a variety of ecosystem effects, including sensitivity to insects and pathogens; frost sensitivity; drought; and fire (Bytnerowicz et al., 2007). Thresholds of nitrogen deposition and ozone concentration likely exist, below which ecosystem effects cannot be detected and at which benefits such as increased ecosystem growth and carbon sequestration are likely. However, the exact levels of these thresholds are not well known, nor are the tipping points above

which negative ecosystem effects sharply increase (Aber et al., 2001). Better identification of such patterns in ecosystem effects could provide vital information for use in future air quality policies.

Mercury Interactions with Atmospheric Sulfur and Nitrogen Deposition

Although most surface waters have very low concentrations, mercury is of environmental concern because it accumulates in living cells and is biomagnified in aquatic and terrestrial food webs to the extent that elevated mercury concentrations are present in fish throughout the United States (Scudder et al., 2009). Mercury is a potent neurotoxin, and human health warnings that suggest limiting the consumption of certain fish due to high mercury levels are widespread in the United States (U.S. EPA, 2007). Atmospheric sulfur and nitrogen are closely linked to mercury because emissions from coal burning are a major source of all three of these pollutants. Moreover, atmospheric sulfur and nitrogen deposition can affect the cycling and bioaccumulation of mercury in ecosystems, primarily because of the interactions of mercury and sulfur.

Great Smoky Mountains National Park protects one of the last wild trout habitats in the eastern United States (photo courtesy of the NPS).

The mercury found in the tissue of biota at high ecosystem trophic levels (e.g., predatory fish and birds) is dominantly in the methyl form (methylmercury), and this is the dominant form found in humans as well. Methylation is the biogeochemical process by which mercury is converted to methylmercury, and most methylation in ecosystems is believed to be carried out by the same bacteria that convert sulfate to sulfide forms. This process tends to proceed in environments (e.g., wetlands, lake-bottom sediments) where sulfate is introduced in runoff and oxygen is absent. Several studies have shown that additions of sulfate increase methylmercury concentrations in waters, and by inference, would be expected to also increase mercury levels in biota (Branfireun et al., 1999; Gilmour et al., 1992; Jeremiason et al., 2006). Similar studies have shown that decreases in sulfate concentrations can likewise decrease methylmercury concentrations in waters and fish tissue, even in the absence of any changes in atmospheric mercury deposition rates (Hrabik and Watras, 2002; Drevnick et al., 2007).

The findings from these studies suggest that improvements in ecosystem health, with respect to mercury bioaccumulation levels, might be achieved by simultaneously decreasing sulfur and mercury deposition levels. These findings also suggest that a multi-pollutant policy, considering both mercury and sulfur, could be used strategically to reduce mercury bioaccumulation in many environmental settings. In addition to direct links between methylmercury and sulfate, studies have found that pH and mercury levels in fish tissue are inversely related in many regions. This suggests that ecosystem acidification and mercury bioaccumulation are also linked, and a variety of mechanisms have been suggested to explain this relationship (Kamman et al., 2004; Wiener et al., 2006; Munthe et al., 2007; Scudder et al., 2009). Another recent study shows that high nitrate concentrations in waters may act to suppress the formation of methylmercury when environmental conditions might otherwise favor high rates of methylation (Todorova et al., 2009), but this type of interaction has not yet been demonstrated in a natural ecosystem where atmospheric deposition is the principal source of nitrogen and sulfur.

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