Reduced Nitrogen in the Atmosphere: A Critical Review of Research and Management Needs

by Charles T. Driscoll, Jana B. Milford, Daven K. Henze, and Michael D. Bell

The 2024 Critical Review examines the topic of atmospheric reduced nitrogen: sources, transformations, effects, and management. The full-length review appears in the June 2024 issue of the *Journal of the Air & Waste Management Association (JA&WMA)*. A brief summary follows.

Emissions of reduced nitrogen are dominated by ammonia, which is the most abundant alkaline gas in the atmosphere. Current understanding of sources, effects, transport, and fate of ammonia are provided in the 2024 Critical Review,[1] as well as the status of efforts to manage ammonia emissions. In the Critical Review, research needs are identified, and policy recommendations included on how the U.S. Environmental Protection Agency (EPA) and other federal agencies could move forward in addressing ammonia emissions.

Why We Care About Ammonia

Over the past century, humans have greatly altered the global nitrogen cycle through production and application of synthetic fertilizer, widespread cultivation of leguminous crops, dramatically increased livestock production, and emissions of nitrogen oxides associated with fossil fuel combustion.[2,3] Concerns over perturbation of the nitrogen cycle and the environmental effects of emissions of ammonia have been recognized for decades.[4] While emissions of nitrogen oxides are trending downward in the United States and globally, emissions of ammonia have been and are projected to continue increasing in the United States and on the global scale[5,6] (see Figure 1). Livestock waste and synthetic fertilizer application dominate ammonia emissions sources, with motor vehicles contributing a significant share in many urban areas.[7] Ammonia emissions are largely unregulated in the United States; voluntary programs and U.S. Department of Agriculture incentives have had limited impact in mitigating emissions.

Ammonia emissions have serious effects on human health, visibility, climate, and ecosystems. Emissions of ammonia are estimated to contribute to hundreds of thousands of deaths per year globally,[8] and more than 10,000 premature deaths per year in the United States[9] due to ammonium compounds associated with fine particulate matter. Ammonium sulfate and nitrate comprise more than half of visibility-reducing haze on the most impaired days at most national parks and wilderness areas across the United States.[10]

Ammonia emissions also affect the climate system in complex ways. Nitrogen deposition generally enhances plant biomass and soil carbon storage,[11] but also increases susceptibility to wildfire.[12] Excess nitrogen deposition increases soil emissions of nitrous oxide, a potent greenhouse gas.[13] Atmospheric reactions with ammonia have decreased radiative forcing through production of reflecting inorganic aerosols and clouds,[14] but may also increase formation of absorbing organic aerosols.[15]

Nitrogen deposition has benefits for croplands and overall plant productivity, but causes widespread harm to extensively managed ecosystems. It reduces biodiversity and contributes to eutrophication of soil and fresh and coastal water bodies that can lead to harmful algal blooms and toxin production, and hypoxia.[3] Nitrogen deposition in the United States has shifted over the past two decades from mainly oxidized forms of nitrogen to largely reduced forms. Locations of highest deposition are now found in agricultural regions of the Midwest rather than the more industrialized eastern United States (see Figures 2 and 3).

Thresholds of nitrogen deposition leading to ecosystem harm, or critical loads, have been set for a variety of ecosystem components, with exceedances indicating high risk of degradation. The increasing role of reduced nitrogen in total nitrogen deposition has resulted in large areas of the United States having exceedances of critical loads for declines in lichen diversity, declines in tree growth and survival, and increases in eutrophication of lakes.[1,16] Understanding the spatial distribution of where reduced nitrogen deposition pushes an ecosystem over the critical load allows managers and policymakers to focus efforts where decreases in emissions can have the greatest impact on ecosystem health (see Figure 4). Additionally, excessive nitrogen inputs contribute to the impairment of coastal ecosystems across the United States, causing eutrophication, toxic algal blooms, hypoxia, and loss

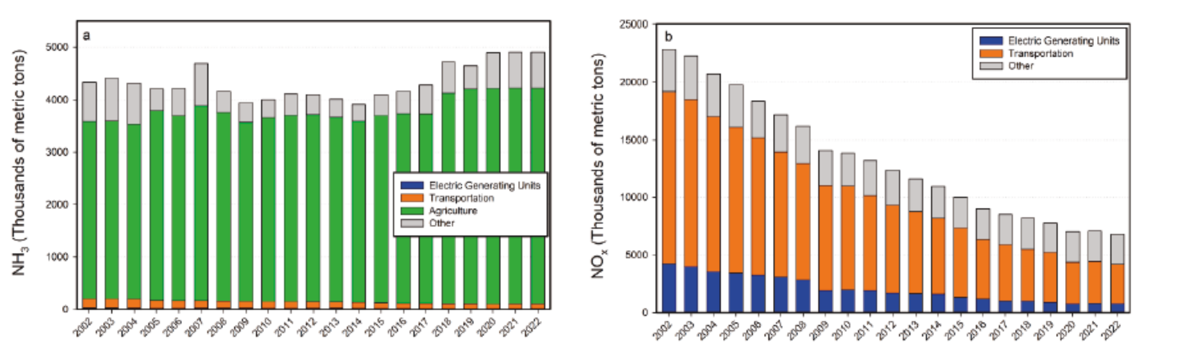


Figure 1. Time series of annual total U.S. anthropogenic emissions of (a) reduced nitrogen (NH_3) and (b) and oxidized nitrogen (NO_x).[6]

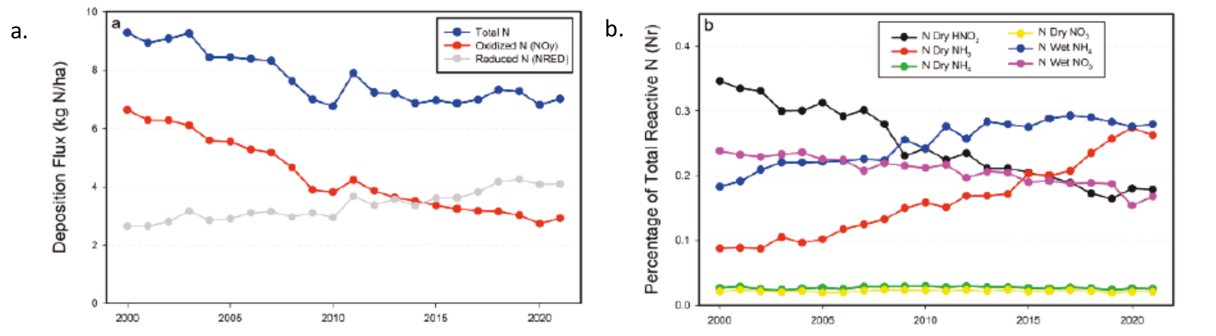


Figure 2. Time series of (a) area weighted mean annual total nitrogen and oxidized (NO_y) and reduced nitrogen (NH_x) deposition and (b) the fractional contributions of components of wet and dry N deposition to total N deposition for the conterminous United States.
Source: Data are from the National Atmospheric Deposition Program Total Deposition (TDep) Committee.

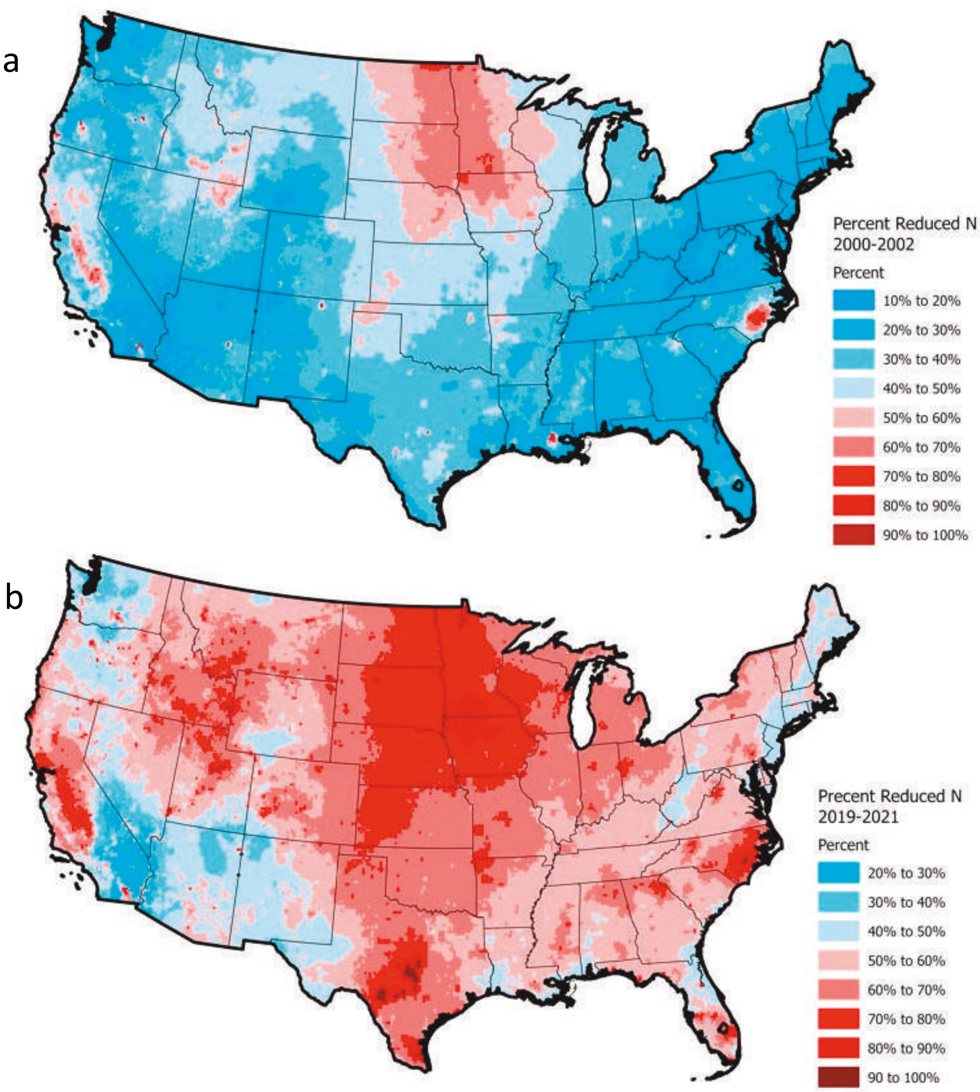


Figure 3. Maps comparing the percent of total N deposition occurring as total reduced nitrogen (NH_x) for the conterminous United States for (a) 2000–2002 with (b) 2019–2021.
Source: Data are from the National Atmospheric Deposition Program Total Deposition (TDep) Committee.

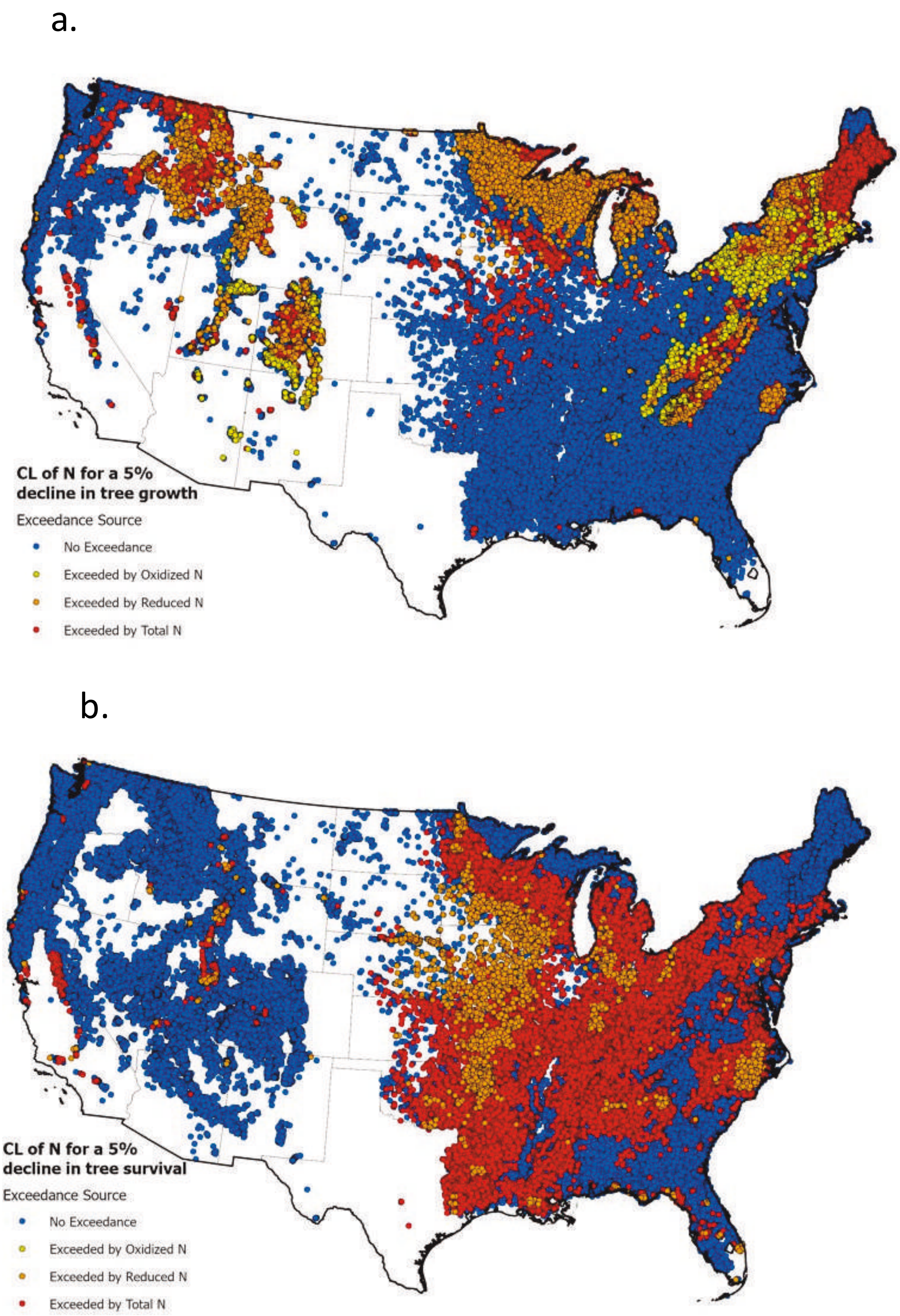


Figure 4. Map of nitrogen critical load exceedances for the conterminous United States for (a) a 5% decrease in tree growth and (b) tree survival.

Notes: Shown are in blue – no exceedance; yellow – exceedance that could be eliminated by oxidized nitrogen; brown – exceedance that could be eliminated by reduced nitrogen; and red – a combination of oxidized and reduced nitrogen is necessary to eliminate the exceedance of important habitat in severe cases.[17] To mitigate these effects, it is essential to consider and control atmospheric deposition to the watershed as well as direct deposition to the waterbody, as atmospheric deposition is a significant source of nitrogen to major U.S. bays and estuaries.

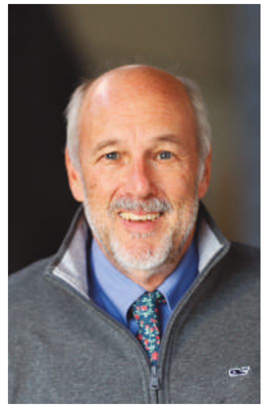
Recent research has improved understanding of the biogeochemical processing of nitrogen, including biomass and soil uptake and loss through leaching and atmospheric emissions.[18] Investigations of ecosystem recovery as nitrogen deposition is declining in some areas show significant hysteresis (lags) in recovery, possibly associated with nitrogen accumulation in woody biomass and soil organic matter.[18,19] Further research is needed to improve quantitative understanding of interactions between nitrogen deposition impacts and climate change, and of ecosystem response to decreases in nitrogen deposition and to increases in deposition of reduced nitrogen while oxidized nitrogen deposition is decreasing. Studies conducted over the past decade have elucidated factors that control spatial variation of ecosystem sensitivity to nitrogen deposition, highlighting the roles of soil pH and available base saturation as important controlling variables. Future assessments of nitrogen critical loads can be refined by accounting for mediating factors, such as changing climate, air pollution, and soil conditions to develop locality-based critical loads.

Ammonia Emissions, Transport, and Fate

Ammonia plays a well-known role in forming secondary inorganic aerosols. However, the long-standing question of whether ammonia, nitrogen oxides or sulfur dioxide emissions limit secondary inorganic aerosols formation for specific locations remains of interest. Recent research has demonstrated the importance of aerosol pH and liquid water content in governing partitioning of sulfate, nitrate, and reduced nitrogen between gas and particle phases.[20] Chemistry and transport models are beginning to explicitly simulate and evaluate performance for these aerosol properties. More research is needed on the site-specific role of ammonia in regulating the magnitude and trends of secondary inorganic aerosol concentrations and nitrogen deposition in the context of changing sulfur dioxide, nitrogen oxides, and ammonia emissions. Key factors include the role of particle pH, liquid water content, size, surface coatings, and associated kinetic limitations to constituent uptake.

Gas-phase ammonia has a short atmospheric lifetime due to the parallel sinks of conversion to condensed ammonium or direct deposition of ammonia.[21] In contrast, condensed-phase ammonium has a longer lifetime and potential for long-range transport. Modeling studies indicate that deposition of reduced nitrogen in the United States is dominated by U.S. emissions, but interstate transport within the United States is significant.[22] As sulfur dioxide and nitrogen oxides emissions are decreasing, the fraction of reduced nitrogen in

About the Lead Author



Dr. Charles T. Driscoll is a Professor of Environmental Systems and Distinguished Professor with the Department of Civil and Environmental Engineering at Syracuse University in Syracuse, NY. He earned a bachelor's of science degree (with distinction) in Civil Engineering from the University of Maine in 1974; a master's in Environmental Engineering from Cornell University in 1976; and a Ph.D. in Environmental Engineering from Cornell University in 1980.

Dr. Driscoll received the Athalie Richardson Irvine Clarke Prize for Outstanding Achievement in Water Science and Technology in 2023; Syracuse University Chancellor's Lifetime Achievement Award in 2020; and the Presidential Young Investigator Award in 1984. He was the lead author of the United Nations Environmental Programme, Intergovernmental Panel on Biodiversity and Ecosystem Services report on Land Degradation and Restoration Assessment (2016–2018). He is a Fellow of the American Association for the Advancement of Science (2018–present); a member of the National Academy of Engineering (2007–present); and a noted Highly Cited Researcher for Engineering and Environmental Science with the Institute of Scientific Information (2003–present).

Dr. Driscoll was a member of the National Academies Committee on Assessing Causality from a Multidisciplinary Evidence Base for National Ambient Air Quality Standards (2021–2022); a member of the National Academies Committee on Independent Scientific Review of Everglades Restoration (2006–present; Chair 2019–present); a member of the International Conference on Mercury as a Global Pollutant, Scientific Advisory Committee (2013–present); a member of the National Research Council Board of Environmental Studies and Toxicology (2011–2017); a member of the National Research Council Committee of Air Quality Management (2001–2004); a member of the U.S. Environmental Protection Agency Committee Reviewing National-Scale Mercury Assessment Supporting the Appropriate and Necessary Finding for Coal and Oil-fired Electric Generating Units (2011); a member of the Clean Air Scientific Advisory Committee Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur (2007–2011, 2014–2018, 2023); and U.S. Representative to United Nations/European Economic Community Workshop of Critical Loads of Sulfur and Nitrogen (1988).

Dr. Driscoll's current research addresses the effects of disturbance on freshwater, forest, urban and marine ecosystems, and recovery and mitigation from disturbance, including air pollution (acid and mercury deposition), land-use, and climate change. Current research focuses on: recovery of eastern forest watersheds and surface waters from acidic deposition; ecosystem restoration; ecosystem response to changing climate; mitigation of harmful algal blooms; atmospheric deposition, watershed and surface water transport and transformations, and biotic exposure of mercury; analysis of costs and benefits associated decarbonization of the electricity sector. The Driscoll laboratory has published more than 525 articles in peer-reviewed journals. These works have been cited over 54,000 times, with an H-index of 117 (Google Scholar).

the gas phase is increasing, causing a shift toward more local deposition and less long-range transport (Figures 2, 3). Continued investigation of the spatial footprint of sources contributing to deposition at sensitive locations is needed as the chemical and climatic environment shifts. Dry deposition of ammonia gas is rarely measured but is estimated from air quality models for purposes of national assessments, so values are relatively uncertain. Further efforts are needed to evaluate ammonia dry deposition estimates from air quality models used for research and regulatory applications.

Inverse modeling studies continue to show limitations in traditional bottom-up ammonia inventories, with a general bias toward underestimation.[23,24,25] Further research is needed to reconcile bottom-up and top-down estimates and to improve mechanistic understanding of ammonia emissions (e.g., from bidirectional exchange), as well as estimates of the magnitude of emissions from specific sectors such as livestock production and motor vehicles. The increasing availability of satellite observations holds promise for greatly expanding atmospheric ammonia measurements, helping track trends, improve emissions estimates, and characterizing deposition.[26] However, satellites do not directly observe surface concentrations, emissions, or deposition, so chemistry and transport models, data assimilation and inverse modeling are critical tools for deriving relevant products. In addition, harmonization of multiple remote sensing records is needed to better understand long-term trends in ambient ammonia concentrations and to identify sources and sinks. Continued research is needed to advance top-down emissions estimation approaches using inverse modeling as more satellite data become available, including instruments on new and potentially upcoming polar and geostationary satellite platforms.

While expanding satellite observations and products are promising, it is vitally important to maintain surface monitoring networks that support research and policy assessments.[27] Surface monitoring networks provide critical data for ground-truthing and constraining remote sensing analyses, evaluating chemistry and transport models, assessing human and ecosystem exposure, and tracking long-term trends. Maintaining support for established U.S. monitoring networks has been a perennial challenge, and multiple sites have been suspended or terminated in recent years.

Managing Ammonia Emissions

To date, EPA has declined to list ammonia as a criteria pollutant or to set emissions limits for industrial, agricultural, or mobile sources.[1,28] EPA has delayed regulating ammonia emissions from animal feeding operations for almost 20 years after signing consent agreements with operators in the mid-2000s.[29] Emissions uncertainties, heterogeneous impacts, administrative complications, and a preference for voluntary approaches for the agricultural sector have been

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Invited Discussants

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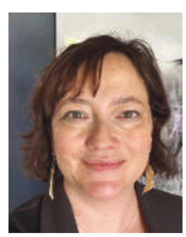

Dr. Carla Davidson is President, Endeavour Scientific Inc. in Calgary and works with Indigenous communities in the Athabasca Oil Sands on issues of environmental management and monitoring.

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cited as reasons not to regulate ammonia emissions. EPA should revisit these decisions, considering improved scientific understanding and the ongoing and multi-pronged harms caused by reduced nitrogen.

EPA has traditionally set secondary standards for protection of human and ecosystem welfare based on ambient concentrations. As recommended by EPA's Clean Air Scientific Advisory Committee, the agency should reconsider this approach and add deposition-based standards for ecosystem effects.[27] Critical loads for lichens, sensitive tree species, and freshwater eutrophication risks provide a sound basis for establishing such standards. Total maximum daily loads for contribution of atmospheric deposition to estuarine watersheds and coastal waters should also be considered.

Confined animal feeding operations contribute disproportionately to U.S. ammonia emissions, with broad regional impacts, as well as heightened health and welfare harms for nearby communities. In 2017, when it denied a petition to set performance standards for confined animal feeding operations, EPA asserted the need to evaluate emissions from confined animal feeding operations and determine further regulatory actions to decrease emissions and cited limited resources and competing priorities.[30] As many health and environmental protection organizations have urged,[31] EPA should reconsider this decision and move forward to set standards. Recommended best management practices, state and local regulations that address ammonia and other emissions from animal feeding operations, and updated European rules for this source sector provide models for feasible work practices that can be used to decrease feedlot emissions.

The European Union has relatively comprehensive regulations to address ammonia emissions through countrywide emissions caps that are guided by critical loads and ambient air quality standards, as well as source-specific emissionslimits and work practice requirements.[32] In contrast to the United States, where ammonia emissions are increasing, emissions across European Union countries declined by 13% from 2005 to 2021.[33] The United States and other impacted countries should follow the lead of European nations in developing an integrated response to the suite of health and environmental harms caused by reduced nitrogen.

As prior assessments have also noted (e.g.,[34]), the sources and natural resources affected by ammonia emissions cross over traditional bureaucratic lines of research and management responsibility in the United States, including air and water divisions at EPA, U.S.D.A., federal land and coastal resource managers, and state and tribal agencies. Europe's ongoing nitrogen assessments and the U.S. National Acid Deposition Assessment Program provide successful examples of the cross-cutting, integrated approach that is needed to fill information gaps and design, implement, and assess mitigation strategies for reduced nitrogen.

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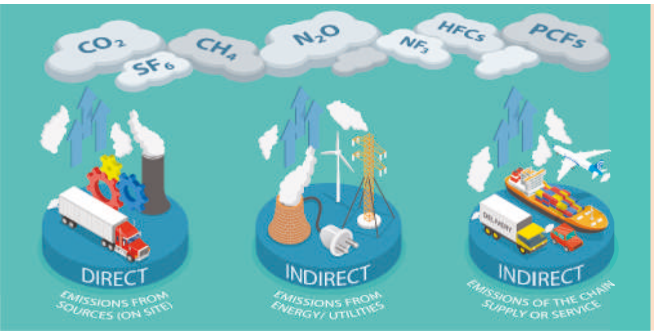

In Next Month's Issue…
Emissions Inventories

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Reduced Nitrogen in the Atmosphere: A Critical Review of Research and Management Needs

Presented Dr. Charles T. Driscoll

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Comments also will be solicited from the floor and from written submissions to the Critical Review Committee Chair. The Chair will then synthesize these points into a Discussion Paper that will be published in the October 2024 issue of *JA&WMA*. Comments should be submitted in writing to Susan S. Wierman, Critical Review Committee Chair, at **susan.wierman@gmail.com** by no later than July 25, 2024.

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Get involved with the Critical Review Committee and help further our scientific understanding by attending the Annual Meeting of the Critical Review Committee on Thursday, June 27, at 10:30 a.m., immediately after the presentation. Please refer to the Conference Program for room allocation.

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About the Critical Review

For more than 50 years, A&WMA has solicited and published in the *Journal of the Air & Waste Management Association (JA&WMA)* an Annual Critical Review on a topic of critical importance to the air and waste management fields. Each year, the review author presents the Annual Critical Review during A&WMA's Annual Conference & Exhibition. The Critical Review Committee, which is a subcommittee of the Publications Committee, selects the review topics, solicits the authors/presenters, offers editorial guidance and critiques to the review authors, reviews the final manuscript before publication, and selects the participants for the panel discussion that follows the review presentation.