ISSUES IN ECOLOGY

Published by the Ecological Society of America

Setting Limits: Using Air Pollution Thresholds to Protect and Restore U.S. Ecosystems

Mark E. Fenn, Kathleen F. Lambert, Tamara F. Blett, Douglas A. Burns,
Linda H. Pardo, Gary M. Lovett, Richard A. Haeuber, David C. Evers,
Charles T. Driscoll, and Dean S. Jeffries

Fall 2011

Report Number 14

esa

Setting Limits: Using Air Pollution Thresholds to Protect and Restore U.S. Ecosystems

SUMMARY

More than four decades of research provide unequivocal evidence that sulfur, nitrogen, and mercury pollution have altered, and will continue to alter, our nation's lands and waters. The emission and deposition of air pollutants harm native plants and animals, degrade water quality, affect forest productivity, and are damaging to human health. Many air quality policies limit emissions at the source but these control measures do not always consider ecosystem impacts. Air pollution thresholds at which ecological effects are observed, such as critical loads, are effective tools for assessing the impacts of air pollution on essential ecosystem services and for informing public policy. U.S. ecosystems can be more effectively protected and restored by using a combination of emissions-based approaches and science-based thresholds of ecosystem damage.

Based on the results of a comprehensive review of air pollution thresholds, we conclude:

- *Ecosystem services* such as air and water purification, decomposition and detoxification of waste materials, climate regulation, regeneration of soil fertility, production and biodiversity maintenance, as well as crop, timber and fish supplies are impacted by deposition of nitrogen, sulfur, mercury and other pollutants. The consequences of these changes may be difficult or impossible to reverse as impacts cascade throughout affected ecosystems.
- The effects of too much nitrogen are common across the U.S. and include altered plant and lichen communities, enhanced growth of invasive species, eutrophication and acidification of lands and waters, and habitat deterioration for native species, including endangered species.
- Lake, stream and soil acidification is widespread across the eastern United States. Up to 65% of lakes within sensitive areas receive acid deposition that exceeds critical loads.
- Mercury contamination adversely affects fish in many inland and coastal waters. Fish consumption advisories for mercury exist in all 50 states and on many tribal lands. High concentrations of mercury in wildlife are also widespread and have multiple adverse effects.
- Air quality programs, such as those stemming from the 1990 Clean Air Act Amendments, have helped decrease air pollution even as population and energy demand have increased. Yet, they do not adequately protect ecosystems from long-term damage. Moreover they do not address ammonia emissions.
- A stronger ecosystem basis for air pollutant policies could be established through adoption of science-based thresholds. Existing monitoring programs track vital information needed to measure the response to policies, and could be expanded to include appropriate chemical and biological indicators for terrestrial and aquatic ecosystems and establishment of a national ecosystem monitoring network for mercury.

The development and use of air pollution thresholds for ecosystem protection and management is increasing in the United States, yet threshold approaches remain underutilized. *Ecological thresholds* for air pollution, such as critical loads for nitrogen and sulfur deposition, are not currently included in the formal regulatory process for emissions controls in the United States, although they are now considered in local management decisions by the National Park Service and U.S. Forest Service. Ecological thresholds offer a scientifically sound approach to protecting and restoring U.S. ecosystems and an important tool for natural resource management and policy.

Cover photo credit: *Loch Vale in the Colorado Rocky Mountains. Photo by SteveB in Denver (http://www.flickr.com/people/darkdenver/) and used in this publication under a Creative Commons Attribution license.*

Setting Limits: Using Air Pollution Thresholds to Protect and Restore U.S. Ecosystems

Mark E. Fenn, Kathleen F. Lambert, Tamara F. Blett, Douglas A. Burns, Linda H. Pardo, Gary M. Lovett, Richard A. Haeuber, David C. Evers, Charles T. Driscoll, and Dean S. Jeffries

Introduction

Natural ecosystems have been altered in various ways by nitrogen, sulfur, and mercury deposited in rain, snow, or as gases and particles in the atmosphere. Through decades of scientific research, scientists have documented how local, regional, and global sources of air pollution can produce profound changes in ecosystems. These changes include acidification of soils and surface waters, harmful algal blooms and low oxygen conditions in estuaries, reduced diversity of native plants, high levels of mercury in fish and other wildlife, and decreased tolerance to other stresses, such as pests, disease, and climate change. Advancing our understanding of the linkages among pollutant deposition rates or concentrations, ecosystem effects, and associated policy decisions is a priority in policy-relevant science in the U.S.

Air pollutants that affect human health and ecosystems are primarily emitted from electric power generation, industrial, transportation, and agricultural activities. The benefits and necessities of these activities must be considered in light of the often detrimental effects of atmospheric emissions on human health, visibility, ecosystems, and on the services provided to society by these ecosystems (Table 1). The 1990 Clean Air Act Amendments and other air quality regulations have led to marked declines in emissions of nitrogen, sulfur and mercury. Some emissions from power generation and other sources have decreased by over 50% since the 1970s, even as population and energy demand have increased. As the emissions and deposition of most pollutants have declined, some impacted ecosystems have started to recover. In many parts of the country, however, ecological conditions are still declining due to the increase in other forms of pollution such as ammonia (NH_3), the long term accumulation of sulfur and nitrogen compounds in soils, and the ongoing biomagnification of mercury in food webs.

The purpose of this report is to distill advances in the science of air pollution thresholds and to describe their use to assess, protect and manage the nation's ecosystems and the vital services they provide. We focus here on the environmental impacts of nitrogen, sulfur, and mercury and refer to connections to climate change. The discussion draws on the published research of hundreds of scientists over the past several decades with a focus on U.S. ecosystems and lessons from Canada and Europe.

Air Pollution Thresholds

Thresholds of air pollution in the U.S. have been widely discussed in the scientific literature since the 1970s, when research established that sulfur deposition was above levels at which damage occurs in many sensitive ecosystems in the eastern U.S. More recently, nitrogen deposition has been shown to impact sensitive ecosystem components and processes throughout the United States. Defining the specific concentration or deposition input of an air pollutant that will cause adverse or significant ecosystem effects has been the subject of much scientific research. Pollutants can accumulate with little noticeable impact on plants or animals until major changes occur as a *tipping point* is reached (Box 1). These changes are measured by scientifically determined chemical or biological indicators (Box 2). Such environmental changes might eliminate a single sensitive species, or a broad shift may occur in biodiversity throughout an ecosystem. Once a species or ecosystem has passed a tipping point, a return to the previous state may not be possible.

Air pollution thresholds can be defined based strictly on scientific research (*ecological thresholds*) or based on a balance of policy considerations spanning law, economics, ecological effects, human health and risk assessment (*policy thresholds*) (Box 1) (Figure 1). One tool increasingly used to integrate the science and policy of air pollution thresholds for ecosystem protection and management is critical loads (Box 4).

Advances in the Science of Air Pollution Thresholds

Based on research over the past decade, a strong scientific foundation exists for defining air pollution thresholds using critical loads approaches (Box 4). In the following sections we synthesize the state of the science related to the ecological effects, key indicators, and critical loads approaches for acidifying deposition, nitrogen pollution and mercury contamination.

1. ACIDIFYING DEPOSITION

A. Effects of Acidifying Deposition

Acidifying deposition (or "acid rain") is caused by emissions to the atmosphere of sulfur dioxide (SO_2), nitrogen oxides (NO_x), and other acidifying compounds such as ammonia (NH_3)(see Box 3 for definition of chemical names and symbols). These pollutants return to Earth in rain, snow, fog, mist and gases in forms such as nitric and sulfuric acids and ammonium (NH_4^+) and can have long-term negative impacts to terrestrial and aquatic ecosystems. Ecosystems in the western U.S. have not been greatly affected by acidification because acidifying deposition is relatively low in much of the region and because in many arid or semi-arid regions the soils are relatively insensitive to acid inputs. Some high elevation streams in the Colorado Rockies and the

Box 1. DEFINITION OF TERMS

ACIDIFYING DEPOSITION. Deposition of substances from the atmosphere as rain, snow, fog, or dry particles that have the potential to acidify the receptor medium, such as soil or surface waters. Emissions of sulfur and nitrogen oxides and ammonia are the most common sources of acidifying air pollutants.

ACID NEUTRALIZING CAPACITY. A measure of the ability of a solution to neutralize inputs of strong acids, commonly applied to surface water or soil solution. The acronym ANC is widely used in referring to acid neutralizing capacity.

ATMOSPHERIC DEPOSITION. The transfer of air pollutants from the atmosphere to the Earth's surface. Atmospheric deposition occurs as wet (e.g., rainfall, fog, or snow) and dry deposition (e.g., gaseous or particulate deposition).

BASE SATURATION. The fraction of exchangeable cations in soil which are nonacid forming cations (Ca^{+2}, Mg^{+2}, K^+ and Na^+), also referred to as 'base cations'. The higher the amount of exchangeable base cations in soil, the more acidity can be neutralized.

BIOACCUMULATION. The increase in concentration of a contaminant in an individual organism relative to the surrounding environment or medium (e.g., water, sediment).

BIOMAGNIFICATION. The increase in concentration of a contaminant from lower trophic levels to higher trophic levels in the food chain.

CRITICAL LOAD. The quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge.

ECOLOGICAL THRESHOLD. The dose of a pollutant at which a measurable change occurs in the response of some component of an ecosystem (e.g., NO_3^- leaching at nitrogen deposition of 8 kg/ha/yr).

ECOSYSTEM SERVICES. Benefits to society from a multitude of resources and processes that are supplied by natural ecosystems (e.g., clean drinking water).

ENDPOINT. The ultimate ecological, biological or human condition or process to be protected from harm. Two examples of endpoints are human health and forest sustainability.

INDICATOR. A measurable physical, chemical, or biological characteristic of a resource that may be adversely affected by a change in air quality (e.g., ANC).

NITROGEN SATURATION. Syndrome of effects occurring in an ecosystem caused by an overload of nitrogen, usually from long term atmospheric nitrogen deposition.

POLICY THRESHOLD. A quantitative value of desired ecological condition established by policy and selected based on a balancing of science and land management or policy goals.

SENSITIVE RECEPTOR. The indicator that is the most responsive to, or the most easily affected by a type of air pollution.

TARGET LOAD. The acceptable pollution load that is agreed upon by policy makers or land managers. The target load is set below the critical load to provide a reasonable margin of safety, but could be higher than the critical load at least temporarily.

TIPPING POINT. The point at which an ecosystem shifts to a new state or condition in a rapid, often irreversible, transformation.

Table 1. Linking air pollution impacts to ecosystem services, indicators and thresholds. Ecological thresholds given are typical values that can vary depending on ecological and environmental conditions.

Impact	Ecosystem	Ecological Response	Ecosystem Services Impacted	Indicator	Ecological Threshold
Sulfur and Nitrogen Deposition					
Acidification	Terrestrial	1. Decreased forest growth 2. Increased susceptibility to disease	1. Timber production 2. Climate regulation 3. Biodiversity 4. Resilience to disturbance	Ca: Al^{+3} ratios in soil	<1 – heightened risk to trees >10 – low risk
				Soil percent base saturation	<10% - risk of nutritional deficiencies in sensitive trees >30% - low risk
				Foliar chemistry	<5000 ppm Ca and <700 ppm Mg – limiting to growth of sugar maple
	Freshwater	1. Reduced species richness 2. Degraded water quality	1. Recreational fishing 2. Biodiversity 3. Water quality	Acid neutralizing capacity	0 μeq/L – risk for chronic acidification 20-50 μeq/L – risk for episodic acidification >100 μeq/L – low risk to aquatic biota
				Base cation surplus in soil	0 μeq/L – risk of Al^{+3} leaching to streams
				pH	<6.0 – reduced number of fish species
				Inorganic Al^{+3}	>2 μmol/L – toxic to aquatic biota
				Calcium	<1.5 mg/L – sub-optimal for crustaceans
Nitrogen Deposition					
Nitrogen Enrichment	Terrestrial	1. Loss of sensitive plant species 2. Increase in invasive plants 3. Increased tree mortality	1. Biodiversity 2. Soil fertility	Shifts in lichen communities	5 kg N/ha/yr atmospheric deposition
				N concentration in plant or lichen tissue	1.0% in lichen (*Letharia vulpina*)
				C:N ratio in soil	<20-25 or less - elevated risk of nitrate leaching
	Freshwater	1. Loss of sensitive diatom (single-celled algae) species 2. Degraded water quality	1. Biodiversity 2. Water quality	Shifts in diatom communities	1.5 kg N/ha/yr wet deposition
				Nitrate concentrations	<2 μeq/L – low risk >20 μeq/L - degraded
	Coastal	1. Increased algal blooms 2. Decreased dissolved oxygen	1. Habitat preservation 2. Commercial fishing 3. Recreational fishing 4. Swimming, tourism, aesthetics	Dissolved nitrogen	High (≥1 mg/L) Medium (≥0.1 and <1 mg/L) Low (≥0 and <0.1 mg/L)
				Dissolved phosphorus	High (≥0.1 mg/L) Medium (≥0.01 and <0.1 mg/L) Low (≥0 and <0.01 mg/L)
Mercury Deposition					
Mercury toxicity	Terrestrial	1. Toxicity to fish-eating wildlife 2. Toxicity to wildlife	Wildlife health	Hg in songbirds	1.3 μg/g in blood
				Hg in bats	10.0 μg/g in hair
	Freshwater	1. Mercury bioaccumulation	1. Recreational fishing 2. Food production 3. Human health	Hg in fish	0.2 - 0.3 μg/g
			Fish and wildlife health	Hg in diet	0.16 μg/g in prey fish
				Hg in fish-eating birds	3.0 μg/g in blood

Sierra Nevada Mountains do experience acidic episodes when pollutants retained in the snow pack over the winter are released into soils and streams during snowmelt. In the eastern United States, depletion of available calcium and magnesium pools and acidification of forest soils is widespread and well documented in the Appalachian Mountains, including the Catskills and the Adirondacks, and in the Shenandoah Mountain region of West Virginia.

Mountain forests of the northeastern and southeastern United States receive high rates of acidifying deposition due to frequent exposure to acidic clouds, fog, rain and snow. Changes associated with acidifying deposition have reduced the ability of some tree species to cope with the cold temperatures common to these mountain environments. This effect contributed to large-scale red spruce deaths in these regions in the 1980s and 90s, and remains a problem today. In eastern U.S. hardwood forests at lower elevations, many sugar maple, white ash, flowering dogwood, and other trees have high calcium requirements and therefore are also sensitive to acidification. Tree declines have negative consequences for forest productivity and ecosystem services, including timber production and climate regulation (lower productivity means less removal of carbon dioxide from the atmosphere). Research has attributed sugar maple declines in western Pennsylvania to acidification acting in concert with insect outbreaks, and research in New Hampshire has shown improved growth and reproduction of sugar maple, and less frost damage to red spruce, when calcium was added to an acidified forest for experimental purposes.

Acidification of sensitive surface waters has resulted in well documented adverse effects on fish, zooplankton, aquatic insects, microorganisms, and other aquatic biota. In many sensitive areas receiving elevated acidifying deposition, surface waters are too acidic to support any fish species. The reduction in the number of aquatic species and in the number of fish supported diminishes biodiversity and recreational fishing opportunities. Long-term research on acidification impacts on forests, lakes and streams has produced a wealth of data, from which are drawn the most commonly applied indicators for assessing acidification status and effects (Table 1). Although terrestrial and aquatic indicators are treated separately below, recognition should be given to the connection of soil acidification to aquatic acidification.

Box 2. INDICATORS AND AIR POLLUTION THRESHOLDS

Just as physicians use a range of diagnostic measurements to monitor human health, scientists track chemical and biological indicators to monitor ecosystem health. When many different studies confirm an association between a pollutant amount and an ecosystem response, threshold pollutant levels can often be identified for indicators that signal likely problems. Chemical indicators are often used as surrogates for biological effects because chemical indicators are typically simpler and less expensive to measure. Chemical indicators are imperfect surrogates since accurate prediction of just how plants and animals will respond to chemical changes in their environment is not always possible.

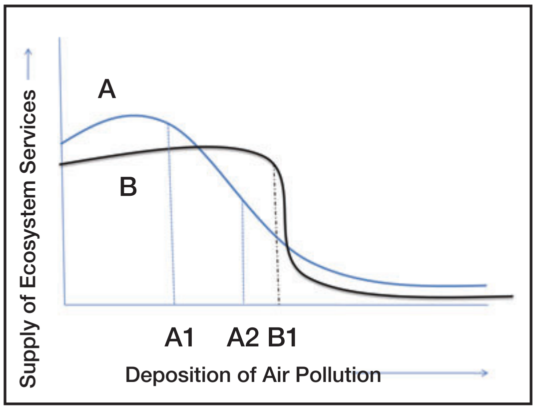

Figure 1. Conceptual representation of how ecological and policy thresholds may be developed. Both lines show estimates of ecosystem degradation as pollutants increase in ecosystems. Line "A" represents a gradual decline in ecosystem condition, where managers, policy makers, and regulators can set policy thresholds at any number of different points depending on goals (for example, A1, at beginning of decline or A2, at midpoint of decline). Line "B" represents a rapid decline in ecosystem condition, with a clearly identified, ecological threshold at which a tipping point occurs (B1).

B. Indicators - Acidifying Deposition

Indicators of Soil Acidification and Forest Health

One way to assess the risk to acid sensitive tree species such as red spruce and sugar maple is by tracking chemical indicators in the soil and in the leaves and needles of plants (i.e.,

Box 3. CHEMICAL NAMES AND SYMBOLS, AND UNITS OF MEASURE

Chemical Names and Symbols:
Sulfur dioxide, SO_2
Nitrogen dioxide, NO_2
Nitrogen oxides, NO_x
Sulfur oxides, SO_x
Ammonia, NH_3
Ammonium, NH_4^+
Mercury, Hg
Methylmercury, MeHg
Sulfate, SO_4^{-2}
Nitrate, NO_3^-
Dissolved organic carbon, DOC
Phosphorus, P
Nitrogen, N
Carbon, C
Aluminum, Al^{+3}
Calcium, Ca^{+2}
Magnesium, Mg^{+2}
Sodium, Na^+
Potassium, K^+

Calcium to aluminum ratio, Ca:Al
pH, a measure of acidity or hydrogen ion concentration
Nutrient ratios (e.g., N:P, N:Ca, C:N)

Units of Measure:
Equivalents per hectare per year, eq/ha/yr
Kilograms per hectare per year, kg/ha/yr
Parts per million, ppm
Microequivalents per liter, µeq/L
Milliequivalents per square meter per year meq/m^2/yr
Micrograms per liter, µg/L

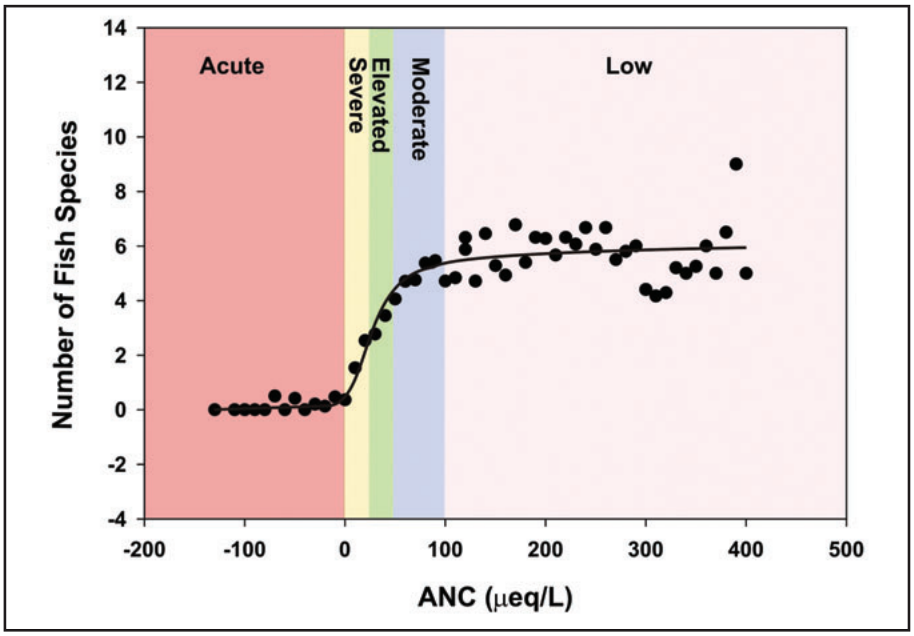

Figure 2. *Number of fish species per lake as a function of acid neutralizing capacity (ANC) in Adirondack lakes. The data are presented as the mean of species richness for every 10 µeq/L ANC class. Lakes are also classified into five descriptive categories ranging from low to acute impacts. (Adapted from: Sullivan, T.J. and others 2006. Assessment of the Extent to Which Intensively-Studied Lakes are Representative of the Adirondack Mountain Region. Final Report 06-17. New York State Energy Research and Development Authority. Albany, NY).*

foliage) (Table 1). Three elements naturally present in soils, calcium (Ca^{+2}), magnesium (Mg^{+2}), and aluminum (Al^{+3}), influence the extent to which trees and other plants may be adversely affected by acidifying deposition. Calcium and magnesium are nutrients needed for a variety of plant functions and their supply helps neutralize acid inputs to soils, whereas Al^{+3} can be harmful to plants at high concentrations when present in the readily available exchangeable form. Acid deposition slowly removes readily available exchangeable Ca^{+2} and Mg^{+2} from soils and replaces them with exchangeable Al^{+3} and hydrogen ion (or acidity), setting off a cascade of adverse changes.

In general, greater availability of Ca^{+2} and Mg^{+2} and low Al^{+3} provides favorable conditions for many acid-sensitive tree species such as sugar maple and red spruce. *Calcium to aluminum ratio* (Ca:Al) in soils and soil solutions is one indicator used to assess the health risk to acid sensitive tree species such as red spruce and sugar maple. *Soil percent base saturation* is another useful indicator for assessing sensitivity and extent of acidification. Scientists generally concur that where soil percent base saturation is low there is a high risk of damage to the vitality of sensitive tree species due to nutritional deficits resulting from acidification. The risks to forest vegetation associated with a range of Ca:Al ratios and soil percent base saturation values are shown in Table 1. Other studies have focused on the concentration of exchangeable Ca^{+2} and Mg^{+2} as a useful indicator since soils can have widely varying amounts of these nutrients that are essential to the health of forest vegetation. *Concentrations of Ca^{+2} and Mg^{+2}* in the leaves and needles of plants (foliage) have recently been identified as valuable indicators for evaluating acid deposition impacts. For example, low concentrations of these nutrients have been identified as limiting the growth of sugar maple (Table 1).

Indicators of Acidification in Aquatic Ecosystems

Indicators of acidification in lakes and streams are generally based on changes in water chemistry. Water chemistry strongly affects the numbers and types of aquatic organisms that are present in a water body. The indicators most commonly used to track changes in surface water acidification are ANC, pH, and/or concentrations of key elements.

Acid neutralizing capacity (ANC) is a commonly used chemical indicator of lake or stream sensitivity to acidification. ANC, measured in microequivalents per liter (µeq/L; See Box 3 for a list of chemical units of measure), characterizes the ability of water to neutralize strong acids including those introduced by atmospheric deposition. ANC is a good general indicator of acidity-related water quality because values are typically strongly correlated with pH, Al^{+3} concentrations, and Ca^{+2} concentrations. Specific concern levels have been identified and are used to estimate critical loads (Table 2). The diversity of fish species declines precipitously with decreases in ANC in Adirondack Lakes (Figure 2). In Shenandoah National Park (Virginia) streams researchers found that one fish species, on average, is lost for every 21 µeq/L decline in ANC. Recent studies have demonstrated that another useful chemical indicator is *base cation surplus*. Low values indicate that the soil has become sufficiently acidified to enable toxic forms of aluminum to be transported from the soil into streams at concentrations of concern.

The pH value of a water body is a fundamental measure of acidity or the hydrogen ion concentration. A pH of 7 is neutral, and pH values below 7 are increasingly acidic while values above 7 are increasingly basic or alkaline. Like ANC, decreases in pH are associated with decreases in the richness of aquatic species (Table 1). Studies have shown that in lakes of the Adirondack Mountains of New York and the White Mountains of New Hampshire, one fish species is lost for every pH decline of 0.8 units as values decrease from 6 to 4. Few fish species can survive at pH values of 4 or less (Figure 3).

Decreases in pH and ANC are often paralleled by changes in element concentrations including increases in Al^{+3} concentrations and decreases in Ca^{+2}. High dissolved *Al^{+3} concentrations* can have toxic effects on many types of aquatic biota, and at extreme levels few aquatic species can survive (Table 1). Organic forms of Al^{+3} are much less toxic than inorganic forms. Emerging research suggests that *Ca^{+2} concentrations* in streamwater are also an important biological indicator. Acidifying deposition has accelerated the leaching of Ca^{+2} from soils to surface waters gradually decreasing the available pool of Ca^{+2} in soils and lowering Ca^{+2} concentrations in runoff. This soil depletion together with decreases in leaching associated with declines in acidifying deposition is contributing to decreases in surface water Ca^{+2}. Many lakes in the boreal forest of the Canadian Shield now have Ca^{+2} concentrations that are considered sub-optimal for water fleas, crayfish and other crustaceans and may be limiting the species richness of lakes in this region.

C. Critical Loads – Acidifying Deposition

Critical loads represent the deposition rate that can occur without surpassing tipping points for a given species or ecosystem based on established indicators and effect levels. The critical load for a specific pollutant or group of pollutants will vary depending on differences in landscape sensitivity and in the endpoints for which the critical loads are calculated (e.g., forest soils, lake chemistry).

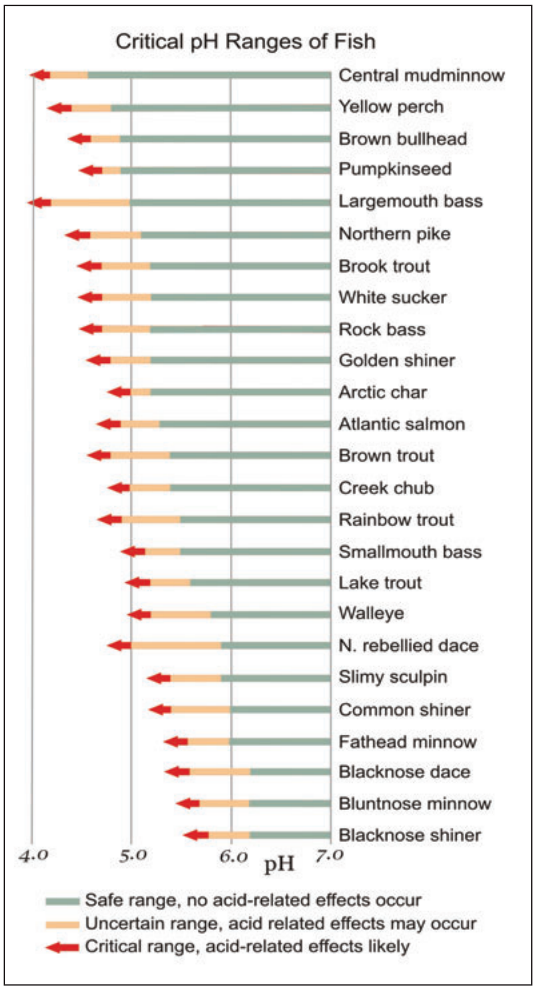

Figure 3. Critical aquatic pH ranges for fish species. (Source: Baker, J.P. and Christensen, S.W. 1991. pp. 83-106, In: Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Charles, D.F. (ed). Springer-Verlag, New York. Figure redrawn in Jenkins, J. and others 2005. Acid Rain and the Adirondacks: A Research Summary. October, 2005. Adirondack Lakes Survey Corporation, Ray Brook, NY). Image used with kind permission of Springer Science+Business Media.

Advances in understanding of chemical and biological indicators of acidification have supported the development of critical loads for sulfur and nitrogen in parts of the U.S. and Canada.

Table 2. Expected ecological effects and concern levels in freshwater ecosystems at various levels of acid neutralizing capacity (ANC). (Source: USEPA)[a].

Category Label	ANC level (μeq/L)	Expected Ecological Effects
Low Concern (No Effect)	>100	Fish species richness may be unaffected. Reproducing brook trout populations are expected where habitat is suitable. Zooplankton communities are unaffected and exhibit expected diversity and distribution.
Moderate Concern (Minimally Impacted)	50-100	Fish species richness begins to decline (sensitive species are lost from lakes). Brook trout populations are sensitive and variable, with possible sub-lethal effects. Diversity and distribution of zooplankton communities begin to decline as species that are sensitive to acid deposition are affected.
Elevated Concern (Episodically Acidic)	0–50	Fish species richness is greatly reduced (more than half of expected species are missing). On average, brook trout populations experience sub-lethal effects, including loss of health and reproduction (fitness). During episodes of high acid deposition, brook trout populations may die. Diversity and distribution of zooplankton communities declines.
Acute Concern (Chronically Acidic)	<0	Near complete loss of fish populations is expected. Planktonic communities have extremely low diversity and are dominated by acid-tolerant forms. The numbers of individuals in plankton species that are present are greatly reduced.

[a] Based on data from Southern Appalachian streams and from Shenandoah National Park.

Box 4. UNDERSTANDING THE CRITICAL LOADS APPROACH

Critical loads, and other approaches that use models or empirical observations to link deposition with effects, provide tools that enable resource managers and policymakers to evaluate tradeoffs between the costs of more stringent emissions controls and the benefits of ecosystem services provided by healthy ecosystems.

A critical loads approach can be used to synthesize scientific knowledge about air pollution thresholds that cause adverse impacts or ecosystem change. Describing air pollutant effects on ecosystems in critical load terms quantifies estimates of “cause and effect” in a way that allows researchers to communicate science to air quality regulators and natural resource managers. Critical loads are most commonly applied to evaluate the effects of nitrogen and sulfur pollutants and their associated acidity or the eutrophying effects of nitrogen. When critical loads are exceeded there is increased risk for a range of problems including ecosystem acidification, excess nitrogen effects, declines in forest health, and changes in biodiversity.

Critical loads are typically expressed as deposition loading rates of one or more pollutants in amount per area per year (e.g., kilograms per hectare per year (kg/ha/yr)). Critical loads are based on changes to specific biological or chemical indicators such as species composition of a given ecosystem (e.g., grassland) or biotic community (e.g., understory plants or tree-dwelling lichens) or acid neutralizing capacity (ANC) in soils, streams or lakes. Because different sensitive receptors (e.g., forest soils, high elevation lakes, species of lichen) or species may have varying sensitivities to air pollutant loads, multiple critical loads can be used to describe a continuum of impacts with increasing deposition at a given location (See Figure 5).

In addition, even for the same organism, multiple critical loads may be associated with biological thresholds for different negative effects, such as stunted growth, reduced reproduction, and increased mortality. Several different threshold levels may therefore be included in a critical load assessment. The policymaker, air regulator, or land manager can assess all the critical loads (science-driven ecological thresholds) and select target loads (policy thresholds) based on the level of ecosystem protection desired, economic considerations, and stakeholder input at a given location.

Forests

U.S. researchers use models to develop critical loads for forest soil acidification (Box 4). A recent study estimated the critical acid loads for forest soils across the conterminous U.S. The critical acid loads for S and N throughout the Appalachian Mountain Range and Florida are estimated to be less than 1,000 eq/ha/yr (critical loads for combined sulfur and nitrogen are expressed in terms of ionic charge balance as equivalents per hectare per year). This study estimated that about 15% of U.S. forest soils exceed their critical acid load by at least 25% including much of New England, West Virginia, and parts of North Carolina. By comparison, critical load modeling in Canada estimated that 30 to 40% of upland forest areas in Canada are in exceedance of the critical load for acidification, while more than 50% are in exceedance in eastern Canada (Ontario, Quebec, New Brunswick, Nova Scotia and Newfoundland).

Surface Waters

Regional critical loads for surface waters have been developed for acidifying deposition of sulfur and nitrogen in sensitive regions of the Adirondack Mountains of New York and in the central Appalachians of Virginia and West Virginia. The median critical load for a target ANC of 50 µeq/L is 129 milliequivalents per square meter per year (meq/m^2/yr) in the Adirondacks and 45 meq/m^2/yr in the central Appalachians with values ranging from less than 0 to over 1,000 meq/m^2/yr in relatively insensitive ecosystems. The number of aquatic ecosystems exceeding the critical loads is still quite high, but has declined with decreases in acid deposition from the early 1990s to the late 2000s (Figure 4). Currently, 44% of Adirondack lakes evaluated exceed the critical load and in these lakes recreationally valuable fish species such as trout are missing due to acidification. In the Shenandoah area, 85% of streams evaluated exceed the critical load resulting in losses in fitness in fish species such as the blacknose dace. The persistence of critical load exceedances despite significant decreases in SO_2 emissions is related to continued high inputs of acidifying NO_x, low initial ANC conditions, and soil depletion of nutrient cations (Ca^{+2} and Mg^{+2}) that have left many watersheds more sensitive to acid deposition over time.

A similar study of 2053 lakes in six northeastern states and four eastern Canadian provinces estimated critical loads for acidifying deposition of sulfur and nitrogen for a target ANC of 40 µeq/L. Results show that 28% of the lakes studied have a critical load in the categories of ≤20 and 20–40 meq/m^2/yr, suggesting vulnerability to acidification with relatively moderate atmospheric deposition. It is estimated that the critical load is exceeded in 12% of the study lakes, based on deposition levels in 2002. These studies point to the importance of long-term monitoring and research for assessing the impact of emissions control programs on deposition and ecological recovery (Box 5).

2. NITROGEN

A. Effects of Excess Nitrogen on Ecosystems

The nitrogen gas that makes up most of the Earth's atmosphere is inert, with little impact on ecosystems. Nitrogen converted to its reactive forms such as NH_3 and NO_x, however, can cause profound biological changes. Activities such as fertilizer manufacturing, intensive livestock production and the burning of fossil fuels convert nitrogen to these reactive forms which can then enter and potentially over-fertilize ecosystems. This can lead to problems such as algal overgrowth in lakes, reduced water quality, declines in forest health, and decreases in aquatic and terrestrial biodiversity by favoring "nitrogen loving" species at the expense of other species with low nitrogen preferences. For example, most estuaries and bays in the Northeast U.S. and Mid-Atlantic regions experience some degree of eutrophication (where excess nutrients promote a proliferation of plant life, which can deplete oxygen in the deeper waters), as a result of nutrients from atmospheric deposition and agricultural, urban and industrial runoff. Excess nitrogen can also change species composition. In Waquoit Bay, Massachusetts elevated nitrogen allows tall cord grass to thrive but not eelgrass, which decreases critical fish habitat.

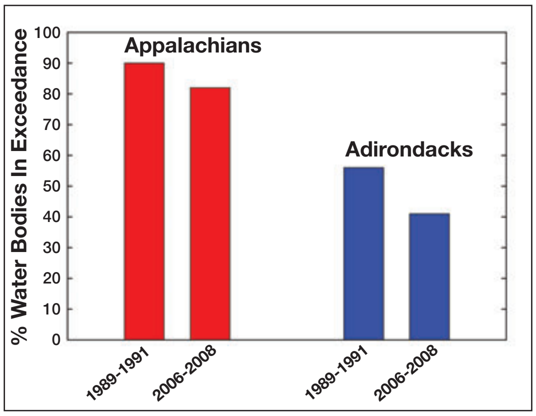

Figure 4. *Percentage of lakes in exceedance of the critical load for sensitive eastern US surface waters in the Adirondacks (169 lakes in NY) and the central Appalachians (92 streams in VA and WV). The percent exceeding the critical load has declined as emissions and deposition have been decreasing (Source: Jason Lynch- USEPA).*

Adding nitrogen to forests whose growth is typically limited by its availability may appear desirable, possibly increasing forest growth and timber production, but it can also have adverse effects such as increased soil acidification, biodiversity impacts, predisposition to insect infestations, and effects on beneficial root fungi called *mycorrhizae*. As atmospheric nitrogen deposition onto forests and other ecosystems increases, the enhanced availability of nitrogen can lead to chemical and biological changes collectively called "*nitrogen saturation*." As nitrogen deposition from air pollution accumulates in an ecosystem, a progression of effects can occur as levels of biologically available nitrogen increase (Figure 5). Because of the multiple potential effects of nitrogen deposition in terrestrial and aquatic ecosystems, the ecosystem services affected vary depending on the sensitive receptors found within a given ecosystem and the level of atmospheric deposition. Prominent examples of affected ecosystem services in forests include timber production, climate regulation, recreational use, and biodiversity loss. In

Box 5. THE ROLE OF LONG-TERM MONITORING AND RESEARCH

Long-term studies measure baseline ecosystem conditions and trends and can show how ecosystems respond when atmospheric deposition decreases below a threshold that was previously exceeded. The trajectory of recovery is not always consistent with model simulations, illustrating the importance of long-term monitoring and research to improve the capabilities of simulation models. A number of regional- and national-scale air, water, soil, and biota monitoring networks collect high-quality data that are useful in assessing ecosystem thresholds. However, current efforts are not enough to provide continuous data at sites across the country, and often lack the coordination needed to effectively combine datasets for maximum benefit. We recommend that existing monitoring and research programs be continued, expanded and better integrated. Some examples of federal monitoring programs include:

- Federal agency air pollution monitoring programs such as the Interagency Monitoring of Protected Visual Environments (IMPROVE) *http://vista.cira.colostate.edu/improve/* and the National Atmospheric Deposition Monitoring Program (NADP), *http://nadp.sws.uiuc.edu/*, and the Clean Air Status and Trends Network (CASTNET) http://www.epa.gov/castnet/
- The U.S. Forest Service's Forest Inventory Analysis and Forest Health Monitoring (FIA/FHM)
- The Environmental Protection Agency's Temporally Integrated Monitoring of Ecosystems/Long-term Monitoring (TIME/LTM) network *http://www.epa.gov/airmarkt/assessments/TIMELTM.html* and National Surface Water Surveys
- The U.S. Geological Survey's National Water-quality Assessment Program (NAWQA) *http://water.usgs.gov/nawqa/* and Biomonitoring of Environmental Status and Trends (BEST) programs
- The U.S.Forest Service's wilderness area surface water-monitoring programs *http://www.fs.fed.us/waterdata/*
- The NSF-sponsored National Ecological Observation Network (NEON) *http://www.neoninc.org/* and Long-Term Ecological Research (LTER) network - *http://www.lternet.edu/*

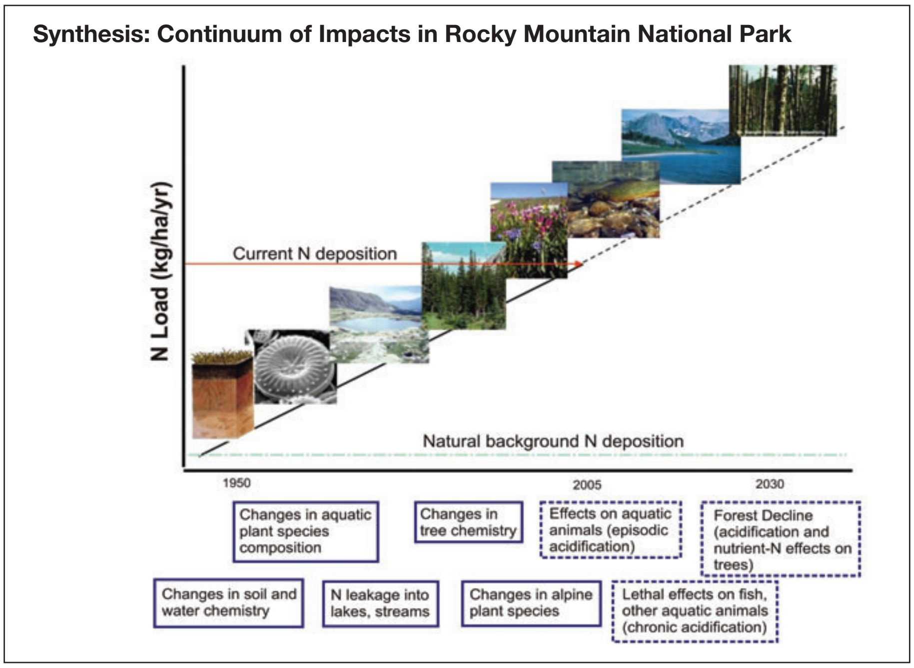

Figure 5. Continuum of nitrogen deposition impacts as demonstrated from past observations and potential future effects in Rocky Mountain National Park in Colorado. As ecosystem nitrogen accumulation continues, additional acidification or eutrophication impacts occur to various ecosystem receptors. Note that the trajectory line is conceptual even though the effects below the current nitrogen deposition level have been documented. Similar trajectories of additional ecosystem effects as nitrogen accumulates in the ecosystem likewise occur in other ecological regions. (Figure courtesy of Ellen Porter, National Park Service).

freshwaters affected ecosystem services include recreational fishing, other forms of recreation, and provision of high quality drinking water (Table 1).

B. Indicators – Nitrogen Pollution

Nitrogen impacts on ecosystems can be identified by examining changes in biota, or by measuring chemical indicators. Ideally, a chemical indicator provides a warning suggesting that sensitive biota are at risk before biological harm occurs.

Nitrate Leaching

One of the most notable symptoms of nitrogen saturation is increased leaching of nitrogen from soils into lakes, streams and groundwater, primarily in the form of nitrate (NO_3^-). Streamwater NO_3^- concentration is a useful and simple indicator of the nitrogen status of a catchment because this measure integrates many nitrogen cycling processes that occur within the catchment, including the processing of atmospheric nitrogen deposition. In nitrogen limited ecosystems in the western United States, U.S. Forest Service land managers have set *policy thresholds* of <2 µeq/L NO_3^- in surface waters indicating that the waters are in good condition relative to the effects of excess nitrogen leaching and >20 µeq/L as a concern level indicating potentially over-enriched systems. Studies in Europe and the northeastern U.S. show that nitrogen leaching begins to increase in forests receiving levels of atmospheric nitrogen deposition greater than 8-12 kilograms of nitrogen per hectare per year (kg N/ha/yr), although not all forests receiving those levels of deposition show NO_3^- leaching, due to land disturbance history, the presence of wetlands and other characteristics.

Nutrient Ratios

Other commonly used chemical indicators of nitrogen enrichment include nutrient ratios in foliage such as nitrogen:phosphorus (N:P), nitrogen:calcium (N:Ca^{+2}), or carbon:nitrogen (C:N). C:N ratios in organic or mineral horizons of the soil also indicate ecosystem nitrogen status or the predisposition to nitrogen saturation. The C:N ratio of the soil and the growth rate of the forest influence nitrogen leaching. Forests with soil C:N ratios less than 20-25 (indicating high N availability) are more likely to exhibit nitrogen leaching than forests with higher C:N ratios.

Biological Indicators

Biological indicators of nitrogen over-enrichment or eutrophication include shifts in species or biological communities, enhanced establishment of invasive species, or other measures of biodiversity change (Box 6). Local extinction of sensitive species or functional groups can also occur. The most sensitive organisms exhibiting such changes in response to nitrogen enrichment in freshwater ecosystems such as lakes are small single-celled algae known as diatoms. Even a small amount of additional nitrogen deposition from air pollution that is transported to waters can induce major shifts in the species of diatoms. Each diatom species has specific patterning in the shell, so by studying lake sediment cores from western lakes that extend back 100 years or more, researchers have been able to document whether recent diatom species shifts correspond with changes in nitrogen deposition. Tree-inhabiting lichens (epiphytes) are highly sensitive indicators of nitrogen air pollution in forests and woodlands. The lowest critical loads for nitrogen effects are typically based on diatom or lichen community responses, making them good early warning indicators for ecosystem changes. Nitrogen also affects the biodiversity of herbaceous or grassland plant communities at relatively low levels (Box 6).

Nitrogen effects on these biological indicators are linked to changes in ecosystem functions including alteration of food webs, increased risk of fire, and reduction in important nursery habitat for commercial fisheries. For example, among the nitrogen sensitive lichen species are those that are vital forage species for deer, elk, and the northern flying squirrel; the latter is the major prey of the federally endangered spotted owl. Nitrogen deposition effects on plant biodiversity can result in major impacts on ecosystems, including enhanced invasion by exotic grasses, increased fire danger, vegetation type change, and disappearance of biological species that depend on declining native plant species and communities (Box 6). Finally, negative impacts to eelgrass beds from the over-enrichment of coastal waters can diminish the quality of important nursery, habitat, and feeding grounds for commercially important fish and shellfish in the eastern U.S., such as scallops.

C. Nitrogen Critical Loads

Air pollution thresholds at which excess nitrogen effects on ecosystems occur can be determined using field studies or estimated by modeling. Nitrogen critical loads can also be extended over wide geographic areas or predicted through time by use of models. Critical loads based on field observations across spatial gradients of varying air pollution exposure or from field experiments are known as empirical critical loads. Empirical critical loads for nitrogen deposition effects in selected ecoregions of North America are presented in Table 3. Recent studies demonstrate that exceedance of empirical critical loads for nitrogen is common across the U.S.

By comparing modeled and empirical critical load values to current and future deposition data and estimates, policymakers can assess current ecosystem condition, set goals for ecosystem recovery, and track improvement. This information can also aid decision making processes for air pollution controls or mitigation programs for damaged ecosystems. For example, the low end of the critical load range in Mediterranean California mixed conifer forests (3 kg N/ha/yr) describes the point where impacts begin in the most sensitive parts of these ecosystems, specifically, changes in lichen communities. Such a low nitrogen critical load provides a ‘canary in the coal mine’ threshold that is indicative of initial ecosystem responses to added nitrogen. Ecoregions in the United States where lichen communities are likely affected by nitrogen air pollution based on nitrogen deposition in

Box 6. BIODIVERSITY

Biological diversity (or “biodiversity”) may simply be defined as the species richness of a geographic area. Biodiversity loss has accelerated in modern times due to land use change, the introduction of invasive species and other disturbances. Climate change and air pollution also contribute to changes in plant community composition and biodiversity. In polluted regions, the occurrence of sensitive species may decrease and lead to replacement by pollution-tolerant species. When air pollution alters the biodiversity or the composition of biological communities, detrimental effects on the provision of valued ecosystem services can occur. The implications for biodiversity shown by long-term studies of acid deposition and nitrogen pollution are highlighted in the case studies below.

Acid Rain: Diminishing Aquatic Diversity in the Northeast
Aquatic organisms vary in sensitivity to acidity with sensitive species showing limitations at pH 6.0 and many organisms declining in abundance and richness at pH levels of 5.5 and lower. As acidity increases, sensitive species or sensitive life history stages of species either die or seek refuge in less-acidified habitats leaving the original habitat less productive and diverse. The impacts are most severe in sensitive high elevation ecosystems that have experienced chronic deposition. Of the 53 fish species recorded in lake surveys in the Adirondack Mountains of New York, half are absent from lakes with pH less than 6.0. Recreational fishes, such as Atlantic salmon, tiger trout, bluegill, walleye and alewife, are among those absent from low-pH lakes. These acidity effects can extend further down the food chain. In lakes of the Adirondacks and the White Mountains (New Hampshire) an average of 2.4 zooplankton species are lost with each pH unit decrease. Long-term monitoring of acidifying deposition and surface water chemistry confirm that decreased emissions of acidic pollutants have resulted in lower deposition and some recovery in pH. The long and complex process of biological recovery including the restoration of soil base saturation is only just beginning.

Nitrogen Pollution, Plant Communities and Biodiversity in California
Biodiversity of plant communities is sensitive to N added by air pollution. Nitrogen-loving species are often favored and increase in prominence as ecosystem nitrogen availability increases. Forests and woodlands in many regions of the world show large changes in epiphytic lichen communities in response to chronic atmospheric nitrogen deposition. These lichen community impacts occur at nitrogen pollution thresholds as low as 3-6 kg/ha/yr. Ecologically important lichen species have been eliminated from forests over large areas of California. Similarly, in coastal central California, native serpentine grassland plant communities are exterminated when the N deposition load is 6 kg/ha/yr or greater and are replaced by exotic grasses and other native plant species. These changes result in the loss of the plant species needed for reproduction and survival of the endangered Bay Checkerspot butterfly, and has caused local population extinctions of the butterfly. Likewise, nitrogen from air pollution at levels around 8 kg/ha/yr in desert and scrub vegetation communities of California favors the growth of exotic annual grasses, crowding out native species and increasing fire risk in some areas.

Table 3. Critical Loads (CL) of Nitrogen Deposition for Effects on Selected North American Ecosystems

Ecosystem	Chemical or Biotic Response	CL for N Deposition (kg N/ha/yr)
Arctic Tundra	Plant community change; grass growth	1-3
Arctic Tundra	Changes in shrub, bryophyte, lichen cover	6-11
Boreal Shrublands	Decrease in shrub cover; increase in grass cover	6
Northern Forests	Change in soil community structure	5-7
Northern Hardwood and Coniferous Forests; Eastern Temperate Hardwood Forests	Increased surface water nitrate leaching	8
Northwest Forested Mountains and Mediterranean California Mixed Conifer Forests	Lichen community change from oligotrophic to eutrophic species dominance	3-5
Alpine	Changes in herb and grass species composition	4-10
Great Plains Tall Grass Prairie	Change in biogeochemical N cycling, plant and insect community shifts	5-15
Mediterranean California Mixed Conifer Forests	Increased surface water nitrate leaching	17
Mediterranean California Serpentine Grasslands	Native herbs replaced by annual grasses; loss of checkerspot butterfly habitat	6
Rocky Mountain Western Lakes	Freshwater eutrophication	2

From: Pardo, L.H., Robin-Abbott, M.J., and Driscoll, C.T., eds. 2011. Assessment of nitrogen deposition effects and empirical critical loads of nitrogen for ecoregions of the United States. Gen. Tech. Rep. NRS-80. Newtown Square, PA: U.S. Department of Agriculture, Forest Service, Northern Research Station.

exceedance of the critical load for lichen effects are shown in Figure 6.

Recent research has shown that by stimulating increased growth of non-native grasses, nitrogen deposition may increase the frequency of wildfires in southwestern U.S. desert areas because these grasses provide fuel to sustain the spread of fire in areas with little or no previous fire history. Simulation models have estimated that the lowest threshold level of nitrogen that initiates these changes in pinyon-juniper ecosystems is about 3 kg N/ha/yr of nitrogen deposition. Fire risk increases exponentially above this level to about 5.7 kg N/ha/yr, at which level grasses are generally fully established. This provides an example of how policymakers or stakeholders could select a variety of *policy thresholds* (3.0, 5.7 kg/ha/yr, or levels in between) depending on risk tolerance and goals for protecting native vegetation in different types of ecosystems. Nitrogen deposition currently exceeds these *ecological thresholds* in many southwestern U.S. ecosystems, indicating the importance of this information in air pollution policy or land management decision making in this region.

Another example of nitrogen deposition effects relative to *ecological thresholds* occurs in the Colorado Rocky Mountains. In this region, alpine vegetation has begun to shift toward a higher proportion of grasses. This shift occurs at a threshold of around 4 kg N/ha/yr, which approximates current nitrogen deposition levels in areas of the Rockies most influenced by agricultural and urban emissions. The responses of individual plant species to nitrogen (critical load of 4 kg N/ha/yr) is a much more sensitive indicator of nitrogen effects than soil acidification (critical load of 10-15 kg N/ha/yr). Nitrogen deposition also has initiated shifts in diatom species to those that favor higher nitrogen levels in some high-elevation lakes at levels of 1.4 to 1.5 kg N/ha/yr as wet deposition in national parks and Class I Wilderness areas in Colorado, the greater Yellowstone Ecosystem, and the eastern Sierra Nevada Mountains. This threshold is among the lowest identified for any ecosystem changes resulting from nitrogen deposition, making diatoms ideal early warning indicators of decline in aquatic ecosystem condition from nitrogen air pollution. Current nitrogen deposition levels are higher than these thresholds for most of the western U.S.

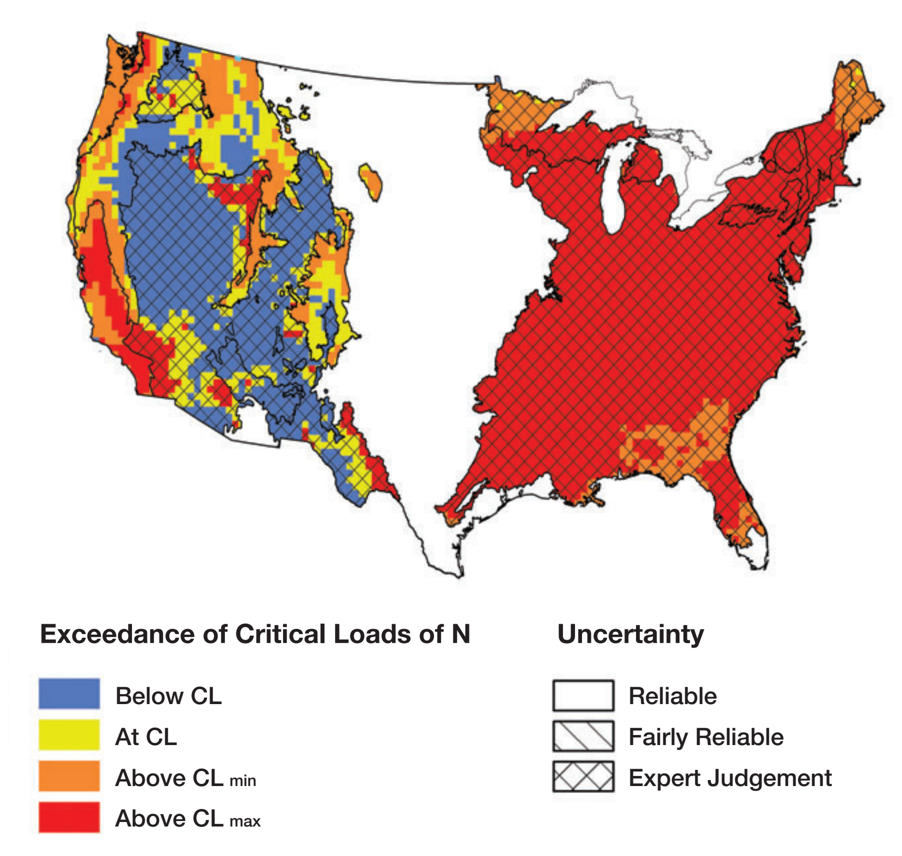

Figure 6. *Lichen Based Critical Load Exceedance Map. Areas shown in red and orange received atmospheric nitrogen deposition at levels deleterious to communities of epiphytic (tree dwelling) lichens. This map shows that these effects occur in over half of the forested land area, including urban forests, of the continental U.S. Levels of certainty in the critical load exceedance estimates vary among ecoregions depending on the amount of available lichen community data. (From Pardo, L.H. and others 2011. Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States. Ecological Applications, doi: 10/1890/10-234.1).*

3. MERCURY POLLUTION

A. Effects

Mercury is a naturally occurring metal and a local, regional and global pollutant. Mercury emissions from electric utilities, incinerators and industrial manufacturing are among the largest sources of mercury to the environment in the U.S. Mercury in the air and water are not direct public health risks at levels commonly found in the U.S. The risk to human and ecological health typically occurs through consumption of mercury-contaminated fish and other biota. Inorganic mercury (Hg) is deposited to the landscape, transported from soils to wetlands and surface waters, and converted by bacteria to methylmercury (MeHg) – the organic form of mercury that is readily absorbed by fish and other organisms. Once ingested, mercury can bioaccumulate in organisms and biomagnify through the food web to elevated concentrations in fish and other organisms that are consumed by people and wildlife. Fish contamination by MeHg poses a widespread problem in freshwater, and in coastal and marine recreational and commercial fisheries.

As mercury sampling in lakes and rivers has expanded, the extent of waters known to be impaired by mercury pollution has increased. In 2008, all 50 states, one U.S. territory, and three Native American tribes issued mercury advisories for human fish consumption covering 16.8 million lake acres and 1.3 million river miles. That was a 19% increase in lake area under advisory and a 42% rise for rivers compared to 2006. The number of statewide mercury advisories for coastal waters increased from 12 in 2004 to 15 in 2008. These increases likely do not reflect increases in Hg deposition, but rather increases in measurements documenting the widespread nature of mercury contamination. In addition to contamination of fish, MeHg poses risks to fish-eating wildlife such as loons, mink, eagles and otter. MeHg concentrations can also be elevated in organisms that feed on aquatic insects, such as songbirds and bats, and in organisms that dwell in wetlands and upland environments such as the Bicknell's thrush – a migratory songbird that breeds in the forested mountains of the Northeast.

B. Indicators - Mercury

Indicators for Human and Ecological Health

Mercury concentrations in fish and other animals routinely exceed human and wildlife health levels. Human health indicators for mercury are based on the concentration of MeHg in fish tissue that is considered safe for the average consumer (Figure 7). The U.S. Environmental Protection Agency has recommended a human health criterion of 0.3 parts per million (ppm) in fish tissue which represents the maximum advisable concentration of MeHg in fish and shellfish that protects the average consumer among the general population. Particularly sensitive groups of people including women in child-bearing years and children under 12 years of age are advised to limit consumption to fish low in mercury. Many states have set even more stringent

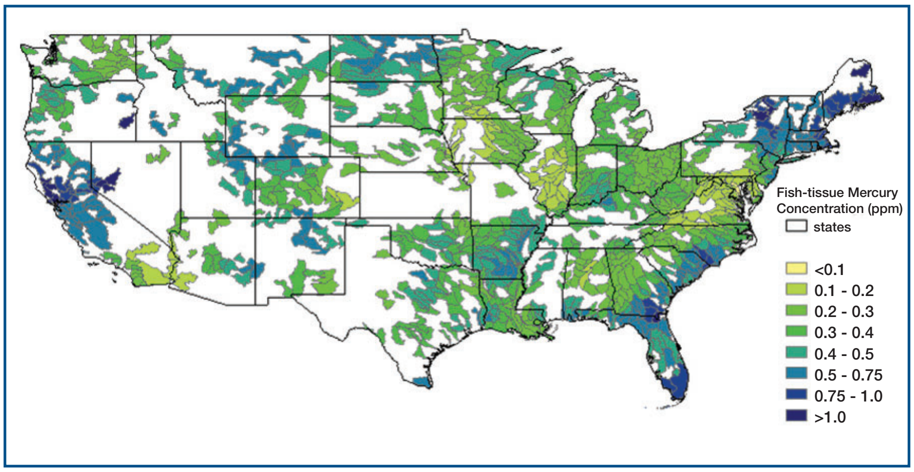

Figure 7. Fish tissue mercury concentration (ppm; same as µg/g) across the U.S. All data standardized to 14 inch largemouth bass, skin-off fillets. (Figure is derived from a model and national dataset described in: Wente, S.P., 2004. U.S. Geological Survey Scientific Investigation Report 2004-5199, 15 p.).

human health protection levels. Maine and Minnesota use 0.2 ppm as the human health threshold, as does Canada.

Ecological effects thresholds for mercury are generally based on the concentrations of mercury in the tissue, blood, or diet (often fish) of an organism that are associated with adverse impacts. Adverse impacts to biota from mercury exposure include reduced reproductive success, decreased egg incubation time and other behavioral changes, and neurological problems such as the loss of movement known as ataxia. Several ecological effects thresholds have been defined in the literature and many are lower than human health effect thresholds. For example, reproductive effects in fish-eating birds are reported at levels of 0.16 ppm in their prey fish. Significant adverse reproductive impacts on loons are commonly cited at the threshold of 3.0 ppm in loon blood.

These indicators of human and ecological effects can be used to assess and communicate risk, to determine the presence of biological mercury hotspots where concentrations exceed established thresholds, to establish targets for critical loads estimates, and to assess the effectiveness of mercury emissions reductions on target species.

Chemical Indicators of Freshwater Sensitivity to Mercury

There is large variation in the degree to which mercury deposited onto the landscape will be transported to lakes and streams through drainage waters and converted from inorganic mercury to MeHg by bacteria in soils, wetlands and lake and river sediments. The rate of methylation by these bacteria is affected by pH, sulfur and dissolved organic carbon (DOC) concentrations and, in Canadian shield lakes, was found to increase with increasing water temperatures. Once converted to the MeHg form, mercury can bioaccumulate in individual organisms and biomagnify in the food web. As a result, total mercury and MeHg concentrations in surface waters may not correlate well with mercury concentrations in biota, such as fish. In areas where mercury deposition is low or moderate, levels in fish and wildlife may be disproportionately high if conditions are conducive to MeHg production and bioaccumulation. This has been observed in some Alaskan ecosystems, such as Noatak and Gates of the Arctic National Parks, in Kejimikujik National Park of Nova Scotia, and large areas of the Northeast including the Adirondacks.

In recent years scientists have turned their attention to understanding the specific conditions that make an ecosystem sensitive to mercury. Sensitive systems may be more efficient at converting inorganic mercury to MeHg or more efficient at bioaccumulating mercury at each level in the food chain. In general, acidic ecosystems with low productivity and high sulfate and DOC tend to be sensitive to mercury inputs and to exhibit higher fish mercury concentrations. Several of these chemical indicators are influenced by inputs of acidifying deposition leading to interactive effects among two or more atmospheric pollutants. A study of freshwater ecosystems in the northeastern U.S. used the following chemical indicators of mercury sensitivity: total phosphorus, DOC, ANC, and pH (Table 4). These water chemistry indicators provide managers with a means for evaluating where MeHg concentrations in fish are likely to be high and can help prioritize monitoring and assessment efforts.

Watersheds particularly sensitive to mercury are more commonly found in the southern and eastern U.S., Great Lakes, and isolated areas in the western U.S. The sensitive regions shown in Figure 8 were identified based on the physical and chemical characteristics of a watershed that cause it to convert inorganic mercury to MeHg at a higher rate than other watersheds.

Recovery from mercury deposition has been studied in watersheds where emission controls have been implemented for large sources such as municipal waste incinerators. Studies from southern New Hampshire suggest that even though mercury is persistent and bioaccumulates in the environment, decreased inputs from local sources have been accompanied by decreased concentrations in top predators including fish and loons. In New Hampshire mercury emissions upwind of a biological mercury hotspot declined by 45% between 1997 and 2002. Mercury concentrations in yellow perch and loon blood in the region declined 32 and 64% respectively between 1999 and 2002 – rates much greater than observed elsewhere in the region. These results suggest that reduced emissions and deposition of mercury from local and regional sources are needed to restore healthy wildlife and safe fisheries and that return to levels consistent with human health criterion is likely to occur within decades, not centuries. However continued monitoring is essential in light of increasing contributions of mercury from global sources and the need to better understand potential interactions with other pollutants and with climate change.

C. Mercury and Critical Loads

Science-based air pollution thresholds and critical loads for mercury are not as well established as those of sulfur, nitrogen and acidity. Efforts are underway to develop and refine critical loads for mercury by investigating the linkage between atmospheric deposition levels, methylation processes and chemical and biological thresholds for human and ecological health. Critical loads for mercury and other trace metals have been estimated for parts of Europe using a set of very general assumptions relating atmospheric Hg to Hg concentrations in groundwater, food crops, and aquatic organisms. Available data for calculating exceedances are quite limited. However the results suggest that a large part of the landscape (approximately 50%) exceeds mercury critical loads for ecosystem effects.

Table 4. Indicators of Surface Water Sensitivity to Mercury. The following thresholds are associated with concentrations of methylmercury in yellow perch greater than 0.3 ppm in lakes in the northeastern U.S.

Indicator	Threshold
Total phosphorus	<30 μg/L
Dissolved organic carbon (DOC)	>4 mg/L
Surface water pH	<6.0
Acid neutralizing capacity (ANC)	<100 μeq/L

From: Driscoll, C.T., Han Y.-J., Chen, C.Y., Evers, D.C., Lambert, K.F., Holsen, T.M., Kamman, N.C., and Munson, R.K. 2007. Mercury contamination in forest and freshwater ecosystems in the Northeastern United States. BioScience 57: 17-28.

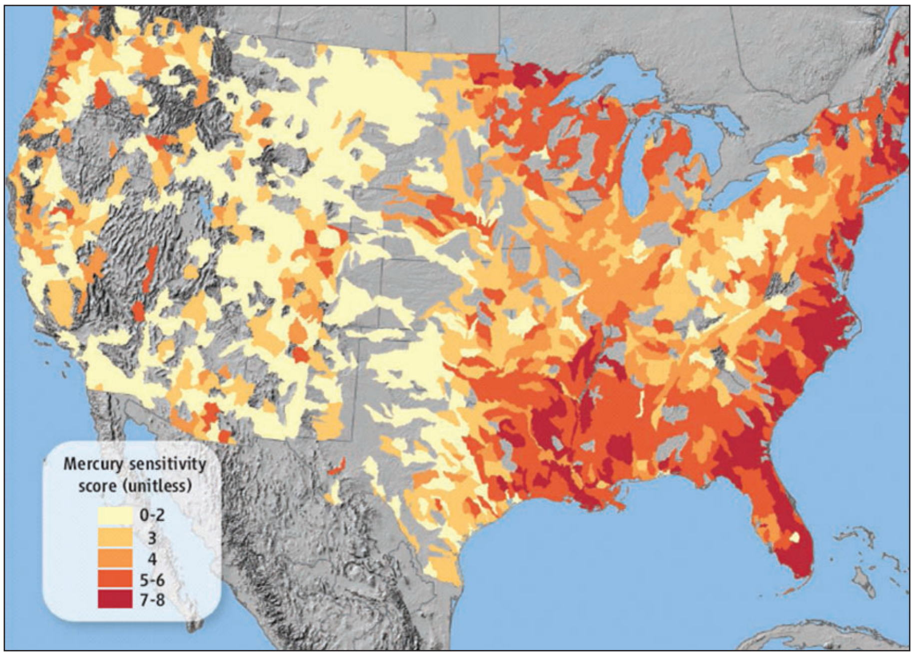

Figure 8. Nationwide mercury sensitivity map for aquatic ecosystems. The higher the number, the more sensitive the system. (From Myers et al., 2007. Science 318:200-201).

Current understanding of the links between mercury emissions and deposition and biological responses in humans and ecosystems is hampered by a lack of consistently collected long-term data on mercury levels in waters, soils, and biota. Mercury monitoring efforts vary widely among states and are difficult to integrate and synthesize to establish response patterns. In 1996 the Mercury Deposition Network (MDN) began measuring mercury deposition in precipitation (wet deposition) (http://nadp.sws.uiuc.edu/mdn/) and currently includes 115 sites in the United States and Canada as part of the National Atmospheric Deposition Program (NADP). In 2009, the Atmospheric Mercury Network (AMNet) also joined NADP and includes 21 sites that track the concentration of different forms of mercury in precipitation. There is further need for a comprehensive environmental mercury monitoring network.

USING AIR POLLUTION THRESHOLDS IN POLICY & MANAGEMENT

Ample evidence exists to advance the wider use of air pollution thresholds in policy, management and regulatory issues. Policy and regulatory decisions in response to air pollutant emissions are based on many economic, political, human health, environmental, scientific and sociological considerations and tradeoffs. Air pollution thresholds can be used to help evaluate and monitor these tradeoffs and several examples of their effective application exist in the U.S., Europe and Canada.

1. U.S. Policy Use of Thresholds

Although the 1970 U.S. Clean Air Act mandates protection of human health and welfare (which includes ecological effects), neither the Clean Air Act nor its 1990 amendments specifically mandates a critical loads approach for addressing air pollution. Class I areas are designated federal wilderness areas that were given special protection from degradation by air pollution under the Clean Air Act Amendments of 1977. It is becoming increasingly evident that critical loads for effects on terrestrial and aquatic ecosystems are exceeded in many Class I areas, even though the human health-based standards for NO_2 and SO_2 are rarely exceeded in these areas. State and federal environmental and regulatory agencies and multi-stakeholder organizations are increasingly turning to critical loads as a type of threshold that can aid in the development of air quality standards, the assessments of emissions regulations, and other policies aimed at protecting or improving ecosystem condition. In Rocky Mountain National Park in Colorado, the critical load for nitrogen deposition impacts on aquatic diatom communities provides the basis for a nitrogen deposition goal to achieve resource protection. The National Park Service, the State of Colorado Department of Public Health and Environment, the U.S. Environmental Protection Agency, and interested stakeholders collaborate in the Rocky Mountain National Park Initiative to develop strategies to reduce air pollutant emissions that contribute to nitrogen deposition in the Park.

The EPA recently started using critical loads to describe threshold effects in its annual Acid Rain Progress Reports. In the 2009 Acid Rain Progress Report, critical loads for acid deposition were calculated for over 1,300 lakes and streams in the Northeast and Mid-Appalachian Highlands regions of the eastern United States. By comparing critical loads to deposition data before and after implementation of the Acid Rain Program, it was determined that 37% of lakes and streams in those regions where atmospheric deposition was in exceedance of the critical load in the 1989-91 period were no longer receiving sulfur and nitrogen deposition loads in 2007-2009 that threatened the health of these ecosystems.

Critical loads are also presented and discussed in the recent US-Canada Progress Reports, which detail progress achieved in implementing the US-Canada Air Quality Agreement. These assessments rely on ecosystem element cycling models from which critical loads can be estimated and the effects of the Clean Air Act and other emissions reductions laws and policies can be evaluated. Long-term measurements have generally shown improvements in some surface-water indicators of acidification, such as sulfate concentrations and pH, in the Northeast U.S. over the past 30 years. Most of these waters have not recovered to pre-acidification conditions, and many remain in excess of critical load thresholds.

Dynamic ecosystem modeling can be used to simulate likely ecosystem responses in a specific year to future emissions reductions or increases. Recent modeling work simulates that future decreases in SO_2 and NO_x emissions of greater than 50% (relative to clean air laws implemented as of 2005) will be necessary to decrease the number of chronically acidic lakes in the Adirondacks by one-third to one-half by the year 2050. Despite these improvements, model results indicate that many of the currently chronically acidic lakes will improve only to an episodically acidic status, so the net change in the sum of chronic plus episodically acidic lakes is likely to stay about the same or improve slightly depending on the extent to which emissions decrease. This work shows how models can simulate whether emissions policy goals for ecosystem recovery are likely to be met within a specific time frame.

Finally, critical loads can inform the development of national air quality standards known as the “secondary standards” that are aimed at protecting environmental resources from air pollution. The Clean Air Act requires EPA to set national air quality standards for six criteria pollutants (nitrogen oxides, sulfur oxides (SO_x), particulate matter, ozone, carbon monoxide, and lead) based on primary (health-based) and secondary (welfare-based) considerations. “Welfare” includes consideration of environmental harm. The law also requires EPA to periodically review the scientific criteria upon which the standards are based.

While EPA generally reviews criteria and standards for each of the six criteria pollutants individually, EPA decided to jointly examine NO_x and SO_x compounds in a recent review of the secondary standards. In a policy assessment completed as a part of the NO_x/SO_x secondary standard review, EPA staff found that although the current secondary standards serve to protect vegetation from direct damage associated with exposures to gaseous SO_2 and NO_2, "currently available scientific evidence and assessments clearly call into question the adequacy of the current standards with regard to deposition-related effects on sensitive aquatic and terrestrial ecosystems, including acidification and nutrient enrichment" (USEPA 2011). They further conclude that "consideration should be given to establishing a new ecologically relevant multi-pollutant, multimedia standard to provide appropriate protection from deposition-related ecological effects of oxides of nitrogen and sulfur on sensitive ecosystems with a focus on protecting against adverse effects associated with acidifying deposition in sensitive aquatic ecosystems" (USEPA 2011). Finally, the policy assessment recommends the use of a critical loads approach in establishing and monitoring this suggested ecologically relevant standard.

The conclusions of the EPA staff policy document were supported in a review by the independent Clean Air Science Advisory Committee (CASAC), which advises EPA on scientific issues related to Clean Air Act implementation. Similar to the CASAC findings, the data and information presented here support the advance of secondary air quality standards to enhance recovery of sensitive ecosystems from acidifying atmospheric nitrogen and sulfur pollution. In particular, secondary standards for NO_x and SO_x could be developed using a critical loads approach to link pollution concentrations in the air with deposition and ecological effects based on established indicators for surface water chemistry and biology. In addition, the wealth of research-scale data on thresholds and critical loads becoming available for U.S. ecosystems support the exploration of other collaborative policy and management approaches to strategically reduce pollution in areas where demonstrated impacts exist. Such a process was used in Colorado, as discussed above.

2. European Policy Use of Thresholds

European scientists and policymakers have used a critical loads approach for addressing nitrogen and sulfur effects in ecosystems since 1994. The air pollution abatement strategies under the European Union Convention on Long-range Transboundary Air Pollution and under the European Commission's Thematic Strategy on Air Pollution are linked to acidity and nitrogen critical loads as the basis for negotiating national emissions maxima. Meanwhile, ecosystem monitoring has demonstrated that pollution abatement efforts have decreased acidification, and to a lesser degree, nitrogen levels in ecosystems. Once scheduled emissions controls are completely implemented, European areas that exceed acidification critical loads will be reduced from 93 million hectares in 1990 to an estimated 15 million hectares.

In Europe, application of critical loads has connected science to policy by providing methodologies for using scientific evidence to define pollution limits and to assist in setting emission control targets within a broad multination policy framework. As a result, nitrogen critical loads have been developed for NO_3^- leaching from forests and for changes to European plant species diversity for most major vegetation types found in Europe. Similarly, acidification critical loads have been developed to protect terrestrial ecosystems and thousands of lakes and streams. Critical loads for mercury, lead and cadmium have also been developed in Europe based on *endpoints* (e.g., human health and ecosystem function) and *indicators* (metal concentrations in soil, food crops, and biota).

3. Canadian Policy Use of Thresholds

Soil nutrient declines caused by acidifying deposition have resulted in extensive timber

Figure 9. Equipment used to monitor air quality at Great Smoky Mountains National Park.

productivity loss in Atlantic Canadian forests. Fish declines in lakes and rivers of eastern Canada have significantly impacted the Canadian recreational fishing industry. Concern about acidification impacts to ecosystem services (Table 1) motivated Canadian policymakers to apply critical loads when establishing regional and, later, national emissions reduction policies. Sulfur-based critical loads that initially served as the basis for emission control policy have been lowered over time as understanding of air pollution effects on forests and surface waters expanded. Canada periodically reviews its critical loads to ensure that they remain consistent with the latest scientific information and policy requirements. Since the 1990s both S and N are considered in critical load analyses, although airborne S pollutants continue to be the predominant, anthropogenic acidifying agent in Canada.

The national emission control policy agreed to in 1998 by the federal and all provincial and territorial governments is called the Canada-Wide Acid Rain Strategy for Post 2000. It seeks to meet the environmental thresholds of critical loads for acid deposition across Canada; decreasing SO_2 emissions where needed to meet critical loads, and minimizing growth in emissions of both SO_2 and NO_x where acid deposition is currently below critical loads. As of 2008, the application of this policy has decreased SO_2 emissions in Canada 63% below1980 levels. Based on critical loads projections indicating that acid rain will continue to damage sensitive ecosystems even after full implementation of current Canadian and U.S. control programs, further emission controls will be needed.

4. Global Policy Use of Thresholds

Fifty-one European countries, Canada, and the U.S. participate in the United Nations Convention on the Long-Range Transport of Air Pollutants (CLRTAP). Under this treaty, the Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone went into effect in 2003, and established more stringent emissions targets for SO_2, NO_x, NH_3, and other pollutants. Under CLRTAP, European countries compare critical loads of S and N deposition for acidification and eutrophication to estimates of current deposition as a means of evaluating the effectiveness of emissions control measures. Countries are encouraged to work together to reduce harmful impacts to surface waters, soils, and vegetation. In addition to Europe, both Canada and the United States have ratified CLRTAP based on an understanding of the global nature of air pollution transport and the need to reduce air pollution impacts.

There is currently no mechanism in place for decreasing global sources of mercury. However, the February 2009 United Nations Environment Program Governing Council, including the U.S., China, India and 138 other countries established a process to create a legally binding agreement to control global mercury pollution by 2013. Negotiations started in June 2010, and will address a broad set of issues such as decreasing mercury emissions from human sources, improving management of mercury in the waste stream and at storage sites, curbing demand for mercury in products, decreasing mercury mining, remediating contaminated sites, and enhancing global monitoring. Once the framework is established, participating nations must each ratify the treaty in order for it to go into effect. The impact of this framework will be assessed over time based in part on the extent to which fish tissue mercury concentrations have declined to below human health thresholds.

ADVANCING THE USE OF AIR POLLUTION THRESHOLDS IN POLICY

Nitrogen, sulfur, and mercury pollution are altering the Earth's ecosystems. Coupled with other stressors including land use changes, invasion of exotic species, and climate change, these pollutants are threatening the provision of ecosystem services such as air and water purification, waste detoxification, climate regulation, soil regeneration, biodiversity maintenance, and production of crops, timber, and fish. Ongoing and future projected climate change is likely to become an increasing environmental stressor in coming decades. However, little research to date has quantitatively explored the interaction of climate change with the effects of nitrogen, sulfur, and mercury pollutants in ecosystems.

In the face of large-scale global change, natural resource managers, air regulators, and public stakeholders need to know whether emissions controls are effective in producing the ecosystem benefits that were anticipated. While scientists can often determine an air pollution threshold where ecological change is likely to occur and define the nature and degree of change, decision makers must weigh a number of scientific and societal considerations in deciding which ecosystem changes are of concern and what level of protection is desired to address these concerns. Furthermore, long-term monitoring programs for tracking trends in air pollution, soil and water chemistry, and aquatic and terrestrial biota are a critical component for protecting natural resources and for the development, refinement, and application of ecological and policy thresholds.

This interaction between science and decision-making often proceeds iteratively. Once ecosystem components that should be protected (*sensitive receptors*) have been identified, scientists can identify response thresholds (e.g., *critical loads*) for those components, land managers and stakeholders can determine desired protection levels (*policy thresholds* such as *target loads*), and environmental regulators can evaluate tradeoffs to determine whether emissions controls are warranted to achieve these goals. Field studies and modeling can help by linking potential threshold responses (ecological thresholds) to stressors. Monitoring is essential to determine whether the desired response is achieved.

The National Ambient Air Quality Standards (NAAQS) for air pollutants such as NO_x and SO_x are based on concentrations of these pollutants in ambient air rather than on deposition levels experienced by ecosystems. Scientific progress has improved our ability to relate ambient air concentrations to atmospheric deposition inputs and effects through the estimation of critical loads. The secondary standards provide a framework for addressing these issues and ample evidence exists for applying existing research and modeling to the case of acidifying deposition impacts on sensitive aquatic ecosystems. Similar applications of secondary standards toward protection of terrestrial ecosystems from the effects of nitrogen and sulfur pollution would also be of great benefit. Nitrogen as ammonia and ammonium (NH_3 and NH_4^+), are increasingly important sources of nitrogen air pollution, but are not regulated by EPA as criteria pollutants in the NAAQS.

Air pollution thresholds based on science provide a mechanism for evaluating the extent to which ecosystem services have been compromised and for restoring impaired ecosystems. Establishing priorities such as the levels at which various ecosystem services should be maintained will require the mutual engagement of public stakeholders, policymakers, and scientists. Use of *ecological thresholds* for assessing the impacts of air pollution on essential ecosystem services and for informing public policy is gaining ground. These ecological thresholds provide a strong basis for development of *policy thresholds* and offer a scientifically sound approach to protecting and restoring U.S. ecosystems.

For Further Reading

Aber, J.D., C.L. Goodale, S.V. Ollinger, M.-L. Smith, A.H. Magill, M.E. Martin, R.A. Hallett, and J.L. Stoddard. 2003. Is nitrogen deposition altering the nitrogen status of northeastern forests? *BioScience*. 53: 375-389.

Burns, D.A., T. Blett, R. Haeuber, and L.H. Pardo. 2008. Critical loads as a policy tool for protecting ecosystems from the effects of air pollutants. *Frontiers in Ecology and the Environment*. 6: 156-159.

Driscoll, C.T., G.B. Lawrence, A.J. Bulger, T.J. Butler, C.S. Cronan, C. Eagar, K.F. Lambert, G.E. Likens, J.L. Stoddard, and K.C. Weathers. 2001. Acidic deposition in the northeastern United States: Sources and inputs, ecosystem effects, and management strategies. *BioScience*. 51: 180-198.

Driscoll, C.T., et al. 2007. Mercury Matters: Linking mercury science with public policy in the northeastern United States. Hubbard Brook Research Foundation. *Science Links*^{TM} Publication. Vol. 1, no. 3, Hanover, NH.

Dupont, J., T.A. Clair, C. Gagnon, D.S. Jeffries, J.S. Kahl, S.J. Nelson, and J.M. Peckenham 2005. Estimation of critical loads of acidity for lakes in northeastern United States and eastern Canada. *Environmental Monitoring and Assessment*. 109: 275–29.

Evers, D.C., Y.-J. Han, C.T. Driscoll, N.C. Kamman, W.M. Goodale, K.F. Lambert, T.M. Holsen, C.Y. Chen, T.A. Clair, T.J. Butler. 2007. Biological mercury hotspots in the northeastern United States and southeastern Canada. *BioScience*. 57: 1-15.

Fenn, M.E., J.S. Baron, E.B. Allen, H.M. Rueth, K.R. Nydick, L. Geiser, W.D. Bowman, J.O. Sickman, T. Meixner, and D.W. Johnson. 2003. Ecological effects of nitrogen deposition in the western United States. *BioScience*. 53: 404-420.

The Heinz Center. 2010. Indicators of Ecological Effects of Air Quality. The H. John Heinz III Center for Science, Economics and the Environment. Washington, D.C. Available online at: http://heinzctr.org/Programs/Reporting/Air_Quality/index.shtml

Lovett, G.M., and T.H. Tear. 2008. Threats from Above: Air Pollution Impacts on

Ecosystems and Biological Diversity in the Eastern United States. The Nature Conservancy and the Cary Institute of Ecosystem Studies.

Pardo, L.H., M.J. Robin-Abbott, and C.T. Driscoll (eds.). 2011. Assessment of Nitrogen Deposition Effects and Empirical Critical Loads of Nitrogen for Ecoregions of the United States. Gen. Tech. Rep. NRS-80. Newtown Square, PA: U.S. Department of Agriculture, Forest Service, Northern Research Station.

USEPA. 2009. 2008 Biennial National Listing of Fish Advisories. EPA 823-F-09-007. http://water.epa.gov/scitech/swguidance/fishshellfish/fishadvisories/upload/2009_09_16_fish_advisories_tech2008-2.pdf

USEPA. 2009. Acid Rain and Related Programs: 2008 Environmental Results. http://www.epa.gov/airmarkt/progress/ARP_3/ARP_2008_Environmental_Results.pdf

USEPA. 2011. Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur. EPA-452/R-11-005a. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards Health and Environmental Impacts Division, Research Triangle Park, North Carolina. 364 p. http://www.epa.gov/ttnnaaqs/standards/no2so2sec/data/20110204pamain.pdf

Acknowledgements

Funding for this project was provided by Purchase Order EP09H001132 from the US Environmental Protection Agency to the Ecological Society of America.

About the Scientists

Mark E. Fenn, USDA Forest Service, Pacific Southwest Research Station, Riverside, CA 92507
Kathleen F. Lambert, Harvard University, Harvard Forest, Petersham, MA 01366
Tamara Blett, National Park Service, Air Resources Division, Denver, CO 80225
Douglas A. Burns, U.S. Geological Survey, Troy, NY 12180
Linda H. Pardo, USDA Forest Service, Northern Research Station, Burlington, VT 05403
Gary M. Lovett, Cary Institute of Ecosystem Studies, Millbrook, NY 12545
Richard Haeuber, U.S. Environmental Protection Agency, Clean Air Markets Division, Washington, DC 20460
David C. Evers, BioDiversity Research Institute, Gorham, ME 04038
Charles T. Driscoll, Department of Civil and Environmental Engineering, Syracuse University, Syracuse, NY 13244
Dean S. Jeffries, Environment Canada, National Water Research Institute, Burlington, ON L7R4A6

Science Writing and Layout

Mark Schrope, Science writer
Bernie Taylor, Design and layout

About Issues in Ecology

Issues in Ecology uses commonly-understood language to report the consensus of a panel of scientific experts on issues related to the environment. The text for *Issues in Ecology* is reviewed for technical content by external expert reviewers, and all reports must be approved by the Editor-in-Chief before publication. This report is a publication of the Ecological Society of America. No responsibility for the view expressed by the authors in ESA publications is assumed by the editors or the publisher.

Jill S. Baron, Editor-in-Chief,
U.S. Geological Survey and Colorado State University, jill.baron@colostate.edu.

Advisory Board of Issues in Ecology

Ex-Officio Advisors

ESA Staff

Clifford S. Duke, Director of Science Programs
Jennifer Riem, Science Programs Coordinator

Additional Copies

This report and all previous Issues in Ecology are available electronically for free at

www.esa.org/issues.

Print copies may be ordered online or by contacting ESA:

Ecological Society of America
1990 M Street NW, Suite 700
Washington, DC 20036
(202) 833-8773, esahq@esa.org