Clean Air Status and Trends Network (CASTNET)
Identifying Approaches for Optimizing CASTNET Monitoring

Workshop Summary
August 2009
Research Triangle Park, NC

Introduction to the Workshop

As an air quality monitoring network operated by the federal agencies, CASTNET periodically reviews the state of the network, evaluates the ongoing monitoring and assessment needs of EPA and NPS, and solicits recommendations from within the agencies and from stakeholder groups to optimize the network. A 2-day workshop was organized and held in August 2009 in Research Triangle Park, North Carolina to review the state of the CASTNET monitoring network and provide recommendations to the EPA and NPS that would ensure the network continues to provide relevant and useful information to stakeholders. Approximately 70 scientists and interested parties who are familiar with and use CASTNET measurements participated in the workshop. The operating agencies were particularly interested in recommendations on improvements to the current instrumentation and methods that would address contemporary and projected assessment needs. Because agency budgets have been decreasing in real terms when adjusted for inflation, one of the overall goals of this workshop was to provide recommendations that, once implemented, would allow the program to continue to operate within the current budget constraints. Thus, the costs of any additions or improvements were required to be balanced with savings or improved efficiency of the network.

Organization of the Workshop

The workshop was organized into distinct sections. First, presentations were made that provided background on the network, descriptions of current monitoring technologies, and anticipated agency monitoring needs, including those for assessment and numerical model development and evaluation. These presentations can be found in Appendices A, B, and C. Following the introductory presentations, two major themes were explored in sessions entitled, *Strengthening the Foundation*, and *Building on the Foundation*. Participants in both sessions broke up into smaller groups of 10-12 participants and discussed scenarios for improving CASTNET, while preserving the elements deemed necessary for meeting ongoing EPA and NPS goals. Recommendations from the individual breakout groups were vetted in the plenary sessions.

Strengthening the Foundation

In this session, workshop participants discussed the current CASTNET monitoring technology and focused on how to optimize CASTNET for achieving its contemporary and projected monitoring goals. This theme was extensively discussed and resulted in a number of specific recommendations. Break-out session leaders were provided with a spreadsheet that listed approximate costs of existing program elements, as well as available new instrumental configurations that might be considered. The groups were urged to balance new costs with savings and efficiencies. Discussion in the session focused on providing more accurate and representative data, especially for concentrations and deposition of reactive nitrogen species. The consensus of participants was that these data would be especially valuable in supporting model development and source/receptor evaluations.

Building on the Foundation

In this session, workshop participants discussed new scientific and assessment fields of study and communities that CASTNET could collaborate and engage with in the future. Discussion in the session focused on how the CASTNET program and its infrastructure could be leveraged to broaden its constituency and enhance these areas outside the traditional foci of the program. Two areas that received particular attention were ways in which CASTNET could better contribute to ecological impact evaluations and climate-change research and monitoring.

Evaluating impacts of pollutant deposition in a watershed, alpine/subalpine/forest environment, or other ecosystem requires quantifying the atmospheric inputs of acid-forming chemicals, base cations, and nutrients, including that from dry deposition flux. Many of the ecosystems most sensitive to atmospheric inputs have heterogeneous terrain, making these areal estimates of loading problematic for both wet and dry deposition. Neither the CASTNET measurement system, nor the Multi-Layer Model (MLM) used to make dry deposition estimates from CASTNET data, are designed to address this complexity. Furthermore, many of the sensitive ecosystems where impact evaluations are most needed do not have a CASTNET site nearby. Further development is needed on numerical models that address these complexities and can extend the estimates of dry deposition from CASTNET measurement sites. Specific monitoring needs for model development included accurate partitioning of gaseous and aerosol species of oxidized and reduced nitrogen, and providing data on the size-dependence of base cation concentrations. The latter data is necessary to improve the modeling of deposition fluxes of base cations as a function of particle size.

Collaboration and collocation of instrumentation with monitoring programs whose specific objectives relate to climate-change were cited as ways to increase the relevancy of CASTNET to climate-change research and monitoring. AmeriFlux and the National Ecological Observatory Network (NEON) were two networks that were identified which could significantly improve the linkage between atmospheric and biogeochemical systems. AmeriFlux is a multi-agency program with a goal of quantifying carbon, water, and energy fluxes over major vegetation types and across seasons and years. There are more than 30 sites in the continental United States.
A principal goal of this program is to improve understanding of the biogeochemical cycling of greenhouse gases and AmeriFlux would benefit from the flux measurements of nitrogen, sulfur, and other pollutants performed by CASTNET. NEON is envisioned as a long-term monitoring program that will collect data on the impacts of climate change on natural resources and biodiversity. Communications among scientists in these programs could provide linkages between historical measurements performed by CASTNET and the future ecological monitoring performed by NEON.

Recommendations

The following recommendations represent a consensus of the body of participants and summarize the discussion that led to the recommendations.

(1) *Adopt a two-tiered approach to monitoring in which most sites follow a low-cost protocol, similar to that currently conducted, while a few sites make enhanced measurements of additional species and at higher time resolution.*

Adding higher time-resolution measurements (hourly and/or daily) of many different chemical species would increase the utility of CASTNET measurements. However, there was wide recognition that such measurements are quite expensive and could not be made at all current sites in the network within the current budget. There was also wide recognition that maintaining a significant number and density of sites was essential for CASTNET to achieve its assessment objectives. The consensus of the workshop was that the network should balance these disparate needs and adopt a tiered approach to monitoring, where some specific sites are classified as "high tier", while the majority of sites would be "low tier". At the "high tier" sites, additional chemical species would be measured and additional high-time resolution measurements would be made. At the "low tier" sites, the current CASTNET monitoring protocol would continue. Specific recommendations about particular measurements to be made at the "high" and "low" tier sites are presented in additional recommendations.

(2) *Add measurements of gaseous NH_3 to the suite of routine CASTNET measurements at all sites, and continuous hourly true NO_2, NO_y, and reactive nitrogen (N_r) measurements at selected sites.*

There is wide recognition of the importance of NH_3 deposition in terrestrial and aquatic systems in rural and remote areas (e.g., Rocky Mountain National Park) and near agricultural sources (e.g., the Chesapeake Bay watershed). A recent National Research Council report calls on programs, including CASTNET, to develop the capacity to monitor NH_3, as well as all N_r species. Gaseous NH_3 is chemically labile, deposits rapidly, especially to wet surfaces, and can be deposited to or emitted from vegetation and other surfaces. This latter characteristic is termed bi-directionality and reflects the fact that atmospheric and surface concentrations of NH_3 are in quasi-equilibrium: when atmospheric concentrations are below equilibrium, NH_3 is emitted to the atmosphere, and when atmospheric concentrations are above equilibrium, NH_3 is deposited to the surface. This behavior significantly complicates the use of air concentration data to estimate dry deposition. Quantifying NH_3 deposition may require direct deposition measurements, such as eddy accumulation. Notwithstanding these complications, the consensus opinion supported making routine air concentration measurements of $NH_{3.}$ Two alternatives were discussed: (1) add a phosphorous (or citric) acid-impregnated filter to the filter pack, or (2) deploy a network of passive samplers. Both alternatives require exploratory studies to evaluate sensitivity, accuracy, and repeatability of the measurements. Passive samplers offer the possibility of making a large number of low-technology measurements in rough terrain where power is limited and accessibility is difficult.

In the tiered monitoring approach described in (1) above, the "high tier" sites could be equipped with chemiluminescent analyzers adapted to make continuous concentration measurements of NO_2, NO_y, and N_r. These measurements would support diagnostic model evaluations of fine particle formation. Further, the concentration data could be used to estimate the dry deposition fluxes of the individual nitrogen species. Several participants recommended taking daily organic nitrogen measurements in wet deposition samples at a subset of sites.

(3) Discontinue meteorological measurements at "low tier" sites with more than five years of data.

Currently, dry deposition velocities are computed by the MLM from on-site meteorological measurements and surface characteristics. In the absence of meteorological measurements, an alternative is to derive the deposition velocities from the accumulated record of MLM-computed values at a site. Another alternative is to use the output from a meteorological model as input to the MLM in place of measurements. Where the CASTNET site is in a forest clearing, this approach may yield deposition velocities more representative of the top of the forest canopy than the floor of the clearing. Unlike the meteorological measurements in the clearing, the model output is unaffected by the trees. Improvements are being explored in the way surface vegetation is characterized, as well. Using a mosaic-approach to characterize the vegetation in a grid cell may be more realistic than assuming the vegetation in a cell is uniform. Eliminating meteorological measurements would result in considerable savings.

(4) Replace wind speed and direction instruments with 3-dimensional sonic anemometers at "high tier" sites.

Currently, wind speed and direction are measured with mechanical anemometers and wind vanes. Turbulence is estimated based on the variance of wind direction. The consensus of participants was that 3-dimensional sonic anemometers would provide direct measurements of these variables that are more accurate than current instruments and also provide turbulence vectors in three dimensions (i.e. σ_u, σ_v, and σ_w). These added measurements would be valuable in numeric modeling of dry deposition and model diagnostics.

(5) Pursue development of multi-pollutant instruments that measure both gases and aerosols.

CASTNET should continue to develop and evaluate multi-pollutant instruments and, if successful, install at least three of these instruments at "high-tier" sites. Continuous (or semi-continuous) multi-pollutant measurements at selected sites would be a significant new source of monitoring data for the development and verification of deposition models. These measurements would also be invaluable in improving models for the partitioning of gas and aerosol nitrogen species.

The Monitor for AeRosols and Gases (MARGA) and modified SuperSASS instruments were discussed as potential instruments for deployment. In the MARGA, a wet rotating denuder removes soluble gases from the airstream for measurement by ion chromatography (IC). Aerosols enter a supersaturated chamber, where droplets form on and dissolve the particles. The droplets are captured and analyzed by IC. Airborne gas and aerosol concentrations are derived from the IC concentrations. The MARGA has the capacity to produce semi-continuous measurements of important gases: HCl, HNO_2, HNO_3, SO_2, and NH_3, and aerosols: Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+}. Systems such as the MARGA have the capacity to measure most of the chemical species for assessing atmospheric fluxes of acids, bases, and nutrients. Near real-time concentrations of these constituents could support diagnostic modeling evaluations, source-receptor studies, and deposition flux calculations, among others. These data could also be used to evaluate the potential biases in the CASTNET filter pack system.

The potential for the near real-time multi-pollutant chemical speciation of both gases and particles, offered by the MARGA, requires intricate processing of the airstream coupled with wet-chemical IC measurements. Given this complexity, questions were raised about the MARGA's reliability and ease of operation by a typical CASTNET operator under year-round field conditions. Citing several intensive bench and field tests, workshop participants reported cases of reliable, accurate operations, as well as cases of problems both of reliability and accuracy, especially for reduced nitrogen species. During the workshop, one participant demonstrated MARGA output by remotely accessing real-time data from an instrument operating successfully under field conditions. In summary, there was more discussion of the pros and cons of the recommendation to pursue the multi-pollutant potential of the MARGA than for any other workshop recommendation, with the conclusion that additional testing, development, and evaluation might prove it ready for routine operation at selected sites.

Super SASS samplers combine denuders with filters in various configurations that potentially could be modified to effectively separate gaseous and particulate species, such as HNO_3, and NO_3^- or NH_3 and NH_4^+. The samplers could be configured to collect daily samples at three sites for one year, and then moved to three different sites until covering the range of meteorological and air chemistry conditions encountered in CASTNET. The modified Super SASS measurements would be used to quantify the biases in filter pack measurements of gaseous and particulate nitrogen species over the one-week CASTNET sampling period. The Super SASS samplers could potentially be configured to measure organic and elemental carbon, which could support visibility and climate-related studies. Data from the samplers could also be used for model verification. Like the MARGA system, extensive testing is needed to determine the performance of the modified Super SASS.

Sampling inlets on the Super SASS sampler could be fitted with a cyclone to eliminate particles with an aerodynamic diameter larger than 2.5 μm. There was considerable discussion about the pros of cons of defining this or some other cut point. The CASTNET filter pack system has a somewhat variable cut point (~4 – 5 μm), dependent on wind speed, so there were arguments to evaluate the potential sampling biases in the CASTNET system by configuring the Super SASS sampler as nearly like the CASTNET system as practical. Other arguments were to operate the sampler with a defined cut point so that the coarse-particle effects on the CASTNET system could be quantified. The MLM computes a single deposition velocity for each chemical species, which for aerosols is a fine-particle deposition velocity. The effect of size cut on particulate chemistry measurements needs to be investigated to resolve this issue.

(6) Pursue development of methods for direct dry deposition measurements and collaborations for conducting flux measurements at CASTNET sites.

A clear need was expressed for verifying the inferential dry deposition estimates at CASTNET sites with a variety of land surfaces and vegetation types and conditions. A mobile system was suggested. Also discussed was a modified MARGA system with two analytical benches for measurement of atmospheric gradients (referred to as a GREGOR system), though like the MARGA system extensive evaluations are needed to demonstrate its performance. Recognizing that direct dry deposition measurements, whether by eddy correlation, eddy accumulation, or gradient-measurement techniques, all require considerable expertise, it was suggested that CASTNET sponsors pursue a partnership with the AmeriFlux program. Site operators have experience in high-resolution, fast-response measurements, which may be helpful in evaluating the MARGA and GREGOR systems. The collaboration was viewed as mutually beneficial to the two programs, as CASTNET can offer AmeriFlux scientists information on nutrient inputs and pollutant deposition on vegetation, and AmeriFlux can offer CASTNET scientists expertise and experience in high resolution flux measurements.

(7) *Reduce the number of CASTNET sites to 50-70 by eliminating sites in the Northeast, where the network is most dense.*

Maps of pollutant (e.g., SO_2 or sulfate) concentrations are an effective way to display air pollution data and to visually demonstrate concentration changes over time (see Figure 2). They are an invaluable tool in EPA's assessment process. It is essential that closure of northeastern U.S. CASTNET sites not jeopardize the spatial coverage of maps in that region. Closing sites must be done with attention to the spatial statistics of each concentration field. Every effort should be made to minimize the loss of "signal." In addition, sites located in ecologically sensitive areas should be maintained. A map should be produced that is based on a combination of measurements and model output. Techniques for doing this are not currently available and need to be explored. It is likely that future assessments will be increasingly model-based, yet these models will require substantial further development and validation. The nearly 4-year delay in releasing emissions data is another constraint on timely model-based assessments.

An alternative to closing sites is to collect a one-week sample every other week or even every third week, which may be adequate for producing annual concentration maps. This approach is preferable to lengthening the sampling period, which risks larger biases in the partitioning of gaseous and particulate nitrates, loses of ammonium, and saturation of the filters.

(8) *Participating agencies and stakeholders should advocate for increased resources within their agencies.*

CASTNET data continue to be a valuable resource for scientific and regulatory assessment. There is a long-term need for the network. This workshop has illustrated both its importance and a vision for its future. Its budgetary allocation does not reflect its contributions to atmospheric science, model development and evaluation, and environmental program assessment. The network is currently and chronically underfunded. As laid out in this document, there are very clear additional monitoring needs expressed by the community. The workshop participants did not approve of the "zero sum" flat budget approach for CASTNET (where additional monitoring objectives had to be paired with cutbacks in other areas). The needs expressed by the community represented at the workshop cannot be met by simply reducing the number of sites or by removing particular measurements (e.g., meteorology) to free resources. The cost-saving recommendations should be implemented as soon as possible (once the technical, scientific, and contractual implications have been evaluated) in order to safeguard the current viability of the network. However, there must be a concerted effort by EPA and NPS to work with management to take a strong position on the steps recommended by the workshop community, to demonstrate the utility of the network to upper management, and to advocate for increased resources.

(9) *CASTNET should better collaborate with other monitoring networks and convene regular meetings in order to facilitate the exchange of information and to build a defined user-community.*

This workshop was the first time in 10 years that the CASTNET community has met and participants found it to be quite valuable. Other networks have regular meetings (e.g., NADP has a forum twice per year). Regular meetings/workshops have the effect of building an integrated scientific community and leads to increased exchange of information. CASTNET should work to continue to engage with the community by meeting regularly. CASTNET should meet regularly with other monitoring networks (e.g., IMPROVE, NADP, CSN, AmeriFlux, NEON, etc.) and strive to collocate measurements where possible.

Specific mention was also made to strengthen collaborations with the EPA-sponsored Temporally Integrated Monitoring of Ecosystems and Long-Term Monitoring (TIME/LTM) programs and with programs in other agencies that use and need atmospheric deposition data for their ecosystem effects assessments.

(10) Measurement of CO and sponsorship of NCore sites should be a lesser priority for CASTNET at this time.

Carbon monoxide (CO) is a conservative tracer of combustion sources and could be used in diagnostic evaluations of chemical transformations downwind of these sources. CO concentrations are most useful in tracking plumes from large area (e.g., urban) sources or power plants. However, the consensus of workshop participants was that measuring CO at the mostly rural and remote CASTNET sites would have limited benefit without adding a suite of related tracer measurements (e.g., trace metals).

The National Core multi-pollutant monitoring program is designed to determine compliance with the NAAQS. NCore sites are designed to better characterize the numerous chemical and physical interactions between pollutants than the measurement system at most compliance-oriented monitoring sites. Most sites are or will be located in urban areas. The consensus of workshop participants was that sponsoring the full suite of NCore measurements at CASTNET sites would have limited utility in meeting the objectives of CASTNET and was not an effective use of limited resources.

Appendix A: CASTNET Overview

CASTNET is a long-term program for monitoring the principal air pollutants involved in acidic deposition and evaluating the trends in these pollutants resulting from emissions reductions programs. Established under the Clean Air Act Amendments (CAAA) of 1990, CASTNET is administered and operated by the Environmental Protection Agency (EPA) Clean Air Markets Division (CAMD). Through an interagency agreement, the National Park Service (NPS) sponsors 27 CASTNET sites located in National Parks and other Class - I areas (Figure 1). CASTNET operates more than 80 regional sites for measuring air pollutant concentrations and meteorological conditions and for recording observations needed to compute dry deposition fluxes. Sites are located in areas where urban influences are minimal.

The practice of consistent quality-assured methods for more than 15 years has resulted in a CASTNET data set that is fundamental in evaluating the effectiveness of sulfur dioxide (SO_2) and nitrogen oxide (NO_x) emissions reductions required by the Acid Rain Program (ARP), NO_x Budget Trading Program (NBP), and other programs. CASTNET data have demonstrated clearly that these reductions have resulted in cleaner air and in lower deposition of acids and acidifying chemicals. CASTNET air quality measurements and dry deposition estimates complement the wet deposition measurements made by the National Atmospheric Deposition Program/National Trends Network (NADP/NTN). Together, these data sets can serve as the basis to gauge planned and future emissions reduction programs, especially providing input for the development and evaluation of numerical models that serve as tools for regulatory assessment and for understanding atmospheric processes.

CASTNET was established to evaluate the status and effectiveness of SO_2 and NO_x emissions reductions under the 1990 CAAA. Requirements were to:

- Monitor the status and trends in regional air quality and atmospheric deposition;
- Provide atmospheric data on the dry deposition component of acidic deposition, ground-level ozone, and other pollutant gases and particles that enter the environment; and
- Assess and report the geographic patterns and long-term temporal trends in ambient air pollutant concentrations and acidic deposition.

CASTNET measurements provide data for validating and improving atmospheric models used to assess the effect of air pollutant emissions on air quality and atmospheric deposition. These models address the complex physical and chemical interactions that affect air quality, visibility, and deposition and offer insights into measurement limitations and needs. CASTNET measurements also provide information used to study the impacts of air pollution on aquatic and terrestrial ecosystems.

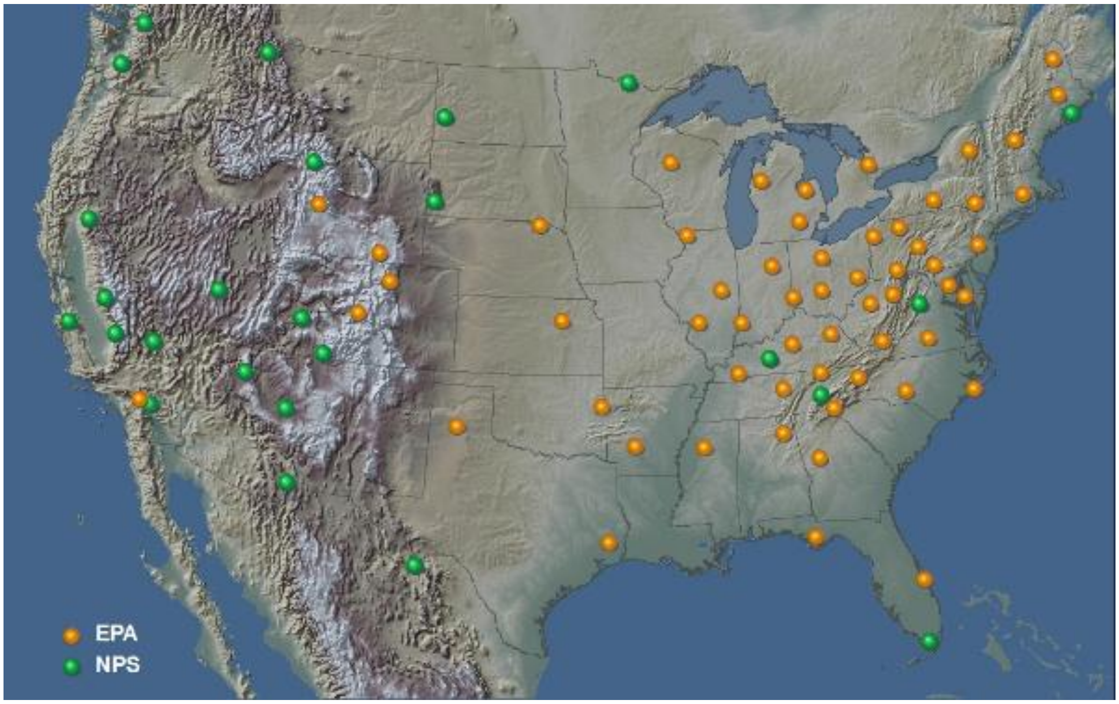

Figure 1. Locations of CASTNET sites sponsored by the EPA and NPS. For complete information on CASTNET sites see www.epa.gov/castnet.

More than half of the CASTNET sites now have at least a 20-year data record. Air quality and deposition data are strongly affected by weather conditions, so isolating long-term changes from weather-induced variations requires consistent long-term data. The duration and consistency of CASTNET data has facilitated the evaluation of spatial and temporal trends that have been used to assess the impact of emissions changes on air quality and deposition. Perhaps the foremost current and future value of CASTNET is its long-term quality-assured data set, which makes it possible to deduce the effects of emissions reductions under the ARP, NBP, and other programs affecting air quality, now and in the future. The network complements urban and source-related monitoring with rural and regional ozone, sulfur, and nitrate monitoring.

Confronted with decreasing inflation-adjusted resources, CASTNET has grown increasingly cost-effective and valuable. It is important that CASTNET continue to provide the data needed for defensible assessments of our nation's air quality programs. Figure 2 displays an example of how CASTNET data have been used to demonstrate that SO_2 emissions reductions, resulting from the ARP, have resulted in decreases in particulate sulfate concentrations.

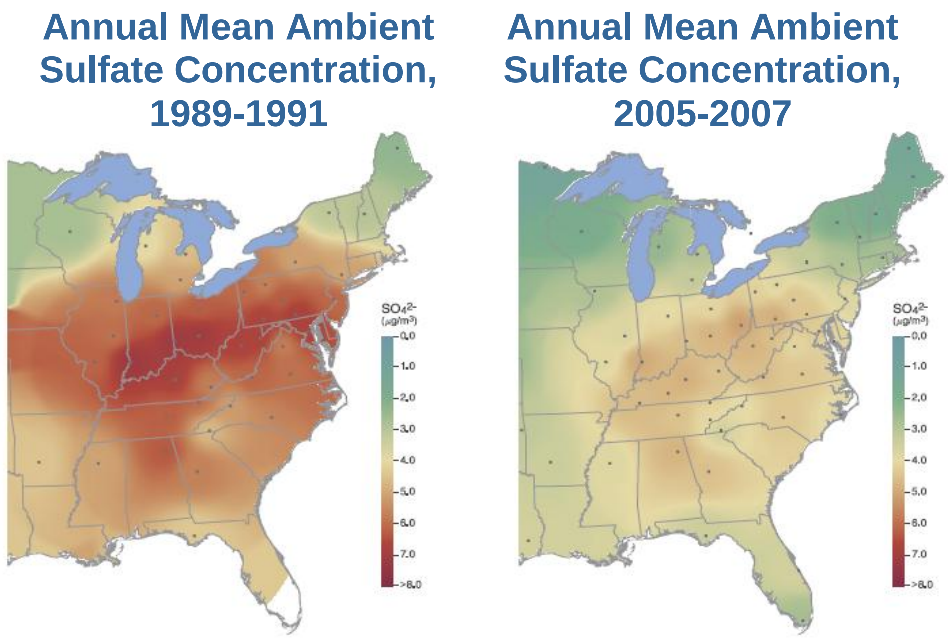

Figure 2. Three-year average sulfate concentrations calculated from one-week measurements at eastern U.S. CASTNET sites. The ARP required phased reductions of sulfur dioxide emissions beginning in 1990. For more information see www.epa.gov/castnet.

Appendix B: Description of Measurements.

Each CASTNET site reports the same set of measurements and related observations every Tuesday. A temperature-controlled shelter at each site houses a computer, data logger, and continuous ozone (O_3) monitor. Ambient particulate matter and select gases are captured in an open-face 3-stage filter pack which samples air at a mass-flow-controlled rate (1.5 liters/min. for eastern sites and 3.0 liters/min. for western sites). Filter packs are fixed atop a 10-meter tower and are replaced every Tuesday. The accumulated mass is used to calculate weekly-average air concentrations. A second 10-meter tower holds meteorological instruments for measuring temperature, delta temperature (2- and 9- meter temperature difference), relative humidity, wind speed and direction, solar radiation, and surface wetness. Wind direction data are used to compute the standard deviation of the horizontal wind direction (i.e., sigma theta). A precipitation gauge is used to record precipitation depth and time of occurrence. Regular calibration and periodic audits of instrument performance ensure that reported measurements meet the CASTNET data quality objectives.

Pollutant concentrations reported by CASTNET include:

- hourly average surface O_3;
- weekly average particulate sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), chloride (Cl^-), sodium (Na^+), magnesium (Mg^{2+}), and calcium (Ca^{2+}); and
- weekly average gaseous nitric acid (HNO_3) and sulfur dioxide (SO_2).

Figure 3 is a schematic drawing of the 3-stage filter pack, showing the filter media and particles and gases captured in each of the stages. SO_2 captured by the nylon and Whatman filters is combined to determine the total gaseous SO_2 concentration.

Dry deposition fluxes are calculated as the product of the measured pollutant concentrations and modeled deposition velocities for all pollutants, except smog-forming O_3. Hourly deposition velocities (V_d) for each pollutant are computed using the Multi-Layer Model (MLM), which derives V_d values from the hourly average meteorological measurements taken at each site. Additional MLM input includes land use and vegetative status surrounding the site. The MLM accounts for the atmospheric and surface resistances that affect the rate at which each pollutant is deposited. Daily dry deposition amounts are calculated as the sum of the product of the hourly fluxes.

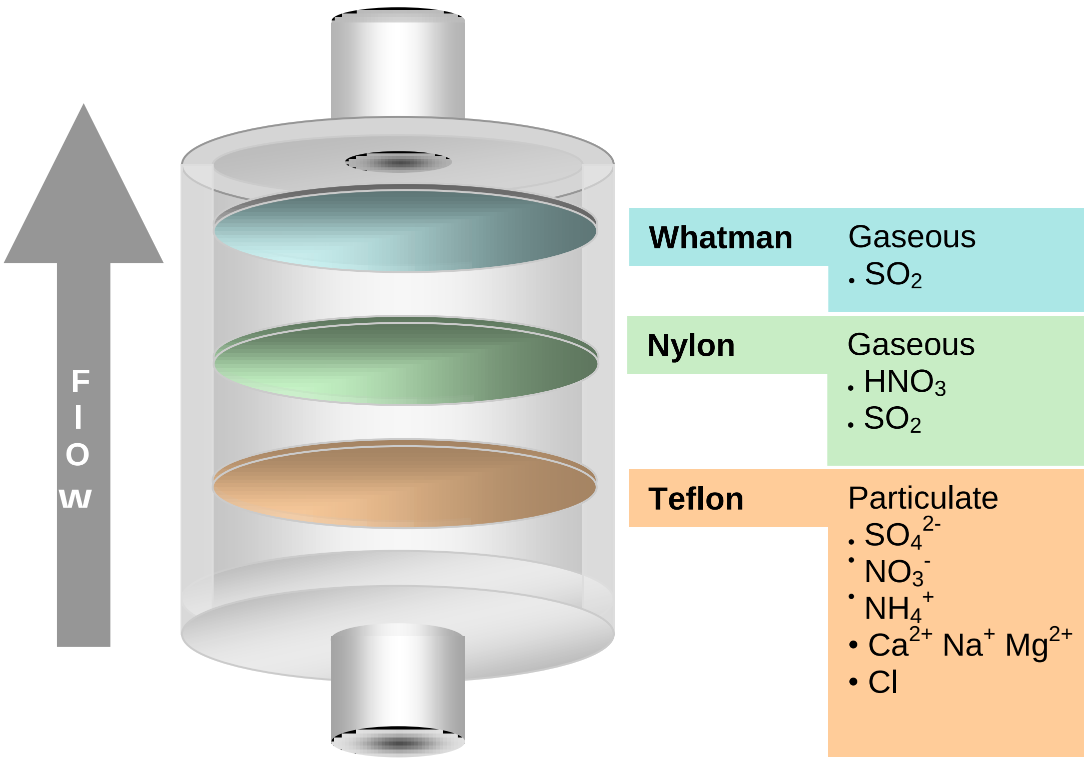

Figure 3. Schematic diagram of the 3-stage filter pack. SO_2 is captured on both the nylon and Whatman cellulose filters. The Whatman filters are impregnated with potassium carbonate.

Appendix C: Summary of Agency Presentations.

Critique. Among the strengths of the CASTNET measurement program is its consistency of quality-assured measurements over 20-plus years at many locations. This constancy facilitates the robust comparison of pollutant emissions trends with spatial and temporal trends in air quality and has led to efficient, cost-effective operations. However, several issues have been identified that should be addressed. These are listed and then described below.

- CASTNET does not measure several species that are important to characterizing total nitrogen inputs to ecosystems, the largest of which is ammonia.
- Characterizing deposition in the West is difficult because there are fewer sites and most western sites are located in heterogeneous terrain that does not comply with the assumptions of the MLM model.
- The MLM model also provides point estimates of deposition that cannot be reliably extrapolated to surrounding areas.
- There are no direct deposition measurements made at CASTNET sites to help validate modeled estimates by the MLM.

Growing evidence points to the need for improved speciation of nitrogenous compounds, especially as they affect aerosol formation and growth and as nitrogen deposition affects the nutrient balance in unmanaged ecosystems. For example, predictions from the Community Multiscale Air Quality (CMAQ) model suggest that up to a fourth of the reactive nitrogen deposited in the Chesapeake Bay watershed goes unmeasured, because it occurs as nitrogen species not currently detected by the CASTNET or NADP measurement programs (see Figure 4). Accurate accounting of all of the important nitrogen inputs in the Chesapeake watershed are needed in order to form comprehensive strategies for mitigating the seasonal eutrophication in this estuarine system. Mounting evidence of the deleterious effects of nitrogen deposition in sensitive alpine and sub-alpine ecosystems in the western United States requires adaptations that will improve the identification and accurate measurement of atmospheric nitrogen in both wet and dry deposition.

The program has not taken advantage of advances in measurement technologies and instrumentation that could improve accuracy, expand coverage, and leverage new applications for the data. Sulfur (SO_4^{2-} and SO_2) measurements are robust and stable and have largely met the needs of the ARP. However, particulate NH_4NO_3 is much less stable and is affected by ambient temperature, relative humidity, and gas-phase NH_3 and HNO_3 concentrations. Changes in these over the week-long sampling period can lead to conditions favoring volatilization of NH_4NO_3 from the first stage of the filter pack, resulting in potentially significant negative biases in particulate NO_3^- and NH_4^+ concentrations and accompanying positive biases in gaseous HNO_3, which is captured on the second stage of the filter pack. Dry deposition estimates are disproportionately affected by these biases, since the deposition velocity of particulate NH_4NO_3 is approximately an order of magnitude smaller than the deposition velocity of HNO_3.

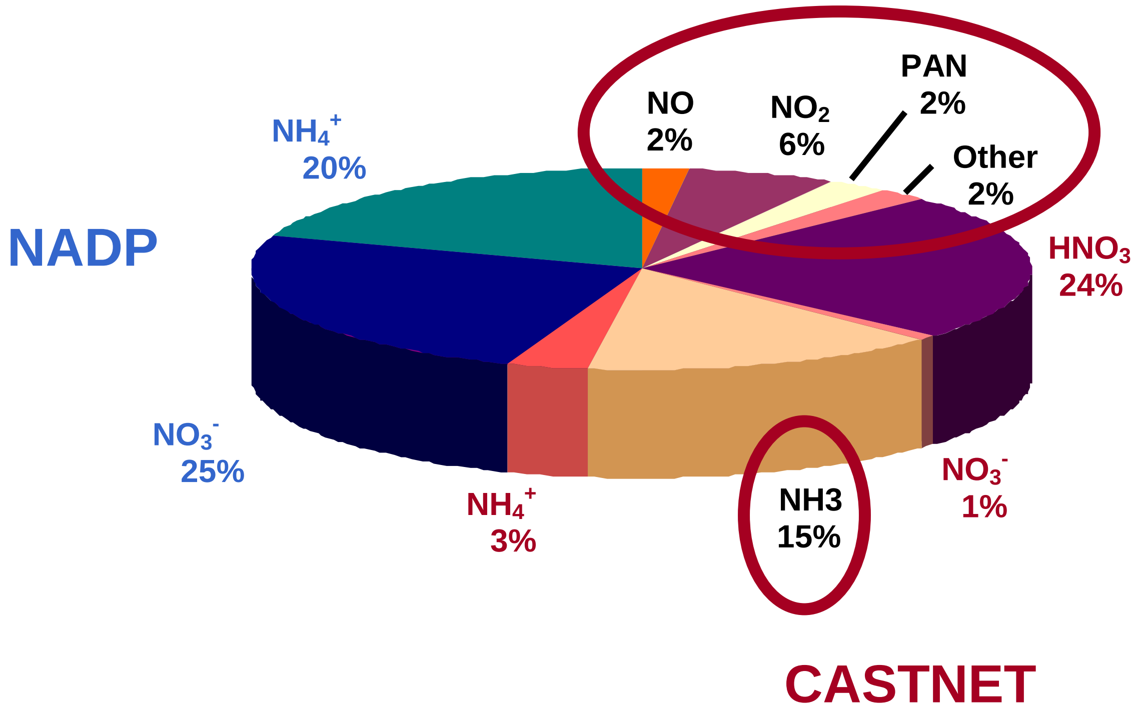

Figure 4. Percentage breakdown of the contribution of reactive nitrogen compounds deposited in the Chesapeake Bay watershed, as predicted by the CMAQ model. Nitrogen species measured by CASTNET are in red letters. Nitrogen species measured by NADP are in blue letters. Circled nitrogen species (black letters) are measured by neither program. NOx is gaseous (NO + NO_2). PAN is peroxyacetyl nitrates, and NH_3 is gaseous ammonia.

CASTNET has more than 50 sites in the 31 states bordering and east of the Mississippi River, which afford adequate spatial coverage for producing maps and depicting concentration changes (e.g., Figure 2). Airborne sulfur concentrations from CASTNET, together with precipitation sulfur concentrations from NADP, have made it possible to evaluate the impact of SO_2 emissions reductions on air and precipitation quality in the eastern United States. Outside of the 31 eastern states, however, CASTNET has fewer than 30 sites, limiting comparisons in space and time to only portions of the 17 western states. The complex western terrain further limits the representativeness of spatial interpolation schemes for both networks.

CASTNET concentration and meteorological data make it possible to calculate dry deposition fluxes as the product of pollutant concentrations and deposition velocities, determined by the MLM. Since dry deposition is a surface phenomenon affected by land cover (soil, rock, water), vegetation (type, leaf area, age, status), and wetness, these calculations are point-only estimates. Models that account for these surface characteristics over a continuous landscape are needed to extend these site-specific estimates to spatial estimates. While CASTNET data make point estimates possible, there are no actual point measurements of dry deposition for verification of these models or validation of this inferential approach. Further, the inferential calculations use weekly-average pollutant concentrations and deposition velocities based on hourly-average meteorological measurements. CASTNET measurement protocols preclude any evaluation of the potential effects of the coincidence of high pollutant concentrations and high deposition velocities. Real-time or near real-time measurements could be used to evaluate whether covariance in concentrations and deposition velocities is important in determining deposition fluxes.

CASTNET developed with a focus on acidifying pollutants, especially the sulfur and nitrogen precursors resulting in acid rain. The purview of the scientific and regulatory communities has expanded to other important environmental and health problems. It is important that the CASTNET program be responsive to the needs of scientists and regulators for additional measurements, enhanced spatial coverage, and increased temporal resolution. New methods and technologies are available for monitoring chemical species that previously could not be measured in a routine network. CASTNET needs a strategy for modernization that can adapt to contemporary needs without compromising ongoing program requirements.

EPA Goals.

CASTNET data are integral to the ongoing EPA requirement for accountability in assessing the efficacy of emissions controls under the ARP, NBP, and other programs. Figure 5 presents the framework in which EPA evaluates progress toward regional and national goals for improved environmental and human health that can be realized from reductions in airborne pollutants. Addressing the question of whether program goals are being met requires the monitoring of emissions, air quality and deposition, and ecosystem response. Monitoring must be complete, accurate, and representative. Linking ecosystem changes to pollutant emissions reductions requires that monitoring be long-term, as well, since emissions reductions are incremental, air quality and deposition are highly variable, and ecosystems generally are buffered from rapid change.

EPA's goal for CASTNET is to provide air concentration and deposition data that best contributes to answering the question of accountability, i.e., providing data that effectively informs policy. This requires measuring the chemical species important in affecting ecosystem and human health. To best address the question of accountability now and in the next decade, CASTNET may need some or all of the following measurements on broad or limited spatial or temporal scales: gaseous NH_3, total reactive nitrogen, and organic carbon, nitrogen, and sulfur.

NPS Goals.

As conservator of U.S. National Parks, the NPS uses CASTNET data to ensure that air quality does not adversely affect the natural resources, scenery, wildlife, or historical objects in the parks. Of special interest is determining that O_3, NO_x, and SO_2 concentrations meet National Ambient Air Quality Standards (NAAQS), and determining that these pollutants, or aerosols derived from them, do not degrade visibility or otherwise interfere with the enjoyment or health of park visitors. In addition, the NPS is investigating whether exposure to ambient air concentrations or chemical deposition is at or above levels (i.e., a "critical load") harmful to naturally occurring vegetation or to terrestrial or aquatic wildlife. CMAQ and ecosystem models are used in these investigations to estimate exposure levels and critical loads.

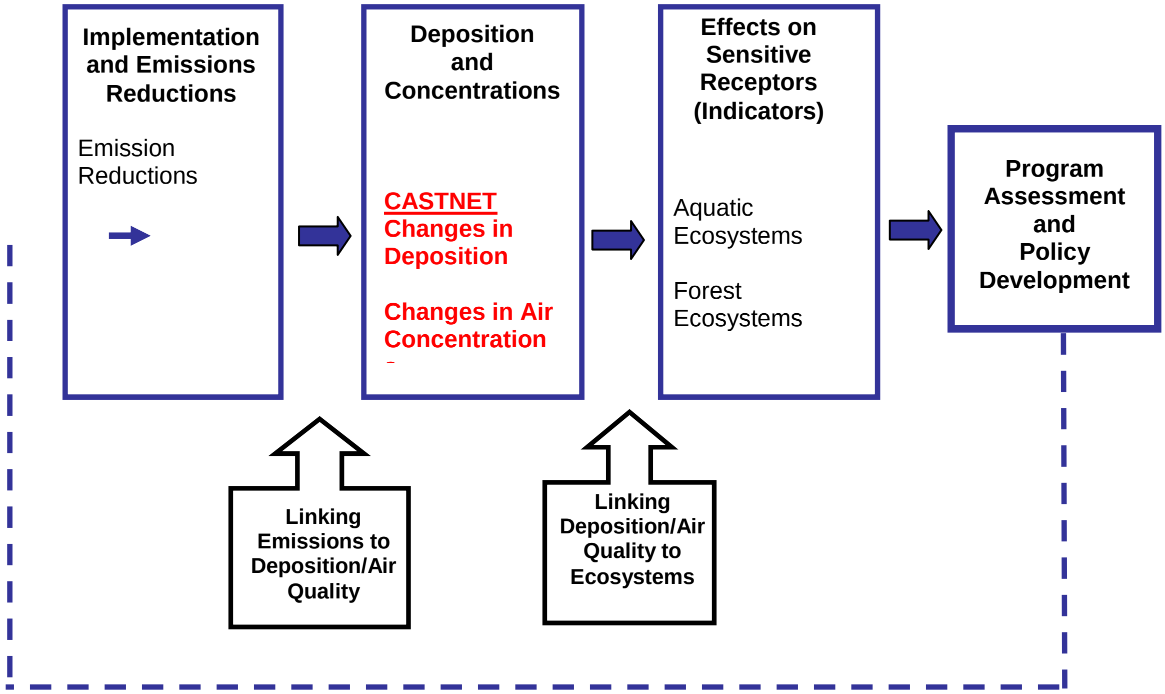

Figure 5. Framework for EPA assessments of emissions reductions programs and policy development and evaluation. The CASTNET monitoring program (red) is integral to the assessment process.

The NPS goal is to utilize CASTNET data to identify locations where National Park resources are potentially degraded by exposure to air pollutants or atmospheric deposition. CASTNET measurements provide data that can be used to evaluate average exposure, seasonal and secular trends in exposure, and estimates of deposition fluxes. Where NAAQS are violated, visibility is impaired, or critical loads are exceeded, however, NPS scientists require real-time or near real-time data to identify pollutant sources and apportion their role in effecting these exceedances.

Recent measurements and model estimates of the dry deposition of nitrogen in Rocky Mountain National Park indicate the importance of gaseous NH_3 and NO_x measurements, not currently measured by CASTNET. These results are consistent with CMAQ estimates of deposition from NH_3 and NO_x in the Chesapeake Bay watershed. In addition, the Rocky Mountain study reported significant amounts of gaseous and particulate organic nitrogen, as well as organic nitrogen in precipitation. Though additional research is needed to identify atmospheric organic nitrogen compounds and their emission sources, monitoring programs may need to consider routine measurements of total organic nitrogen in order to determine its importance in the overall nitrogen deposition budget.

Modeling.

Models are an essential tool for analyzing the complexity of pollutant source-receptor relationships and an integral element in an assessment framework. CMAQ simulations of the forms and amounts of nitrogen deposited in the Chesapeake Bay watershed (Figure 4) provide valuable insights into the limitations of CASTNET measurements in quantifying nitrogen deposition in this estuarine system. Large differences between modeled and measured concentrations reveal the potential for errors or limitations in one or both systems. This information provides the impetus for change and improvement.

The CASTNET measurement system lends itself to operational evaluations of rural O_3 and SO_2, particulate NH_4^+ and SO_4^{2-}, and total nitrate (HNO_3 and NO_3^-), though these evaluations are complicated by the potential biases in nitrogen measurements (NH_4^+, HNO_3, and NO_3^-). In addition, CASTNET O_3, SO_2, and SO_4^{2-} data are adequate for trends analyses and as input for models that examine the linkages between emissions changes and trends in these species. The week-long sampling frequency, however, does not provide the temporal resolution needed by models designed to examine and diagnose atmospheric transport and transformation processes. And, model verification would benefit from direct dry deposition measurements, rather than the estimates calculated from CASTNET measurements.