Integrated Assessment of Ecosystem Effects of Atmospheric Deposition

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Ecosystems obtain a portion of their nutrients from the atmosphere. Following the Industrial Revolution, however, human activities have accelerated biogeochemical cycles, greatly enhancing the transport of substances among the atmosphere, water, soil, and living things. The atmosphere is an important pathway for local, regional and global scale transport, and atmospheric deposition is an important process by which substances are removed from the atmosphere, by wet deposition, dry deposition, and/or cloud or fog deposition processes. Atmospheric deposition includes beneficial nutrients, inert materials, and substances which are toxic depending upon their concentration or the sensitivity of the organisms or ecosystems exposed. Since humans have altered the chemical climate of the Earth, it is essential that we understand the sources, transport, transformations, and effects of airborne substances on the health and productivity of the ecosystems on which the quality of life depends.

Our goal in this article is to discuss how inter-disciplinary, multi-disciplinary, and trans-disciplinary research and assessment programs have helped inform managers on the effects of atmospheric deposition on ecosystems and how this understanding has been used to guide air quality management programs in Europe and North America.

Atmospheric Deposition and its Effects on Ecosystems

Acidification and eutrophication are important mechanisms by which airborne substances alter the physical structure, and biological composition and productivity of both aquatic and terrestrial ecosystems, thereby affecting the services these ecosystems provide. The principal air emissions contributing to acidification and eutrophication of ecosystems are sulfur dioxide (SO_2) and nitrogen oxides (NO_x), largely from combustion sources, and ammonia and organic nitrogen, largely from agricultural sources. Note, agricultural emissions of total reactive nitrogen in the U.S. in 2002 were about three times larger (18.6 Tg/y) than reactive nitrogen emissions from both transportation sources (3.8 Tg/y) and electric utilities (1.9 Tg/y).[1] However, NO_x control programs have been focused primarily on electric utility and transportation sectors. Reactive nitrogen includes all biologically, chemically, and radiatively active nitrogen compounds in the atmosphere or biosphere. In this article, we use the terms "acid rain," "acid deposition," "acidifying deposition," and "nutrient enrichment" to refer to atmospheric deposition induced acidification and eutrophication of terrestrial and aquatic ecosystems.

Scientific recognition of acid deposition extends back to the 1850s.[2] However, only since the 1960s has acid deposition been recognized to involve long-range transport (i.e., hundreds of km) and subsequent deposition of air pollutants, documenting the need for research, measurements and models by atmospheric scientists.[3] A key role in scientifically documenting and politically highlighting the "acid rain" problem was played by Svante Odén, who in 1967 published an insightful and provocative article in the Swedish newspaper *Dagens Nyheter*. His conclusions on the consequences of acid deposition were largely based on the international network on atmospheric deposition (The European Air Chemistry Network) set up in the mid-1950s to better understand atmospheric circulation and the input of elements, in particular nutrients.[4]

Early limnologists and aquatic scientists also discovered acidic conditions in remote surface waters far removed from emission sources and observed associated impacts on aquatic organisms, further demonstrating the linkages from emissions of air pollutants to long-range transport to atmospheric deposition to ecological effects. Interest and concern over the ecological effects of acid deposition accelerated in the 1960s and 1970s with studies first in Europe and later in North America (Figure 1).[4, 5]

Early studies on ecosystem effects during the 1960-70s focused on sulfur compounds that were then believed to be the major cause of acidifying deposition. Research on the impacts of nitrate, ammonium, and organic nitrogen deposition was relegated to secondary status. During the mid-1980s, field studies reported unexpectedly marked leaching of nitrate from remote forest watersheds.[6] This observation lead to the nitrogen saturation hypothesis and the concept of the nitrogen cascade, which motivated a new line of research examining the acidification and nutrient enrichment effects of all chemical forms of reactive nitrogen.[7, 8] Research has shown that recent increases in deposition of reactive nitrogen affect the structure and function of remote forests, alpine and grassland ecosystems, coastal estuaries, and even open-ocean ecosystems.

In addition to cross-disciplinary research and synthesis, implementation of multi-faceted research programs has led to integrated understanding and management of ecosystem effects of atmospheric deposition. These approaches include long-term measurements, experimental manipulations, synoptic surveys, and both conceptual and mathematical modeling.[9] A series of experimental whole ecosystem manipulations have been effective in demonstrating cause-and-effect relationships between the chemical perturbations associated with atmospheric deposition and ecological effects .[10,11] Synoptic-scale surveys have enabled researchers and managers to quantify the spatial extent of the impacts of atmospheric deposition. Researchers who had the foresight to initiate long-term measurements that have continued today have allowed natural resource scientists and managers to characterize and quantify how atmospheric deposition and ecosystems respond to changes in emissions and document the effectiveness of emission control programs.[12, 13] Finally, a series of atmospheric transport models (e.g., ADOM, RADM, CMAQ AURAMS, EMEP) were developed to improve scientific understanding of processes affecting the transport, transformations and deposition of air contaminants, and watershed acidification and nutrient retention models (e.g., PnET, MAGIC, SPARROW) have been developed to quantify the effects and recovery of impacted ecosystems in response to emission control programs.

HISTORICAL SURVEY OF PROGRESS IN EUROPE

Beginning in 1970, the Air Management Group within the Organization for Economic Cooperation and Development (OECD) established a 5-year project to monitor atmospheric concentrations and deposition, and evaluate the transboundary flux of sulfur over Western Europe. Building on the early observations by Oden and others, Sweden presented the first comprehensive assessment of the acid deposition problem at the UN Conference on the Human Environment in Stockholm in 1972.[14] In 1977, the OECD monitoring project concluded that "sulphur compounds do travel long distances (several hundred kilometers and more) in the atmosphere" and that "air quality in one European country is measurably affected by emissions from other European countries".[15] Thus, long-range transport and deposition of sulfur and resulting acidification became an issue of international concern within Europe. As a result, the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) was established as a pan-European monitoring and research program. This program became a standard for regional air pollution measurements and modeling, and most countries in Europe established their own national networks that are linked to EMEP (http://www.emep.int/).

In 1985, the Working Group on Effects under the Convention on Long-Range Transboundary Air Pollution (CLRTAP) set up a series of International Cooperative Programmes (ICPs) to monitor various ecological effects of transboundary air pollution.[16] These programs included: ICP Waters, ICP Forests, ICP Integrated Monitoring, and ICP Modeling and Mapping. The last of these ICP programs was directed to develop emissions inventories and critical loads and levels. Critical loads are the amounts of deposited air pollutants below which adverse effects on specified sensitive ecosystem components are not observed, according to present knowledge. Critical loads provide direct information to managers on the magnitude of decreases in emissions that would lead to the decreases in deposition needed to protect ecosystems.

National Research Programs within Europe

After acid precipitation became an international issue, many countries in Europe established national research programs to understand and evaluate the effects on ecosystems. These observational and experimental research programs often were designed to provide both increased understanding of underlying processes and also to provide the scientific foundation for emission control programs. Through these studies, many countries became aware of their local acidification problems, and a strong scientific community was established throughout Europe.

The first and one of the most comprehensive research programs in support of policy on acid rain was the Norwegian research program "Acid Precipitation: Effects on

Forests and Fish" (SNSF) between 1972 and 1980. SNSF studies integrated the effects of acid deposition on both aquatic and terrestrial systems. In Sweden, an experimental study was established at Lake Gårdsjön watershed in 1978, which has become one of the longest research programs on integrated effects of acidification in Europe. In addition to long-term measurements, the Lake Gårdsjön project includes a series of experimental manipulations to illustrate effects of accelerated acidification, recovery, and mitigation. Other countries also established long-term research programs directed at understanding the effects of acid deposition including Finland, France, the United Kingdom, The Netherlands, Czechoslovakia, and Germany.

Comparative studies were also set-up across Europe in order to learn more about the nature of acidification and ecosystem response under different deposition regimes. These experiments included the Surface Water Acidification Project (SWAP) supported by the British coal and power industry, but conducted through the Royal Society in London and the scientific academies in Norway and Sweden. Even before completion of this project, the British power industry announced plans to install flue gas desulfurization equipment on some of its facilities. NITREX was was an experimental program supported by the European Commission to determine the role of nitrogen in acidification and the dynamic behavior of nitrogen leaching under different deposition scenarios. NITREX forest ecosystems experiencing elevated nitrogen deposition sufficient to cause leaching exhibited an immediate and marked response to the removal of nitrogen inputs in roof exclusion experiments.[17] In contrast ecosystems with limited leaching of nitrogen showed a delayed leaching response to experimental nitrogen additions even after several years of treatment. One of the most important aspects of these highly visible experiments was that they served as effective platforms to clearly demonstrate and communicate to stakeholders and policymakers the effects of acid deposition on ecosystems and the potential for recovery following controls on emissions.

Critical loads as a management tool to assess and mitigate adverse ecological effects

The concept of critical loads was developed largely through a common understanding between scientists and policymakers at two workshops; a Nordic workshop in 1986 and a United Nations Economic Commission for Europe (UNECE) workshop in Skokloster, Sweden in 1988. In response to the success of these workshops, critical loads became an important element in the revision of the NO_x Protocol in December 1988.

The first methods for deriving critical loads were simple and largely based on (semi)empirical data. The calculation and mapping of critical loads was advanced through Integrated Assessment Models, which were first introduced during the preparation of the second Sulfur Protocol. Through application of the unified EMEP model, atmospheric source-receptor relationships were established and steady-state models such as the Steady-State Water Chemistry Model were used to established critical loads. Moreover, models became important tools to develop cost-effective emission control strategies in protocol negotiations. Critical loads are readily adopted as a common basis for policy development because they are continually revised and updated with improvements in scientific understanding. They also form an effective bridge allowing for communication and interaction between scientists and policymakers.

Development of the multiple-pollutant and multiple-effects approaches

In Europe, a series of actions took place starting with protocols for individual pollutants, such as the Sulfur Protocol and the NO_x Protocol. Only after understanding from research had advanced did Europe develop the multi-pollutant Gothenburg Protocol. The use of integrated assessment models to develop critical loads, quantify multiple effects and determine cost-effective emission control strategies was key to this undertaking. “Stove piping” occurred in early assessments, but eventually Europe moved toward a better integration of multiple pollutants affecting ecosystems. This approach became the basis for the Gothenburg Protocol which set national emissions targets for multiple pollutants; the fundamental air pollution control in Europe since 2000. The multiple-pollutant approach was also the basis for the European Union National Emissions Ceilings (NEC) Directive approved in 2001.

Experiments and models based on experiments have demonstrated long-term dynamics associated both with acidification, eutrophication, and recovery of ecosystems. The observed time lag in recovery processes is an important consideration in the development of future control strategies, and has lead to the use of dynamics models to determine the time required for ecosystems to reach so-called dynamic critical loads.

HISTORICAL SURVEY OF PROGRESS IN NORTH AMERICA

In contrast to Europe’s and Canada’s early concern about the acidification and eutrophication impacts of air pollution on ecosystems resulting from long-range transport from major pollution sources, air quality management in the U.S. was initially dominated by concerns about more local sources of emissions and resulting impacts on human health. The Clean Air Act (CAA) of 1970 established “National Ambient Air Quality

Standards" that were focused on limiting concentrations of individual air pollutants in urban areas rather than deposition to more distant rural and remote areas. The so-called "Secondary Standards intended to protect Public Welfare" (including terrestrial and aquatic ecosystems) were generally set equivalent to the "Primary standards" intended to protect public health.

Similar to Europe, early investigations of acid rain in North America were largely *ad hoc* with little national coordination. The Canadian Network for Sampling Precipitation (CANSAP) began in 1976 and the National Atmospheric Deposition Program (NADP) began in the U.S. in 1978. Both networks provide detailed maps of spatial and temporal trends in the chemistry of precipitation. Data from these networks provided important motivation and a scientific foundation for the 1990 CAA in the U.S. and the U.S.-Canada Air Quality Agreement on Transboundary Air Pollution in 1991.

The Canadian program on Long-Range Transport of Air Pollutants (LRTAP-Canada) began during the late 1970s. LRTAP, involved complementary programs in different provinces of Canada, with particularly compelling and visible whole lake acidification studies in Ontario.[10]

Coordinated research on acid deposition in the U.S. began in 1980 under the National Acid Precipitation and Assessment Program (NAPAP). NAPAP was a 10-year multi-agency, multi-disciplinary program of policy-focused research. It culminated with a series of state of science and technology reports that included emissions; atmospheric processes and deposition; aquatic processes and effects; terrestrial, materials, public health and visibility effects; and integrated assessment of both emissions control technologies, future emissions scenarios, and consequent ecosystem-recovery projections.

Nearly coincident with the conclusion of NAPAP, the U.S. Congress passed the 1990 CAA. This path-breaking legislation included plans for a financial markets-based, “cap and trade” program for management of SO_2. This program involved a 50% decrease in emissions of SO_2 from 1980 levels by 2010 and additional controls for NO_x emissions. The extent of emission controls was based on assessments of what was economically acceptable and the belief that some degree of ecosystem recovery would occur. Title IV of the 1990 CAA has proven to be a cost-effective approach to achieve substantial decreases in emissions of SO_2 and NO_x oxides from power plants and industrial boilers, with the targeted “cap” for SO_2 emissions attained by 2007 three years ahead of schedule.[18]

The 1990 CAA and programs that followed had the foresight to include activities that were needed for assessment. Continuous emission monitor (CEM) systems were implemented to track SO_2 emissions to ensure that control targets were met and provide transparency for the cap and trade program. The NADP/National Trends Network (http://nadp.sws.uiuc.edu/) together with the U.S. Environmental Protection Agency (EPA) Clean Air Status and Trends Network (http://www.epa.gov/castnet/; approximately 85 sites to monitor atmospheric chemistry and estimate dry deposition) were used to assess changes in atmospheric deposition in response to emission control programs. The U.S. EPA, the U.S. Geological Survey, the National Park Service (NPS) and state agencies also supported surface water programs as part of the U.S. EPA Office of Air and Radiation: (1) Long-Term Monitoring Program (LTM), and (2) Temporally Integrated Monitoring of Ecosystems (TIME). Together these two monitoring programs provide important information on changes in water chemistry in the eastern U.S. in response to changes in the deposition of air-borne pollutants.[13] Unfortunately, biological measurements were not included as part of these 1990 CAA assessment programs. Monitoring programs provided critical information to determine whether the rate and extent of chemical indicators of ecosystem recovery was occurring as originally forecast with models under NAPAP for the 1990 CAA. These surface water monitoring programs, together with studies showing ongoing soil acidification demonstrated that ecosystem recovery from elevated atmospheric deposition has been delayed.[19]

Meanwhile, models continued to be improved as science and understanding advanced. More advanced biogeochemical models were used to help understand why watersheds and surface waters were not recovering as fast as anticipated. These models have also been paired with new atmospheric models. This effort addressed whether additional emission controls, largely driven by human health concerns, that go beyond Title IV acid rain requirements would be enough: the answer was maybe, but probably not. Further emission controls would be required to accelerate ecosystem recovery.

The U.S. EPA introduced the Nitrogen Budget Trading Program (NBP) to implement the NO_x State Implementation Plan Call in 1998 and the Clean Air Interstate Rule (CAIR) in 2005. The NBP focused on controlling NO_x emissions to decrease inter-state transport of ozone and its precursors in the eastern U.S. CAIR mandated additional decreases in emissions of SO_2 and NO_x from utilities across the eastern U.S. This rule was designed to address regional transport of ozone and $PM_{2.5}$ (particulate matter less than 2.5 μm in diameter) with a cap-and-trade program for both SO_2 and NO_x emissions. Although CAIR was vacated by the U.S. District Court in Washington D.C. in July 2008, this same CAIR rule was reinstated by the Appeals Court of the District of Columbia in

December of that same year. An enduring effect of CAIR has been to encourage the U.S.EPA to continue its progress toward using a multi-pollutant/multiple effects approach in air quality management as was recommended by the National Research Council (NRC) in the report "Air Quality Management in the United States".[20]

Following the recommendations of the NRC, the NPS, USDA Forest Service, and U.S. EPA initiated critical loads pilot projects. These projects have been designed to evaluate the critical loads approach and build experience in using critical loads to prevent significant deterioration in Class I wilderness areas and to help guide air quality management to facilitate recovery of ecosystems that have been impacted by air pollution. North American research and management communities have largely used steady-state biogeochemical watershed models and empirical studies to determine critical loads for sensitive ecosystems. In addition, researchers have documented the impacts of atmospheric nitrogen deposition on terrestrial and aquatic ecosystems in the West.[21] So air pollution effects on ecosystems are no longer a regional issue of the eastern U.S., but clearly a national problem.

FUTURE CONSIDERATIONS

For the first time in its history, the U.S. EPA is conducting an integrated review of secondary National Ambient Air Quality Standards for the combined control of two criteria pollutants -- NO_x and SO_2.[22] This multi-pollutant effort builds on recent multi-disciplinary research which has developed an integrated understanding of the combined effects of NO_x and SO_2 on ecosystems, and uses the concept of ecosystem services to inform decisions about adverse effects on public welfare. Ecosystem services are those outputs of ecological functions or processes that contribute to social welfare or have the potential to do so in the future. Some outputs may be bought and sold, but most are not marketed. In this analysis, the U.S. EPA is also considering the effects of atmospheric deposition of chemically-reduced nitrogen on ecosystems. Although emissions of chemically-reduced forms of nitrogen are not currently regulated in the U.S., the contribution of these pollutants to total nitrogen deposition is quantified to determine the effectiveness of a combined NO_x-SO_2 standard. Finally, critical loads are proposed as a component of the approach for the secondary standard. These shifts in policy suggest that the different paths of air quality research and management taken in Europe/Canada and the U.S. through the 1990s and early 2000s may be coming together in the future.

In Europe, there are presently two parallel policy initiatives. First, after the National Emissions Ceilings (NEC) directive was approved, the European Commission extended its air quality management in 2005 to develop a thematic strategy for air pollution. This strategy considered both health and ecosystem effects, and the proposal for policy measures was directed towards air quality standards, source control legislation and emission ceilings based on integrated assessment models. The revision of the NEC directive is still (as of May 2010) under discussion within the European Commission. Second, the Gothenburg Protocol has been re-assessed and the Protocol will be likely be revised in 2011. With the Gothenburg Protocol, Europe has moved from a focus largely on ecosystem protection to a focus on both human health and ecosystems. Thus, Europe has come closer to the U.S policy driver, while the U.S. has been moving toward the original European policy driver. Both are trying to balance the demands for human health and ecosystem protection, recognizing the need for cost-effective management strategies.

There are several critical research needs, as North American and European efforts to understand, quantify and manage the effects of atmospheric deposition continue. There is an ongoing need for comprehensive monitoring data. These data should include continuous integrated measurements of air chemistry, atmospheric deposition, and both soils and surface water chemistry in regions that are sensitive and have undergone acidification and eutrophication by atmospheric deposition. Such integrated data sets are essential to track the effectiveness of air quality management programs, to test atmospheric transport and biogeochemical watershed models, and to validate critical load calculations (i.e., accountability). In addition to these chemical monitoring programs, there is a need to maintain and develop associated biological monitoring programs to evaluate the impacts of air pollutants on biologic resources, and to quantify their rates of recovery in response to atmospheric emission control programs.

To date, critical load calculations in the U.S. have largely relied on a steady-state modeling approach. Steady-state models have the advantage that they are relatively simple and have limited data requirements. However, ecosystems are dynamic rather than at steady-state. Hence, there is a need to advance the application of dynamic models to improve the calculation of critical loads, and to evaluate the assumptions invoked in steady-state models. Although dynamic models are more complex with greater data requirements, they can be used to assess the time required to obtain a certain environmental quality condition. Such information is essential in environmental management decisions. Another important consideration in future air quality management is a more rigorous and formal understanding of the linkages between atmospheric transport and deposition models and watershed effects models. Increasingly, air pollution is recognized as a multi-media (atmosphere-land-aquatic) disturbance. To date, the applications of atmospheric transport and deposition models have been separate from watershed effects models in air quality assessments. If these tools are to be used effectively in the future, it will be necessary to examine the linkages and the compatibility of these two modeling approaches.

Future research and management should expand the multi-pollutant nature of air pollutant effects on ecosystems, and evaluation of the costs and benefits of these effects on ecosystem services and function. Better quantification of atmospheric deposition of chemically reduced and organic forms of nitrogen and their biological effects in both aquatic and terrestrial ecosystems is needed. Another important and interconnected air pollutant is mercury. The linkages of mercury and sulfur are well established as both are important pollutants from coal-combustion; the methylation of mercury by sulfate reducing bacteria; and the enhanced trophic transfer of mercury associated with surface water acidification. Finally, changing climate can profoundly alter the hydrology and biogeochemistry of ecosystems, and their response to atmospheric deposition. As a result, future research and management of air pollution effects on ecosystems will need to involve: 1) expanded multi-pollutant and multiple ecosystem effects perspectives (i.e., chemically oxidized, reduced, and organic forms of nitrogen, sulfur, mercury, phosphorus, carbon); and 2) quantifying monetized and non-monetized impacts and benefits to ecosystem services.

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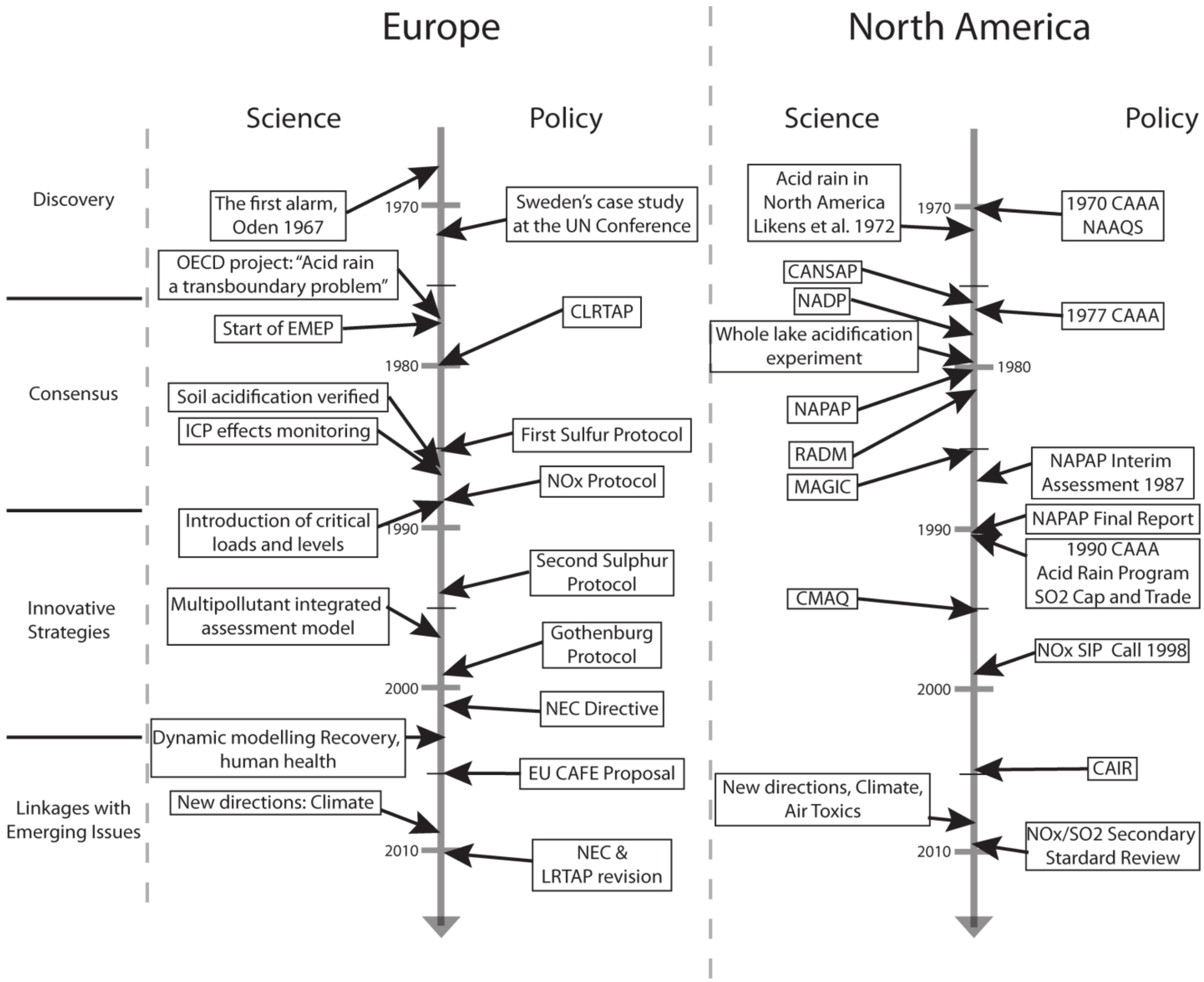

Figure 1. Chronology of major scientific events and policy development programs leading to increased scientific understanding and more effective management of atmospheric-deposition induced acidification and eutrophication of aquatic and terrestrial ecosystem in Europe and North America. Note that there have been four phases in the process: discovery, consensus, innovative strategies and linkages with emerging issues.