TECHNICAL PAPER

ISSN 1047-3289 *J. Air & Waste Manage. Assoc.* **56**:1115–1129

Air Emission Inventories in North America: A Critical Assessment

C. Andrew Miller
U.S. Environmental Protection Agency, Office of Research and Development, National Risk Management Research Laboratory, Research Triangle Park, NC

George Hidy
Envair/Aerochem, Placitas, NM

Jeremy Hales
Envair, Pasco, WA

Charles E. Kolb
Aerodyne Research, Inc., Billerica, MA

Arthur S. Werner and Bernd Haneke
MACTEC Federal Programs, Inc., Durham, NC

David Parrish
National Oceanic and Atmospheric Administration, Earth System Research Laboratory, Chemical Sciences Division, Boulder, CO

H. Christopher Frey
North Carolina State University, Department of Civil, Construction, and Environmental Engineering, Raleigh, NC

Leonora Rojas-Bracho
Instituto Nacional de Ecología, Dirección General de Investigación Sobre la Contaminación Urbana, Regional y Global, Mexico City, Mexico

Marc Deslauriers
Environment Canada, Criteria Air Contaminants, Gatineau, Quebec, Canada

Bill Pennell
NARSTO, Columbia Research and Education Associates, Pasco, WA

J. David Mobley
U.S. Environmental Protection Agency, Office of Research and Development, National Exposure Research Laboratory, Research Triangle Park, NC

ABSTRACT

Although emission inventories are the foundation of air quality management and have supported substantial improvements in North American air quality, they have a number of shortcomings that can potentially lead to ineffective air quality management strategies. Major reductions in the largest emissions sources have made accurate inventories of previously minor sources much more important to the understanding and improvement of local air quality. Changes in manufacturing processes, industry types, vehicle technologies, and metropolitan infrastructure are occurring at an increasingly rapid pace, emphasizing the importance of inventories that reflect current conditions. New technologies for measuring source emissions and ambient pollutant concentrations, both at the point of emissions and from remote platforms, are providing novel approaches to collecting data for inventory developers. Advances in information technologies are allowing data to be shared more quickly, more easily, and processed and compared in novel ways that can speed the development of emission inventories. Approaches to improving quantitative

IMPLICATIONS

Emission inventories are the starting point for managing air quality. Shortcomings in data and methods used to develop current emission inventories can lead to potentially ineffective air quality management strategies. By understanding these shortcomings (and emission inventory strengths), air quality managers can identify what new technologies can be applied and what additional data are most likely to provide the greatest improvement in airshed characterization. The recommendations provide a guide for what improvements are most important and most likely to result in improved air quality management capabilities.

measures of inventory uncertainty allow air quality management decisions to take into account the uncertainties associated with emissions estimates, providing more accurate projections of how well alternative strategies may work. This paper discusses applications of these technologies and techniques to improve the accuracy, timeliness, and completeness of emission inventories across North America and outlines a series of eight recommendations aimed at inventory developers and air quality management decision-makers to improve emission inventories and enable them to support effective air quality management decisions for the foreseeable future.

INTRODUCTION

Air quality management (AQM) in North America focuses on ensuring that concentrations of compounds in the ambient air are below the levels that are considered harmful to human health or the environment. Strategies developed to achieve these standards are based on reduction of emissions from specific source classes. The effectiveness of this approach depends on an accurate understanding of the relative contributions of the sources to ambient atmospheric pollution.

An adequate knowledge of emissions sources and associated fluxes, both before and after emission controls are adopted, has long been recognized as a requirement for designing cost-effective air pollution control strategies.[1] Emission inventories are designed to systematically quantify the temporal and spatial distributions of the fluxes of primary pollutants and secondary pollutant precursors emitted by significant sources. This places emission inventories at the foundation of today's AQM strategies, and significant errors in inventories can, therefore, lead to the adoption of strategies that protect human health and the environment less effectively than possible. Emission inventory errors can be enormously expensive by requiring installation and operation of air pollution controls beyond the minimum needed and by failing to effectively reduce adverse health and environmental damage.

The purpose of this paper is to identify the status of current emission inventory practices, point out the general strengths and weaknesses of existing inventories, and suggest possible directions for improving future inventories. The suggested directions are based on the recommendations developed as part of the recent assessment of emission inventories by NARSTO.[2]

BACKGROUND

Several recent reports have recognized the importance of emission inventories and the challenges that must be overcome to ensure that inventories are able to provide the quality of information needed to support sound AQM decisions. In the latest of these reports, the National Research Council (NRC) noted in 2004 that, "The first step in developing an emission-control strategy for a criteria pollutant is to develop an inventory of pollutant emissions that lists all sources of the pollutant or its precursor and the rate at which each source emits the pollutant to the atmosphere."[3]

In response to the NRC recommendations, the Clean Air Act Advisory Committee, an advisory committee to U.S. Environmental Protection Agency (EPA), created an Air Quality Management Work Group to develop a plan to address the recommendations in the report. The Air Quality Management Work Group agreed with the need to strengthen emission inventories to ensure adequate support for AQM strategy development and concluded that, "A strong national effort is needed to improve emission estimation methods for major source categories, especially for sources that are poorly characterized or whose emissions estimates are uncertain."[4] In other scientific reviews conducted by the NRC[5,6] and NARSTO,[7,8] emission inventories have consistently been seen as needing improvement to enable them to continue to be of use in the development of effective AQM strategies.

Given the consistent call for improvements in inventories of air pollutant emissions by these diverse expert panels, it would be easy to conclude that air pollutant emission inventories are severely flawed and are of little effective use to air quality managers. However, measurements show that U.S. emissions of the pollutants addressed in the original Clean Air Act have decreased, in some cases enormously, over the past 20 yr. Ambient concentrations of those pollutants have also decreased significantly, although economic and personal activities responsible for those pollutant emissions have increased considerably over that period.[9] This would suggest that air quality managers have had a good understanding of what emissions sources to control to improve air quality.

Each of the expert panels cited above examined specific applications of emission inventories and the ability of emission inventories to support future AQM decision-making. Although national emission inventories are currently capable of estimating the average annual emissions on a national scale, those same inventories have shortcomings when used in other contexts, such as estimates of daily emissions in a local area.

Most AQM goals have focused on emissions from major, and relatively well characterized, source categories. As recently implemented regulatory programs take effect, however, emissions from these sources will decline substantially. The remaining emissions will be more evenly distributed over source categories that are more difficult to measure or model. A key example of this changing context is the recent promulgation in the United States of the Clean Air Interstate Rule[10] and the rules to reduce emissions from on- and off-road diesel vehicles.[11,12] Together these rules address emissions from the largest source categories of nitrogen oxides (NO_x) and SO_2, mobile sources and coal-fired electric generating units, respectively. As these rules begin to take effect, these sources will no longer be as dominant in the total U.S. emissions of NO_x and SO_2 (see Figure 1), making other source categories relatively much more important.[13] In this and similar situations, errors in emission estimates from smaller individual sources will have greater consequences than were previously the case. These consequences could range from wrongly identifying a pollutant that should be controlled to overlooking source categories

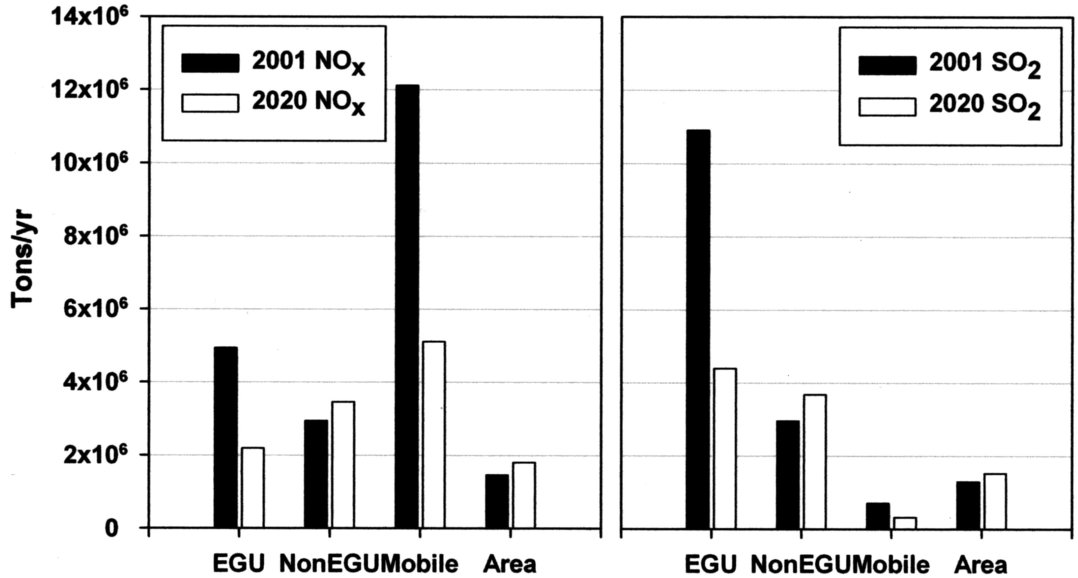

Figure 1. Estimates (2001) and projections (2020) of annual U.S. emissions of NO_x (left) and SO_2 (right) from electric utility-generating units (EGUs), non-EGU, mobile, and area sources.[8]

of which the control could result in more cost-effective emission reductions.

In addition to this loss of dominating sources, future AQM strategies are beginning to consider all emissions into an airshed, including hazardous air pollutants ([HAPs] or air toxics) that have previously been considered separately from the criteria pollutants. This approach is consistent with one of the NRC Air Quality Management recommendations to evaluate the entire load of pollutants entering and emitted within an airshed, rather than managing them individually.[3] To address this recommendation, inventories of all pollutants would need to be at least compatible with one another and ideally integrated into a single coherent inventory.

The changing context in which emission inventories are being used, a greater need to understand the limitations associated with emission inventory data, and a higher expectation for rapid and flexible data availability are placing tremendous pressure on the developers of emission inventories to provide accurate, timely, accessible, and flexible emission inventory databases. To address these issues, NARSTO recently released an assessment of emission inventories across North America, which included eight recommendations for improvements.[2] Although the focus of this assessment was on national emission inventories, the recommendations are applicable to international, regional, state and provincial, and local inventories as well. The recommendations were developed with a North American rather than strictly a U.S. perspective. Although the NARSTO assessment identified differences in emission inventory development and needs across Canada, Mexico, and the United States, the majority of issues and fundamental needs are common to all three countries. Much of the following discussion is drawn from the NARSTO emission inventory assessment, with the same focus on North American inventories.

EMISSION INVENTORY EVOLUTION AND STRUCTURE

Most early emission inventories were developed to help address air quality problems around specific major metropolitan areas,[14–16] as recognition grew during the 1950s and into the 1960s that air pollution was a significant public health problem.[17] To more accurately estimate emissions in these areas with the technology available at that time, factors were developed that related emissions to industrial and other activities. These emission and activity factors allowed air quality managers to estimate how changes in activity levels or technologies impacted total emissions without requiring measurements at every facility, and this approach has formed the foundation of modern emission inventories. Although early emission inventory developers confronted many problems similar to those faced by current efforts, there are also some significant differences. The sampling methods, for instance, have fortunately advanced considerably since 1957 when Rossano and Schell stated that, "Observing the effluents where possible, and even smelling or feeling them may provide useful information."[18]

In their most simple form, emission inventories are developed using emission factors (*EF*s) and associated activity (*A*) information. Emission factors are the mass of pollutant emissions released per unit of the associated process variable. Activities are the related process variable, such as mass of fuel consumed or output produced. The emissions (*E*) are then calculated as:

$$EF \times A \times (1 - [ER/100]) = E \quad (1)$$

where *ER* is the emission reduction (in percent) associated with use of a pollution control system.

In lieu of using an emission reduction factor, a different EF can be used, particularly if the pollution control approach involves a modification of the process. The $EF \times A$ structure assumes that the emission factor is independent of the activity factor. Where the activity is a process variable, for example, load or throughput, this assumption may not be accurate, because it is possible for the emission factor to vary as the process varies. On the other hand, when the activity measure is the population of similar sources, such as the number of dry cleaners, the assumption of independence is reasonable.

Over a large number of sources and a long period of time, variations in the emission rate can be expected to even out so that the emission factor adequately reflects the average emission rate across the activity range; this allows emission factors to be used as the basis for developing national annual emission inventories. EPA compiles a database of emission factors for a wide variety of source types and a range of pollutants in its "AP-42" document. The stationary source volume of AP-42 is currently in its fifth edition and has 15 chapters covering sources as diverse as external combustion sources (such as boilers and process heaters), storage tanks, and ordnance detonation.[19]

In spite of the long history of the emission factor approach, it does have significant shortcomings. As far back as the 1950s, issues such as temporal and spatial allocation of emissions or nonlinear relationships between activity and emission levels were recognized.[20] Even 50 yr later, these complexities continue to present problems for emission inventory developers. To address some of these issues, emission models have been developed to more accurately estimate emissions from sources with complex operating characteristics that cannot be accurately represented by the simple relationship in eq 1. The more well known of these models have been developed for estimating mobile source emissions. Although early mobile source inventories used an average emission factor and fuel consumption data to estimate emissions, current emission models account for changes in fuel type, fuel evaporation, engine deterioration, operation of air conditioning, and engine startup in addition to the variation in emissions because of differences in vehicle design.

Over the years, emission models have grown in scope and complexity. Models for on-road and nonroad mobile sources have been developed by EPA and others.[21–24] Other emission models have been developed to estimate emissions from vegetation and soil.[25] Pollutant-specific models have also been developed, such as the Carnegie-Mellon University ammonia (NH_3) emissions model, which estimates NH_3 emissions from animal feeding operations, wastewater treatment, and mobile sources, as well as from natural processes.[26]

Figure 2 illustrates how these different components (emission factors, activity factors, and emission models) combine to create a complete inventory.[27] For stationary point sources, one can determine emissions using the appropriate emission factors combined with the facility-specific activity factors. Emissions for nonpoint sources are estimated using the EPA MOBILE, NONROAD, and Biogenic Emission Inventory System (BEIS) models, among others. These annual emission estimates are then combined to determine the total estimated emissions. However, modern inventories involve many additional elements to ensure that they are as relevant as possible to current AQM needs.

CURRENT INVENTORIES

The growth of emission models is a reflection of the increasing scope and complexity of emission inventory needs and of emission inventories themselves. The general structure of the current national emission inventories (NEIs) was derived from the early metropolitan-area inventories beginning in the 1970s in the United States and Canada.[28] The 1985 National Acid Precipitation Assessment Program emission inventory was the first attempt of EPA to produce a highly quality-assured national inventory for use by policy-makers, modelers, human and ecological effects researchers, and industry and set the stage for the NEIs today.[29] These inventories (and this

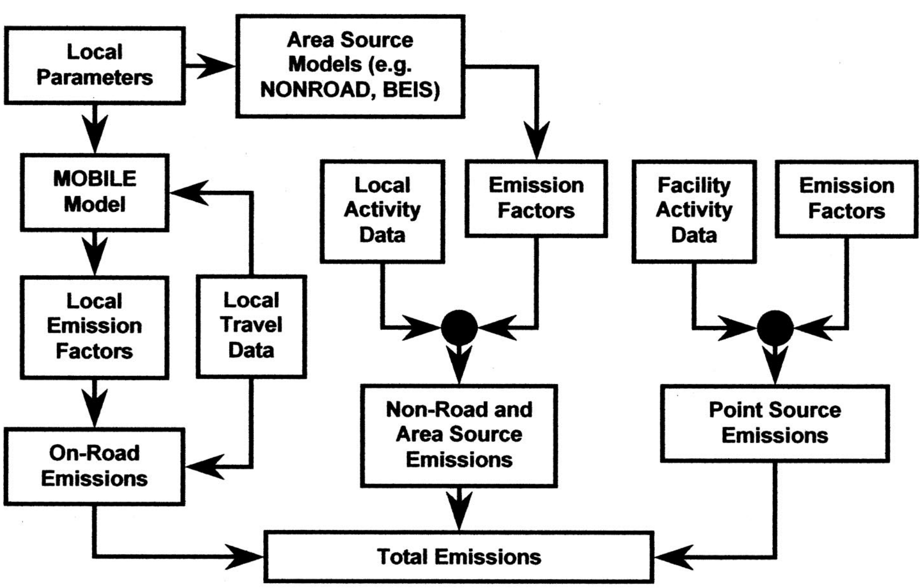

Figure 2. Simplified schematic of the structure of an emission inventory that uses emission and activity factors and emission models.[26]

paper) focus on criteria pollutant emissions and typically include CO, NH_3, NO_x or NO_2, particulate matter ([PM] including specific size fractions such as coarse PM and fine PM [$PM_{2.5}$]), SO_2, and volatile organic compounds (VOCs).[2,30]

The NEIs, in turn, have provided the structure, and often the data, for emission inventories at the local, state and provincial, and regional levels. These inventories are developed to address more specific AQM issues, such as regional haze, implementation of local AQM plans, or cross-border air quality problems.[31–34] Specialty emission inventories, such as those for dioxins and dioxin-like compounds,[35] mercury,[2,36] and black carbon,[37] are also needed to address emissions of specific pollutants. These regional and specialty inventories generally follow the NEI approach that relies on emission and activity factors and emission models as a guide to determine what information, and in what format, is needed. Each of the variations on the NEI challenges the ability of the basic inventory structure to meet the specific inventory needs. Probably the greatest challenge that modern inventories face, however, is their use as the basic source of emissions data for air quality models.

The need to design and implement AQM plans to reduce ambient concentrations of ozone has driven the development of increasingly complex models of atmospheric transport and photochemical reactions, and modeling the atmospheric formation of secondary PM even more complex. Because many of the ozone formation processes occur in the atmosphere on time scales of hours or less and because the chemistry included in these models is necessarily complex, these models require increasingly detailed information on the location, time, and chemical speciation of emission fluxes of the major ozone precursors, NO_x and VOCs.[38] Because emissions data are generally available as annual averages (except for some major source categories, such as electric generating units), annual emission inventory data are fed into processing models (emission processors) that allocate emissions temporally (and spatially for area sources) to simulate the actual hourly, daily, and seasonal changes in emissions. Such simulations must account not only for these temporal emission changes but also changes in meteorological conditions. Processes such as space heating and cooling, evaporative and biogenic emissions, and even traffic patterns change as meteorology changes. These effects add further complexity that must be accounted for when developing emission inventories.

Emissions processors operate on the base inventory data to provide model-ready detailed emissions, as illustrated in Figure 3.[39] Processors such as the Sparse Matrix Operator Kernel Emissions model,[40] the Emissions Processing System, and the Emissions Modeling System generate input files for the atmospheric models. These processors typically provide hourly emissions over the course of a week, allocated to grid cells over the model domain, and with the speciation required by the chemistry model being used. The detailed chemical species are typically contained in a separate database that lists speciation profiles for a range of chemical compounds usually emitted

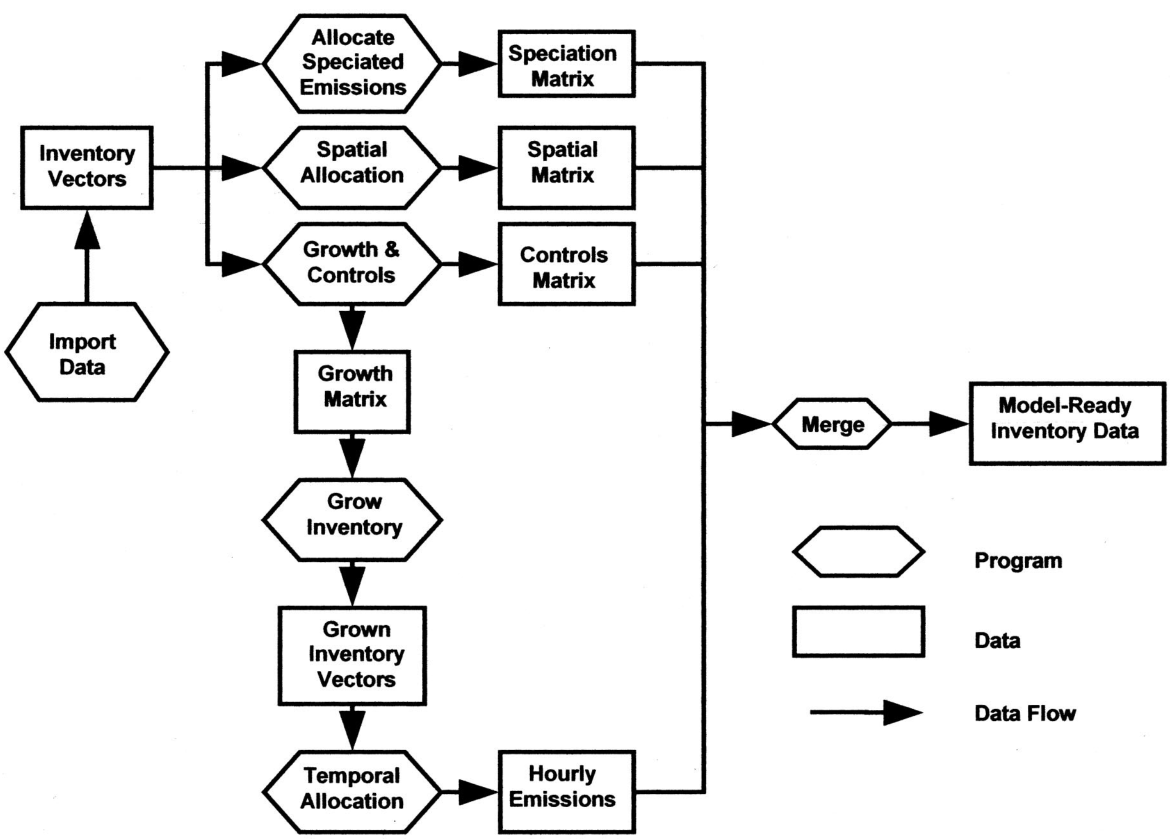

Figure 3. Schematic of data flow in SMOKE emission processor. A linear processor would apply each program consecutively rather than in parallel.[38]

by a given source type. A database that has been widely used is SPECIATE, from EPA, which contains speciation profiles for a wide range of source categories.[41]

As this introduction has illustrated, the scope of the applications of emission inventories is, thus, extremely broad and can range from hours to decades on a temporal scale and from neighborhood to global on a spatial scale. These widely differing scales and purposes can result in significant mismatches between the emission inventory data and the needs for those data, given the temporal or spatial scales or degree of speciation available. These mismatches act to highlight the shortcomings of existing emission inventories, and these will be discussed in more detail below. Although emission inventories are not yet ideal, considerable work has been done over the past 30 yr to strengthen their ability to provide critical information for developing successful AQM strategies.

EMISSION INVENTORY STRENGTHS

Current emission inventories can, in general, be used to compare the relative significance of different source categories. Major insights can be drawn from the current U.S. NEI: the largest fraction of NO_x and SO_2 emissions and a considerable portion of VOC emissions are from stationary sources; mobile sources are the largest contributor to total CO emissions and a considerable contributor to total NO_x and VOC emissions; and biogenic sources contribute the largest portion of total VOC emissions.[9,42] On an aggregated national annual emissions basis, these insights enjoy a high degree of confidence.[2] Similarly, air quality managers in Canada and Mexico are able to identify the key sources of concern based on information derived from their respective national inventories.

Existing emission inventories provide insight into air quality trends over time and overall pollution control efficiency. Comparison of current emission inventories to those from previous years provides the basis for estimating emission trends over time. Such comparisons can give an indication of efficiency of particular control strategies, particularly those that are national in scope, such as reductions in SO_2 and NO_x from large point sources in the United States associated with acid rain provisions of the Clean Air Act Amendments of 1990.

AQM strategies can be developed based on current emission inventories. In addition, priorities for air quality improvements can be set based on the knowledge of emissions contributed from different source categories.[43] With limited resources, such prioritization focuses efforts on those sources with the greatest potential to reduce emissions. For example, in urban areas facing ozone problems, the relative importance of NO_x versus VOC control can be assessed taking into account both urban and regional geographic scales, and the key source categories that should be the focus of control efforts can be broadly prioritized.

Many of the tasks required to develop current emission inventories are completed using existing emission models and tools. Tools such as MOBILE, TANKS (for estimating VOC and HAP emissions from organic liquid storage tanks), or the Emission and Dispersion Modeling System (for estimating emissions from aircraft and airport operations) help automate many of the tasks involved in developing emission inventories, thereby reducing the cost of inventory development. A further benefit is that the use of many of these tools leads to the standardization of emission inventory development efforts, thereby increasing their compatibility across emission inventories.[2]

INVENTORY NEEDS AND SHORTCOMINGS

In an ideal situation, the information reported in the different emission inventories would all be based on a single set of basic data that could then be compiled to provide the information needed at a particular time for a particular application. The basic data would be updated frequently as new information was received and would include the appropriate metadata describing the information source, data collection methods used, limitations (including variability, uncertainty, and applicability), and other descriptors. Inventories would be compiled on request, and updates would be available within days, if not hours, of new information being submitted.

The NRC Air Quality Management panel emphasized such a need for compatible inventory data, given the stress on managing airsheds as a single entity rather than managing individual pollutants. Although the latter approach is driven by the regulatory structures created (in the United States) by the Clean Air Act, the NRC pointed out that the appropriate scientific structure is to evaluate emissions and AQM on a scientifically based foundation of a single airshed that will be affected by each AQM strategy regardless of its regulatory basis.[3] Thus, the NRC recommendations apply equally to emission inventories across North America.

In such a situation, different inventories would be compatible and comparable and near real time. Users would be able to understand how certain the data were and would have the ability to evaluate how uncertainties propagated through analyses that used the inventory data. Air quality managers and policy makers would have the latest information on emissions and a measure of how the data would be likely to change based on the uncertainty and variability information.

Unfortunately, we are far from this ideal situation. There can be considerable uncertainty in the data reported in emission inventories for a number of reasons.[44,45] The shortcomings of current emission inventories are closely related to these sources of uncertainty. For instance, many, if not most, source emission estimates are based on a small number of measurements that do not adequately represent the full range of process designs and operational practices. This limits the accuracy and increases, sometimes significantly, the uncertainty of the estimate.[3,6] Inventories that rely on emission factors typically do not account for changes in emissions during startup and shutdown or often during significant operational changes. Although emissions models can estimate these emissions, they are not often included, particularly for stationary sources.[2] Area source emissions are often spatially allocated at the county level, requiring estimates of spatial allocation factors to be used if the inventory data are to be used for air quality modeling. Similarly, emission inventories are usually based on annual average values, which may be far from actual values over the hour-long time frames of interest to air quality modelers.

The emission inventories developed at the county, state or provincial, and national levels are often inconsistent. Data from different agencies may be developed using different estimating procedures, several similar emissions source types may be consolidated, and information regarding data use limitations may not be carried through as data are combined to create inventories that cover a larger geographical range. Conversely, data that are adequate at a national level can become much more uncertain when disaggregated to estimate emissions at a state or local level because of differences in the source mix, operating conditions, and other factors.[2] Mechanisms are needed to ensure that the aggregation or disaggregation process is appropriate and consistent within and across different countries and, in some cases, across agencies within a country.

Uncertainties are greater for some sources and pollutants than for others. Emissions of NO_x and SO_2 from electric utility generating units (EGUs) in the United States, for instance, are measured using continuous emissions monitors, and the data are reported to EPA each quarter. These data are accurate and well resolved spatially and temporally, but data on other pollutants and other sources are much more difficult to obtain. In particular, emission inventories of HAPs are more uncertain than criteria pollutants, and emissions from natural, area, and mobile sources are more uncertain than those from large stationary point sources. Emissions from anthropogenic sources are much more well characterized than emissions from natural sources, and, hence, the emission estimates for natural sources are often much more uncertain.[42] A relative comparison of the qualitative confidence levels for national inventories of key gaseous pollutants is shown in Table 1.[2,7]

Uncertainties also arise because of the time intervals between updating and reporting of emission inventories and between sampling and reporting of emissions data. As the period between data collection and data reporting lengthens, the uncertainty of the reported data to represent actual emissions increases. The U.S. NEI is updated every 3 yr and, thus, may not provide timely and updated emission information for air quality management decisions. In addition, many of the emission factors and speciation profiles are based on measurements that are more than a decade old, resulting in questionable applicability of the measurements.[2] Efforts are being made to reduce these intervals, notably in Canada, which has as a goal annual updates to their emission inventories.

Typically, the uncertainties associated with the inventory data are not reported, and often, the information needed to quantify the uncertainties is not collected. Quality assurance and quality control procedures are not

Table 1. Estimated relative confidence levels of national emission inventory data for selected pollutants.[2]

		Estimated Confidence Levels in Overall Inventory		
Pollutant	**Source**	**Canada**	**United States**	**Mexico**
SO_2	Utilities	High	High	High
	Other point sources	Medium	Medium	Low-medium
	On-road mobile	Medium	Medium	Low
	Nonroad mobile	Low-medium	Medium	Low
	Area sources	Low	Low	Low
	Biogenic source	Low	Low	Low
	Other man-made sources (noncombustion)	Low	Low	Low
NO_x	Utilities	Medium-high	High	Medium
	Other point sources	Medium	Medium	Medium
	On-road mobile	High	High	Medium
	Nonroad mobile	Medium-high	Medium-high	Low
	Area sources	Low	Low	Low
	Biogenic source	Low	Low	Low
	Other man-made sources (noncombustion)	Medium	Medium	Low
VOC	Utilities	Medium-high	Medium-high	Medium
	Other point sources	Low-medium	Low-medium	Medium
	On-road mobile	Medium	Medium-high	Low
	Nonroad mobile	Medium	Medium	Low
	Area sources	Low	Low	Low
	Biogenic source	Low	Low	Low
	Other man-made sources (noncombustion)	Medium	Medium	Low
HAP	Utilities	Medium	Medium	Medium
	Other point sources	Low-medium	Low-medium	Low
	On-road mobile	Medium	Medium	Low
	Nonroad mobile	Low-medium	Low-medium	Low
	Area sources	Low	Low	Low
	Biogenic source	Low	Low	Low
	Other man-made sources (noncombustion)	Low	Low	Low

Notes: Confidence levels for SO_2, NO_x, and VOC are from the NARSTO PM Assessment.[7]

strictly applied in most emission models or during inventory development, resulting in unknown levels of data certainty. For most emission inventories, documentation of the key assumptions and data sources used during the emission inventory development is inadequate. Although the AP-42 emission factors compilation provides some guidance on the uncertainty of emission factor data, the quality ratings are not quantitative.[19] EPA is in the process of revamping the emission factor program. One goal of this program is to provide quantitative uncertainties and guidance on the use of factors.

FUTURE DIRECTIONS AND TRENDS IN INVENTORY DEVELOPMENT

Many new tools and techniques have been evaluated and applied to reduce the uncertainties associated with emission inventories. Most of these techniques are associated with emission measurements, but a number of developments in information technology and computational models are improving inventories through better information processing and expanded capabilities to evaluate the accuracy of inventories. In many cases, combining innovative measurement and computational techniques can provide new approaches for improving emission inventories.

Remote Sensing

Many of the new emission measurement technologies adopt remote sensing rather than conventional "probe-in-stack" approaches. Ground-based remote sensing methods that rely on absorption spectroscopy include nondispersive infrared ([IR] NDIR) techniques, Fourier transform IR (FTIR) methods, differential optical absorption spectroscopy (DOAS), and tunable IR laser differential absorption spectroscopy (TILDAS).[2] FTIR has been used to measure NH_3 emissions from animal feeding operations,[46,47] a range of organic compounds from combustion processes,[48] and emissions from natural gas flares.[49] DOAS measurements have been combined with dispersion modeling to estimate VOC emissions from gasoline service stations and tanker filling operations.[50] Other ground-based remote sensing methods rely on fluorescence or Raman spectroscopy or light detection and ranging (LIDAR) techniques.[51]

Ground-based remote sensing has been especially useful in evaluating emissions from mobile sources. The most common approach has been to conduct measurements across roadways and intercept the exhaust plumes from vehicles moving across the measurement path. NDIR techniques were used in some of the earliest studies to measure CO and CO_2, and, more recently, emissions of HC and NO_x.[52–56] More advanced TILDAS applications have been developed to measure a wider range of species, including several nitrogen and organic compounds.[57] Still in the development stage are dispersive IR techniques, use of LIDAR, and instrumentation that can measure exhaust PM.[58–60] Future directions for this technique include the use of remote sensing to quantify air toxics, PM and PM precursors, and greenhouse gases.[61,62]

Satellite and Aircraft-Based Sensors

Some remote sensing applications are based on satellites or aircraft. Satellite measurements are currently being used to visually identify the location and intensity of large-area fires, ship plume tracks, major industrial plumes, and incidents of dust storms or regional haze.[2,63] Measurements of surface properties, such as ground cover and temperature, may be the most effective current use of satellite data, but progress has been made in using satellite data to infer pollutant column concentrations or densities and continental-scale emissions. Although the visual images collected by satellites have aided in identifying areas of smoke and haze emissions and transport, these data are limited in their capability to provide quantitative data. Numerous technical challenges to the collection of quantifiable pollutant concentration data remain, including compensating for variations in the air chemistry matrix, aerosol burden, cloud cover, surface albedo, and temperature, as well as dealing with masking effects of the stratospheric overburden, which can be dominant. Current technology does not enable effective satellite measurements to be made beneath cloud cover.[2]

Even so, the ability of satellites to cover large, spatial domains is a major advantage over other approaches. Combining satellite measurements with data from other sources, including air quality and atmospheric process models, can provide valuable information in instances where no other measurements are available. Estimates of emissions over a large geographical area, of the relative amounts of natural and anthropogenic emissions, and of emissions where little or no quantitative data are available are well suited for evaluation by satellite-based methods. Perhaps the most effective use of satellite data has been as an independent source of data to which inventory results can be compared to identify inventory gaps and shortcomings. This technique has been applied to biogenic emissions of isoprene, global NO_x emissions, and CO emissions from wildfires.[64–66]

Aircraft remote sensing is most often used to measure the flux of pollutants that pass through a vertical plane intersecting the pollutant plume that is being observed. DOAS, IR spectroscopy, or LIDAR are the typical techniques used in aircraft remote sensing applications.[67,68] Current aircraft remote sensing is largely limited to NO_x and SO_2 fluxes using DOAS and organic species from biomass burning using IR spectroscopy. Even so, aircraft measurements of this type are particularly useful in determining pollutant fluxes from spatially extended area sources, such as forest fires and total emissions from an urban area. There is potential for DOAS techniques to be extended to some VOC species, including formaldehyde, alkenes, and aromatics, and for LIDAR instrumentation to be applied to ozone and PM.[2]

Mobile Source Emission Inventories

Mobile sources present numerous and significant challenges to inventory developers.[6] Not only are emissions from mobile sources a major contributor to ambient air pollution, there is also enormous variability in the many parameters that influence mobile source emissions in terms of mass and composition. Measuring and estimating those emissions have, therefore, been a major focus of emission inventory development from the outset. Mobile source emissions vary with time of day, week, or year; across and within individual sources; and across and within metropolitan areas. Because we are only able to directly measure emissions from an extremely small fraction of the total number of mobile sources, significant efforts are required to extrapolate those measurements to fleet-wide distributions of emissions. Emission models, remote sensing, and statistical methods are a few of the approaches that are used to more accurately estimate emissions from mobile source categories. Because of the importance and difficulties associated with mobile source emissions, a greater amount of attention will be devoted to this sector.

Mobile source emission inventories are primarily developed using emission models that calculate emissions across a designated area, usually on an urban scale. The most widely used model is the EPA MOBILE model (and its derivatives in Canada and Mexico), which is in its sixth major revision.[21,22] The MOBILE model relies on emissions data from dynamometer testing over standard operating cycles, as well as local characteristics, such as fuel composition, climate, and fleet composition. For example, these local characteristics are considerably different for Mexico than for Canada, and the basic structure of the MOBILE model allows inventory developers to account for such differences.

The EPA NONROAD model uses similar techniques to estimate emissions from nonroad vehicles, such as construction and agricultural equipment, railroad locomotives, marine vessels, recreational vehicles, and small engines, such as those used in lawnmowers and leaf blowers.[23] The resulting emissions estimates are idealized to the extent that the standard driving cycles and fleet characteristics do not fully represent real world conditions.[69] Next-generation models, such as the EPA Motor Vehicle Emissions Simulator (MOVES) model, are designed to enable more complex estimates of mobile source emissions by using emission rates based on operational modes that change with location (a "modal" approach).[70] This allows the model to more accurately estimate the time and location of emissions from operating conditions, such as cold starts, extended idling, aggressive acceleration, and altitude influence.

Modal emissions models such as MOVES will require more detailed measurements under real-world conditions. There are several approaches to measuring emissions during actual operations, including remote sensing, tunnel studies, mobile laboratories and chase vehicles, and portable emission measurement systems (PEMS) and on-board sensors.

Remote sensing is generally able to measure the frequency and impact of high-emitting vehicles and to evaluate the effectiveness of AQM approaches, such as the use of oxygenated fuels and inspection and maintenance programs.[57,71–75] Extensive deployment of remote sensing equipment can also provide near-real-time data on roadway emissions that can be used to maintain current emission inventories and provide better data on spatial and temporal emissions distributions. Although remote sensing is usually used with passenger vehicles, the technique has also been extended to heavy-duty diesel trucks and off-road vehicles.[76–78] Remote sensing data do have limitations; in particular, they are measurements at a limited number of locations and, therefore, may not include emissions over the full range of operating conditions, such as cold starts.[79]

Tunnel studies measure pollutant concentrations at the entrance to roadway tunnels and at the outlet of tunnel exhaust air systems as a means to estimate mobile source emissions. Combined with counts of vehicle number and type passing through the tunnel, these measurements can be used to develop fleet-level emission rate distributions.[69,80–83] Although these measurements are limited to the particular mix of vehicles under particular conditions, tunnel studies provide information on the accuracy of mobile source emission model predictions for aggregate emissions and can identify discrepancies with the predicted mixture of emitted pollutants.[69]

Mobile source emissions can also be characterized using mobile laboratories and chase vehicles, which measure emissions from other vehicles during on-highway operation in normal day-to-day traffic conditions. Both of these approaches rely on fast-response instrumentation, usually mounted on truck beds or in trailers. The instrumentation can be as simple as a single monitor for gaseous pollutants or as complex as a full suite of sampling equipment for gases, particles, and operating parameters. For instance, the mobile laboratory described in Kolb et al.[84] is equipped to quantify exhaust emissions of gaseous CO, NO, NO_2, HONO, NH_3, H_2CO, CH_3CHO, CH_3OH, benzene, toluene, C_2-substututed benzenes, and SO_2, as well as a range of PM properties, including number density, size distribution, and mass loadings of $SO_4^{=}$, NO_3^{-}, NH_4^{+}, organic carbon species, and polycyclic aromatic hydrocarbons.

Mobile laboratories collect data by measuring the elevated pollutant concentrations along the roadway without resolving emissions from specific vehicles. These studies are analogous to tunnel studies with regard to the fleet-level evaluation of emissions.[84–86] A number of groups have focused on characterizing on-road exhaust emissions of PM, with some placing particular emphasis on concentration and properties of ultrafine particles (e.g., those with aerodynamic diameters $<$100 nm).[87–89] CO_2 measurements are often used as markers to which pollutant measurements are correlated to distinguish between background and roadway emissions and also allow estimates of emissions per unit of fuel consumption.

Chase vehicles follow specific vehicles, either cooperating or noncooperating, and sample the individual exhaust plume of the target vehicle. The chase laboratory must shadow the target vehicle and must contain fast response ($\sim$1 sec) sensors measuring CO_2 and the target pollution of interest. If the target vehicle is cooperating with the chase vehicle, operating parameters can be obtained from sensors on board the target vehicle. Otherwise, speed and acceleration must be inferred from the speed and acceleration of the chase vehicle, in combination with range-finding measurements if available. Chase vehicles have been used to characterize emissions from light- and heavy-duty vehicles, both diesel and gasoline.[84,90–93] Chase vehicle measurements appear to be comparable to cross-road remote-sensing data, with the advantages that a wider range of operating conditions can be sampled and, in many cases, many more exhaust species and properties can be measured.

Collection of emissions data during actual operation can also be achieved by using instrumentation that is installed and operated directly on the vehicles of interest. This can provide a means to measure emissions under real-world conditions without significant modification of the vehicle or removal of the engine. The two major approaches to on-vehicle measurements are PEMS and on-board diagnostic sensors (OBDs).

PEMS require analytical instruments that are sufficiently compact to be mounted on or inside the vehicle and robust enough to withstand the temperature and vibrations experienced during actual operating conditions. EPA has led the development of PEMS, with their first-generation system called the Real-Time On-Road Vehicle Exhaust Gas Modular Flow Meter and Emissions Reporting System, better known as ROVER.[94] ROVER and its follow-on system, the Simple Portable Onboard Test, or SPOT (designed specifically for off-road applications), have the dual goals of providing test beds for improving PEMS technology and encouraging private sector development of similar systems. PEMS have been developed by several organizations and are being deployed in support of field test programs to evaluate compliance and evaluate in-use vehicle emissions for a variety of purposes.[95]

Contemporary vehicles include a range of on-board diagnostics, including exhaust-gas oxygen and temperature sensors and engine-load and fuel-consumption monitors. By installing wireless communications devices to transmit the signals from these sensors, OBDs can provide real-time data that can be used to estimate CO, NO_x, and VOC emissions from target vehicles. Although some testing has been conducted using this approach, comparison with other measurement methods has not yet been done.[96] However, OBDs can be used to identify vehicles that are malfunctioning and likely emitting at higher-than-designed rates. Additional work is needed to compare the ability of OBDs to estimate emissions with PEMS and remote sensing techniques.

Further development of sensor technologies is leading toward "microsensors" that require low maintenance and low operating power and can survive in high-temperature, chemically reactive postcombustion environments. The Argonne National Laboratory has been awarded several patents for "smart" microsensors developed from ceramic-metallic (cermet) materials and that use neural network signal processing to relate electrical signals to gas concentrations.[97] Tests have been conducted to measure CO_2 and O_2, and the sensors may be configured and "trained" to detect other pollutants, such as VOCs. The potential for low manufacturing costs ([U.S.]$ 0.25 per sensor) makes it possible to consider microsensors for a range of applications beyond mobile sources, including as continuous emission monitoring systems for small stationary sources for which monitoring costs are currently prohibitive.

DATA UNCERTAINTY, VERIFICATION, AND QUALITY ASSURANCE

Given the critical roles that emission inventories play in the development of AQM strategies, there is a need to better understand the level of uncertainty associated with inventory data. As noted in the background discussion, as emissions become less strongly dominated by a single source category, the need to quantify the uncertainties associated with emissions data becomes more important to ensure the development of effective AQM strategies. This need has been identified by several review panels, in particular the NRC Panel on Air Quality Management in the United States.[3]

The need to understand uncertainty is driven by a number of factors, most importantly whether the uncertainty in the inventory data is significant enough to impact the effectiveness of proposed AQM strategies or whether reported differences between alternative strategies are meaningful.[2] Uncertainties in inventory data are a consequence of uncertainties, variabilities, data entry, and other errors and assumptions in emission measurements, activity data, and emission models. These uncertainties further propagate through air quality models and projections of future emissions, both of which have their own additional uncertainties.[42,69] The impact of emission data uncertainties can, therefore, be very complex and difficult to isolate. Nevertheless, there are approaches that can be used to quantify uncertainties in emissions data and in its subsequent use. Numerous examples of uncertainty analyses have been reported for evaluations of emissions of NO_x, VOCs, greenhouse gases, selected HAPs, and biogenic emissions.[43,98–104] Among the source categories examined in these evaluations are highway vehicles, nonroad vehicles, electric generating units, and biogenic sources.

The impacts on air quality model results because of uncertainties in emission inventory inputs have been evaluated for several cases, particularly for ozone modeling.[105–109] In some studies, the key variables that had the strongest influence on air quality predictions were identified, providing guidance to air quality managers regarding the information of most importance and where additional efforts should be made to improve emission data quality.

Beyond the approach of using direct measurement data to evaluate the uncertainties associated with emission inventory data, the use of "top-down" evaluations can also provide information regarding emission inventory uncertainty. Top-down evaluations use data sources independent of those used to develop the emission inventory, but still closely related to emissions, as the basis for comparison to inventory data. If chosen correctly, these independent data can provide critical tests of the accuracy of an inventory.

There are several techniques that can be used in top-down emission inventory evaluations. A common approach is to compare ambient measurements with inventory data. Other approaches include comparisons of mobile source emissions based on fuel consumption to estimates based on vehicle miles traveled (VMT), comparisons of emission trends estimated several years apart, comparisons with receptor-oriented model data, and inverse modeling using source-oriented models.[2]

Ambient measurements are often used because it is expected that a change in the emissions of a given pollutant will be reflected in a corresponding change in ambient concentrations, particularly when (unlike ozone and some forms of PM) there is little or no chemical transformation of the emissions. Instances in which temporal trends are not consistent between emissions and ambient concentration data can provide an indication of errors in one or both datasets or evidence that the link between emissions and ambient concentrations is not adequately understood. In some cases, ratios of ambient concentrations can be directly compared with ratios of emitted species. For example, the CO-to-NO_x and benzene-to-acetylene ratios have been used to evaluate mobile source emission inventories.[2,110,111]

Fuel consumption data can also be used as the basis for comparing inventory data to related but independent data. Mobile source inventories are largely based on VMT, with emissions estimated from measurements of emitted pollutant mass per unit of distance. Where data are available that estimate pollutant mass emissions per unit of fuel consumed, databases that report fuel consumption can be used as the basis for estimating mobile source emissions and the results compared with the VMT-based emissions or ambient concentrations. This approach has been used to evaluate mobile source inventories in Nashville, TN, and California.[112,113]

Receptor-oriented models estimate the contributions of specific source types to measured ambient pollutant concentrations by fitting measured concentrations of ambient species to a linear combination of source profiles. The contributions are in effect an inventory of emissions that are responsible for the ambient pollutant level at the location of the receptor (an ambient monitoring site). The use of receptor models is only possible for pollutants for which there are differences in the relative mix of chemical species emitted by different source types. This technique has been used to evaluate inventories of $PM_{2.5}$ and VOCs.[7,114]

Inverse modeling is an additional approach to evaluating emission inventory accuracy. Fundamentally, source-oriented air quality models use emission source strengths and other variables, such as winds, solar insolation, and deposition rates, as inputs into equations describing the physical and chemical processes taking place in the atmosphere to compute the ambient pollutant concentrations within the domain of interest. Inverse modeling involves reformulating the equations used in the model so that the emission source strengths are expressed in terms of the observed concentrations. For several reasons, the inverse modeling technique is usually applied to systems that are large in scale (global or continental) and that are characterized by relatively dispersed sources.[66,115,116]

Top-down methods must be applied with caution to ensure that the comparisons are as appropriate as possible. Although the data used in top-down evaluations are (ideally) independently derived relative to the inventory data, top-down data are similarly subject to data uncertainty and limitations. Ambient data, for instance, include contributions from sources other than the source categories being evaluated, and care must be taken to verify that such contributions are minimal relative to the contributions of the categories of interest. The most effective applications of top-down evaluations are those that are combined with concurrent examination of the original bottom-up inventory data, so that the source of errors can be identified rather than simply stating that the inventory is in error.[117,118]

One of the most important insights from the literature on inventory uncertainty is that it is usually far more efficient and less resource intensive to conduct uncertainty analyses at the time the data are developed and incorporated into the emission inventory methodology rather than conducted after the fact.[43,104] After the fact, the original data are often unavailable or are poorly documented, making it difficult or impossible to quantify uncertainty. Even when the original data are available and are adequately documented, the time required to conduct a retrospective evaluation can be prohibitive.

In addition to conducting a technical evaluation of data uncertainty, it is important to ensure that the process of incorporating data from the enormously wide range of data sources into a single inventory is made as seamless as possible. In the United States, the process for developing the NEI is under evaluation with the purpose of making some fundamental changes to that process.[119,120] Other efforts are under way to develop regional inventories that draw on the benefits of an open-source approach to allow users to make modifications to emission models and other algorithms, with the expectation that such an approach will result in a greater understanding of the inventory process and an increased potential for innovations in inventory data processing.[121]

Some of the possible changes are modification of the emission factor quality rating, ability to more routinely incorporate source test results into the emission factor database, and greater use of internet-based information exchange technologies, such as Extensible Markup Language.[2,122] Development and application of online tools and methods for exchanging, processing, and analyzing information are approaches that are currently widely used in most facets of business and throughout government and will continue to be applied to inventory development and reporting.

CONCLUSIONS AND RECOMMENDATIONS

Although the development of emission inventories is often perceived as being a relatively straightforward process, it is, in fact, a complex and involved undertaking to collect, verify, and aggregate the wide range of data required to create the fundamental picture of the location and amount of pollutant emissions. The past (and ongoing) successes in improving air quality across North America are making it more difficult to maintain the gains already achieved and to continue the effort to protect the public from health problems caused or exacerbated by ambient air pollution, because the most readily controlled sources have been or are being controlled. This combination of process complexity and increasing challenges make it imperative that the measurements, data processing, and modeling required to accurately estimate emissions of air pollutants take full advantage of the innovations that have been developed in recent years.

Numerous technologies and approaches have been demonstrated to be of considerable value to improving emission inventories and are ready to be used to support recommendations made by several independent scientific panels to provide more accurate and timely information on where and when emissions occur that are of importance to maintaining and improving air quality. Although it is important to apply the technologies and methods now available, additional efforts are needed. In addition to further research, organizational efforts are also needed to make the collection and processing of inventory data more seamless.

The NARSTO Emission Inventory Assessment has identified eight recommendations to address the shortcomings of existing emission inventories.[2] They are described below.

Reduce Uncertainties Associated with Emissions from Key Undercharacterized Sources

Focus immediate measurement and development efforts on areas of greatest known uncertainty within current emission inventories. Systematically continue to improve emission inventories by applying sensitivity and uncertainty analyses and by comparing them to independent sources of measured data. Such comparisons will help identify subsequent improvement priorities.

Improve Speciation Estimates

Develop new, and improve existing, source speciation profiles and emission factors plus the related activity data needed to more accurately estimate speciated emissions for particulate matter and its precursors, VOCs, and air toxics.

Improve Existing and Develop New Emission Inventory Tools

Continue the development of new and existing measurement and analysis technologies to enable expanded measurements of emissions and ambient concentrations. Apply these technologies in developing emission model and processor capabilities to allow models to more closely approximate actual emissions in time and space.

Quantify and Report Uncertainty

Develop guidance, measures, and techniques to improve uncertainty quantification, and include measures of uncertainty (including variability) as a standard part of reported emission inventory data.

Increase Inventory Compatibility and Comparability

Define and implement standards for emission inventory structure, data documentation, and data reporting for North American emission inventories.

Improve User Accessibility

Improve user accessibility to emission inventory data, documentation, and emission inventory models through the Internet or other electronic formats.

Improve Timeliness

Create and support a process for preparing and reporting NEI data on a yearly basis.

Assess and Improve Emission Projections

Emission projection methodologies for all emission inventory sectors in North America should be evaluated to determine the accuracy of past projections and identify areas of improvement for future projections.

The priority of these recommendations will depend on the particular situation facing each organization, whether at the federal, regional, state or provincial, or local levels. Nevertheless, each recommendation is applicable to all organizations.

Emission inventories face significant challenges in meeting the ever-increasing demands for timely and accurate information to address AQM needs. Here, many currently available technologies and approaches have been described that can improve emission inventories to enable them to meet those challenges. Applying these tools will require investments of time and money, but these investments will ensure that future AQM decisions are based on the best possible information. This, in turn, will lead to the development of AQM strategies that are as effective as possible in terms of health, environmental, and economic measures.

ACKNOWLEDGMENTS

Support for the NARSTO Management Coordinator is provided by the Office of Science (BER), U.S. Department of Energy, grant no. DE-FG02-06ER64170. Additional funding for G.H. was provided by the U.S. Department of Energy, grant no. DF-FG02-01ER63104. We are indebted to the U.S. Environmental Protection Agency, the U.S. Department of Energy, the U.S. National Oceanic and Atmospheric Administration, Environment Canada, Instituto Nacional de Ecología, the California Air Resources Board, Southern Company Services, Inc., the Electric Power Research Institute, and the American Petroleum Institute for support of this project.

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About the Authors

C. Andrew Miller is a Senior Research Engineer in the Air Pollution Prevention and Control Division at U.S. Environmental Protection Agency (EPA) National Risk Management Research Laboratory. George Hidy and Jeremy Hales are principals of Envair/Aerochem. Charles E. Kolb is president and CEO of Aerodyne Research, Inc. Arthur S. Werner is senior principal engineer and Bernd Haneke is project manager at MACTEC Federal Programs, Inc. David Parrish is a research chemist with the National Oceanic and Atmospheric Administration Earth System Research Laboratory. Christopher Frey is professor in the Department of Civil, Construction, and Environmental Engineering at North Carolina State University. Leonora Rojas-Bracho is general director of research on urban, regional and global pollution at the Instituto Nacional de Ecología (National Institute of the Environment). Marc Deslauriers is chief of the Criteria Air Contaminants Section with Environment Canada Pollution Data Division. Bill Pennell is NARSTO management coordinator with Columbia Research and Education Associates. J. David Mobley is associate director of the Atmospheric Modeling Division at U.S. EPA National Exposure Research Laboratory. Address correspondence to: Andy Miller, U.S. Environmental Protection Agency, Air Pollution Prevention and Control Division, E305-01, Research Triangle Park, NC 27711; phone: +1-919-541-2920; fax: +1-919-541-0554; e-mail: miller.andy@epa.gov.